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Obninsk, Russia

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PROCEEDINGS OF THE TRIPARTITE SEMINAR ON

# Nuclear Material Accounting and Control at Radiochemical Plants

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**Hosting:**

Ministry of Atomic Energy of Russian Federation (Minatom)  
State Scientific Centre of Russian Federation  
Institute of Physics and Power Engineering named after A.I. Leipunski

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**Sponsoring:**

Minatom RF  
Department of Energy (DOE) USA  
Joint Research Centre of European Commission (JRC), Ispra, Italy

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## Foreword

Upgrading of the state nuclear materials control and accounting (NMC&A) system at the nuclear installations of Russia, including radiochemical plants, has assumed the primary importance.

The significant intellectual, financial and materials resources from the USA and European Union were aimed in frame of international cooperation to solve this task.

The numerous results gained in the last years requires its discussion and analysis so that it could be possible to use its results and to coordinate future plans with maximum efficiency.

According to the Nuclear Safety Declaration signed at the Moscow meeting of April 19-20, 1996 by the heads of eight states and in view of the necessity to discuss the results of the work which is under way in Russian radiochemical plants, MINATOM took the initiative to organize a tripartite seminar on NMC&A in the above mentioned plants with participation of experts from Russia, European Union and Department of Energy, USA. This initiative was the logical continuation of former one on organisation of similar seminar for the Uranium Fuel Fabrication Plants in April of 1997.

The problems of creation and operation of NMC&A systems and their components at radiochemical plants were discussed in seminar during November 2 – 6 of 1998. There were 63 Russian and 25 foreign participants in seminar.

The seminar programme includes following sessions and articles:

1. The aspects of State nuclear material control and accountancy (NMC&A)
  - principal NMC&A requirements, NMC&A systems development, NMC&A supervision in radiochemical plants;
  - nuclear fuel cycle in Russia, nuclear material flows and characteristics in the course of nuclear reactor spent fuel reprocessing.
2. NMC&A in radiochemical plants and at separate stages of reprocessing of spent nuclear fuel and irradiated fuel elements of commercial reactors
  - radiochemical plants which reprocess irradiated fuel elements of Russian and US commercial reactors;
  - radiochemical plants which reprocess spent nuclear fuel from Russian, French and UK reactors.
3. NMC&A in storage facilities of radiochemical plants
  - plutonium dioxide storage;
  - spent fuel subassemblies storage;
  - recovered uranium storage.
4. NMC&A computerization, material balance assessment, preparation of reports
  - computerized system development
  - data processing and management;
  - computerized system quality testing and verification.
5. Qualitative and Quantitative measurements in NMC&A at radiochemical plants destructive analysis techniques
  - non-destructive analysis and quantitative measurements based on gamma- and neutron radiation
  - mass/volume measurements of solutions
  - requirements for the accuracy and precision of measurements in terms of NMC&A, measurement quality assurance.

Those who organized this seminar hope very much that the exchange of experience gained by the participants of the meeting will be of great help in tackling the task of developing efficient nuclear material control and accounting systems in radiochemical plants of Russia.

This book contains all reports which were presented by seminar participants.

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## **ACHIEVEMENTS, CURRENT STATUS, AND PROSPECTS FOR RUSSIAN-AMERICAN COOPERATION IN NUCLEAR MATERIAL PHYSICAL PROTECTION, CONTROL AND ACCOUNTING - 1998**

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### **1. Major Activities**

Collaboration between the U.S. and Russia on problems related to nuclear material physical protection, control and accounting (MPC&A) began as a result of the "Agreement between the Russian Federation and the United States of America on Safe and Reliable Transportation, Storage and Destruction of Weapons and the Prevention of Weapons Proliferation," signed by the Presidents of both countries on 17 June 1992.

Subsequently, the "Agreement between the Ministry of Atomic Energy of the Russian Federation and the Department of Defense of the United States of America on Nuclear Material Physical Protection, Control and Accounting" was signed by the Minister and Secretary of both countries on 2 September 1993.

Russian-American collaboration on MPC&A promotes U.S. and Russian national interests in protecting nuclear materials (NM) and preventing illegal trafficking in these materials.

The following major scientific and technical activities are being conducted within the framework of this collaboration:

- The development of an MPC&A regulatory basis that satisfies the current requirements of Russian legislation and international agreements;
- Improvements in methodologies and equipment employed to measure NM accounting characteristics, as well as upgrades of security system equipment, which includes: intrusion detection equipment; access control devices; optical and electronic surveillance systems; special communications systems; detection equipment for NM transit (on one's person or by vehicle); and saboteur and terrorist devices (weapons, explosives, chemicals and other hazardous substances);
- Increasing the extent to which modern computer technologies are used in order to improve automated processing and transfer of NM accounting information;
- The creation of special databases on possible threats and the use of modern computer equipment to improve facility threat and vulnerability analysis methodologies;
- The development and implementation of modern physical protection systems; improving physical security equipment, including: buildings, specialized structures, containers and other physical barriers, as well as specially protected vehicles.
- Improvements in controlling the movement of nuclear materials within the country and across its borders;
- Training and qualifying nuclear facility and nuclear material storage area personnel.

## Session 1

Russian and American specialists jointly introduced the concept known as “the three lines of defense” in order to provide a systematic approach to developing technical requirements and organizing research and experimental design activities aimed at upgrading NM and nuclear facility security equipment.

- The first line of defense is the information support and hardware system designed to prevent the theft of NM from nuclear sites and to prevent the unauthorized entry of explosives, chemical substances and narcotics at nuclear sites.
- The second line of defense is the information support and hardware system designed to detect covert stores of NM, explosives, chemical substances and narcotics in areas adjacent to nuclear facilities (at the boundaries of access and customs control points, in vehicle depots, in vehicles, buildings and underground).
- The third line of defense is the information support and hardware system designed to identify and conduct a criminological analysis of NM, explosives, chemical substances and narcotics that have been detected and confiscated (as a result of illegal trafficking).

Fig. 1 presents the specific goals of Russian-American collaboration.

### **RUSSIAN-AMERICAN COLLABORATION IN NUCLEAR MATERIAL PHYSICAL PROTECTION, CONTROL AND ACCOUNTING**

Agreement between the Russian Federation and the United States of America on the Safe and Reliable Transportation, Storage and Destruction of Weapons and the Prevention of Weapons Proliferation  
17 June 1992

Agreement between the Russian Federation Ministry of Atomic Energy and the United States Department of Defense on Nuclear Material Physical Protection, Control and Accounting  
2 September 1993

### **MAJOR ACTIVITIES:**

- Develop regulatory documents based on Russian and international legislation;
- Provide nuclear material physical protection, control and accounting systems using modern instrumentation and equipment;
- Upgrade information systems;
- Analyze threats to and the vulnerability of sites requiring protection;
- Improve controls over the movement of materials;
- Train and qualify personnel.

### **LINES OF DEFENSE FOR NUCLEAR MATERIALS:**

“First line”: Physical protection, control and accounting systems at nuclear sites

“Second line”: Off-site systems that are used at nuclear facilities to prevent the illegal distribution of nuclear materials and the entry of hazardous substances (explosives, weapons and narcotics) into nuclear sites

“Third line”: Criminological analysis of detected contraband for the improvement of protection systems

Fig. 1

At their April 1996 meeting in Moscow on nuclear safety issues, the G-7 plus Russia adopted the “Program to Combat Illegal Trafficking in Nuclear Materials.” This program establishes the main goals of international efforts to stop illegal NM trafficking and consists of three major components:

## General Aspects of the State System of NMC&A

- Reliable and safe storage of NM, along with effective protection, control and accounting measures to prevent their proliferation;
- Joint intelligence, customs and law enforcement measures to prevent international transfers or sales of stolen NM;
- Joint efforts to oppose criminal elements by identifying and eliminating the illegal supply and demand for nuclear materials.

### **2. The Organization of Activities in the Russian Federation**

The beginning of our collaboration in 1994 on nuclear material protection, control and accounting coincided with a painful period when Russia was restructuring its economy and reorganizing and altering the functions of government bodies, the ministries, scientific organizations and production facilities. Minatom of Russia suffered from reductions in funding and the loss of young and energetic employees during this period, and experienced difficulties with defense industry conversion. The financial support provided by the U.S. Department of Energy for modernizing Russian nuclear material control and accounting (MC&A) allowed Minatom to retain and hire highly qualified specialists in nuclear cities such as Sarov (VNIIEF/Arzamas-16), Snezhinsk (National Research Institute for Physics Applications/VNIITF) and Obninsk (IPPE).

The absence of any Russian Federation (RF) laws regarding the use of atomic energy influenced the organization of Russian-American cooperation in 1994.

Fig. 2 presents the legal changes made from 1994-1998 which delegated the responsibilities for creating and operating the National MC&A System.

The Russian Federation adopted the Federal Law "On the Use of Atomic Energy" (hereinafter the "Law") in November 1995. The general provisions of this law led to significant changes in the approach taken to create the National MC&A System.

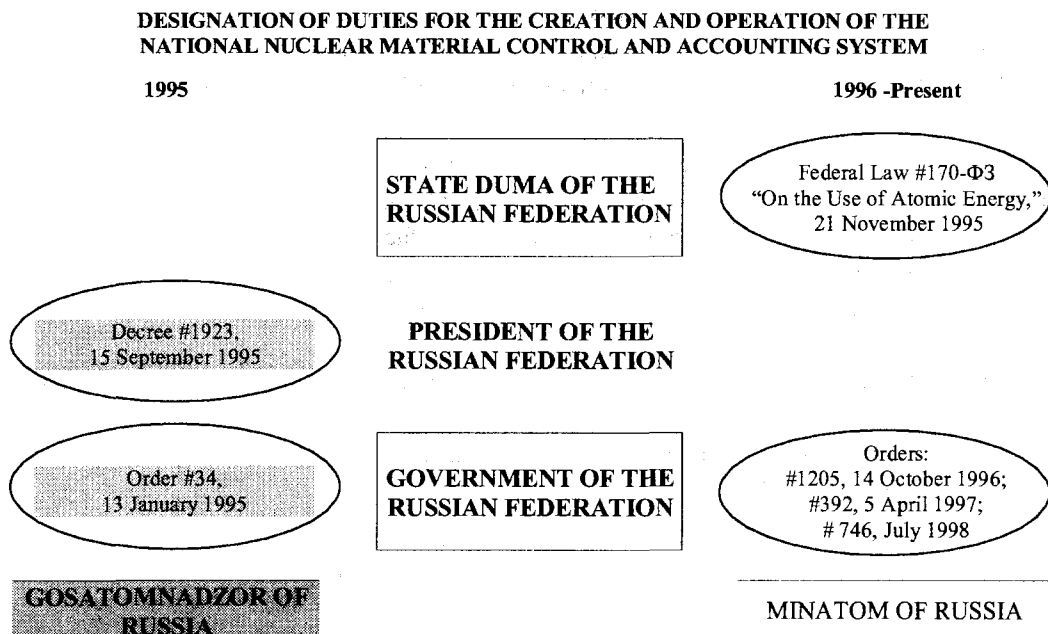


Fig.2

The Law establishes the following general provisions for MPC&A:

- **All nuclear materials are the federal property of the Russian Federation. Entities in possession of nuclear materials shall implement control over their containment and appropriate use** (Article 5 of the "Law").  
From this provision it follows that the MC&A System shall ensure that the appropriate federal executive branch agencies may carry out their designated functions related to the management of nuclear materials under federal jurisdiction.
- **The Government of the Russian Federation shall implement management of nuclear materials under federal jurisdiction** (Article 9 of the "Law").
- **Agencies managing the use of atomic energy are responsible for government nuclear material control and accounting in accordance with established regulations** (Article 20 of the "Law").
- **The National Nuclear Material Control and Accounting system shall ensure government accounting of nuclear materials at the federal and ministry (agency) levels for the following purposes: to establish on-site physical inventory quantities of these materials; to prevent their loss, unauthorized use or theft; and to provide government bodies and government safety regulatory agencies with information on the location and movement of nuclear materials, as well as on their import and export. The Government of the Russian Federation shall develop a procedure for organizing the National Nuclear Material Control and Accounting System** (Article 22 of the Law).
- **Government safety regulatory agencies, within the scope of their authority, shall regulate physical protection for nuclear facilities, radiation sources, storage areas, nuclear materials and radioactive substances. These agencies shall also regulate unified government control and accounting systems for nuclear materials, radioactive substances and radioactive wastes** (Article 25 of the "Law").
- **Operating organizations and specially authorized government agencies shall ensure that nuclear facilities, storage areas and nuclear materials are physically protected. Government safety regulatory agencies shall regulate physical protection** (Article 49 of the "Law").

Minatom of Russia, Gosatomnadzor of Russia and other relevant organizations jointly developed a set of major legal documents in 1996-1997 based on the Federal Law "On the Use of Atomic Energy." These documents, approved by the Government of the Russian Federation, develop the general provisions of the "Law" regarding MPC&A. They include:

- "The Conceptual Design of a National Nuclear Material Control and Accounting System (Order # 1205, 14 October 1996);
- "Physical Protection Rules for Nuclear Materials, Nuclear Facilities and Nuclear Material Storage Areas" (Order # 264, 7 March 1997);
- Organizational Rules for the National Nuclear Material Control and Accounting System (Order #746, June 1998).

These documents contain the following provisions, which are critical for determining the organization and future development of international collaboration in this area:

- Minatom of Russia is established as the government client for activities related to the creation and operation of the National MC&A System. Minatom of Russia is also designated as the management agency for the federal level of the National MC&A System.

## General Aspects of the State System of NMC&A

- Minatom of Russia shall carry out the functions of the central government agency and shall serve as the point of contact in accordance with the provisions of the international "Convention on the Physical Protection of Nuclear Materials." It shall also assume the functions of the national agency authorized by the RF to fulfill its physical protection obligations to the IAEA and other international organizations.
- Minatom of Russia shall create an automated Federal Information System (FIS) for governmental nuclear material control and accounting.
- With respect to MPC&A, Minatom of Russia shall serve as the liaison for federal executive branch agencies, government bodies of the autonomous republics of the Russian Federation, and organizations with hazardous nuclear sites which are under ministry jurisdiction.

Minatom of Russia, in its role as the management agency for the nuclear complex of scientific organizations and production facilities, has had many years experience in research, experimental design and implementation activities, which were conducted in accordance with procedures established by RF government standards (GOST).

These procedures stipulate mandatory compliance with the following requirements:

1. The utilization of statements of work, approved by the clients, for all activities to be conducted, as well as for cost-benefit analyses of these activities;
2. Selection of contractors for all activities on a competitive basis;
3. Compliance with established standard phases and reporting requirements for research, experimental design and implementation activities;
4. Receipt inspection by client committees of individual phases and activities, which shall include the issuance of acceptance certificates for these activities.

These procedures ensure that activities shall be actively monitored for completion and quality.

An Expert Review Commission for Nuclear Material Control and Accounting and a Coordinating Commission for Countermeasures against Illegal Trafficking in Hazardous Substances have been formed at Minatom of Russia and are successfully conducting expert reviews of activities in their respective areas of specialization.

### **Russian-American Collaboration Between 1994-1997**

As a result of contracts with U.S. National Laboratories and International Science and Technology Center (ISTC) projects, 27 science and production organizations of Minatom of Russia have been working with the Americans since the beginning of 1994. This program has increased the level of nuclear material safety and control at Russian nuclear, chemical and fuel production enterprises, nuclear material storage areas, and at the two major nuclear weapons laboratories.

Figs. 3-8 provide a basic outline of Russian-American collaboration between Minatom enterprises and the U.S. Department of Energy.

The specific technical results of our collaboration have been presented in detail at the following meetings:

- At the meeting of the G-7 plus Russia on nuclear safety and radiation protection which was held in Moscow in April 1996;
- At the Russian International Conference on Nuclear Material Control and Physical Protection in Obninsk in March 1997;
- At joint Russian-American workshops (on radiation monitoring, information systems, etc.);
- At the annual INMM conferences.

## Session 1

### **US-RF COLLABORATION ON PHYSICAL PROTECTION, CONTROL AND ACCOUNTING FOR NUCLEAR AND CHEMICAL PRODUCTION**

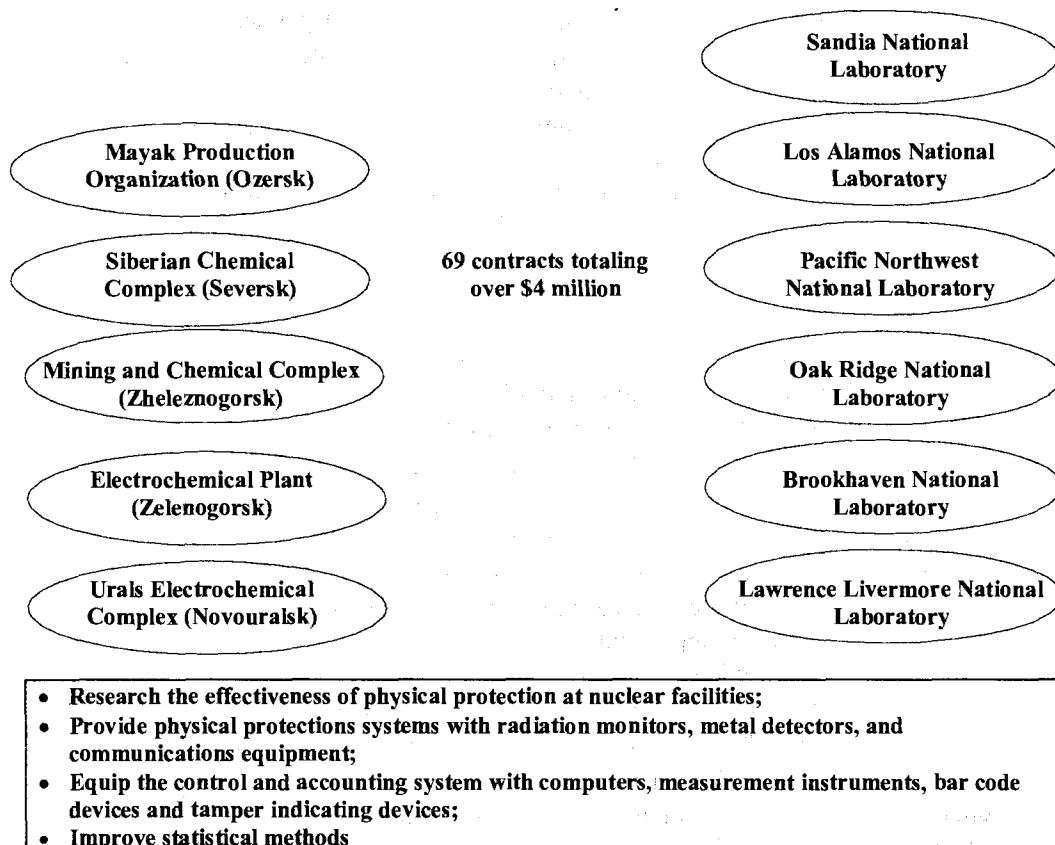


Fig. 3

At the November 1995 international conference at the Livermore National Laboratory on criminological analysis of nuclear contraband, there was a call for scientific and technical collaboration against illegal trafficking in NM—one of the most important goals of the second and third lines of defense. Members of the conference formed an International Working group made up of specialists from several countries to combat illegal trafficking in NM.

This group held three meetings during 1996-1997, at which members worked out approaches to resolving technical problems related to halting illegal trafficking in NM.

During a visit of Russian specialists from Minatom and the Kurchatov Institute to the Argonne National Laboratory (ANL) at the end of 1995, proposals were made for collaboration on technical problems related to the second line of defense. These proposals were further developed at the international workshop "Nuclear Physics Detection Methods for Covert Stores of Explosive Substances and Fissile Materials," which was conducted by the Department of Information, Nuclear Material and Site Security of Minatom, the ISTC and ANL in April 1996 at Obninsk.

There were a number of meetings and discussions between Russian and American specialists from 1995-1997: at the U.S. Department of Energy (November 1995); at the U.S. Department of Defense (November 1995 and November 1997); at Minatom of Russia (October 1997); and

**US-RF COLLABORATION ON PHYSICAL PROTECTION, CONTROL AND ACCOUNTING AT NUCLEAR WEAPONS CENTERS**

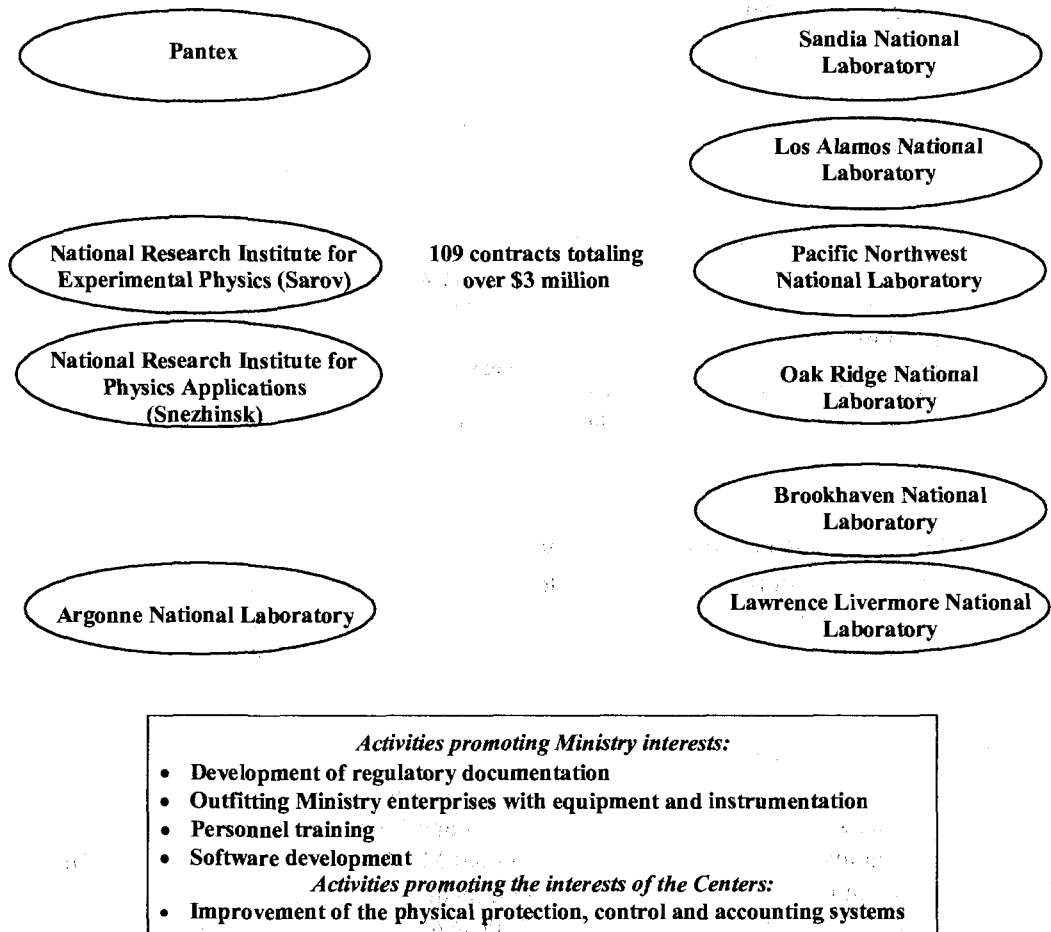


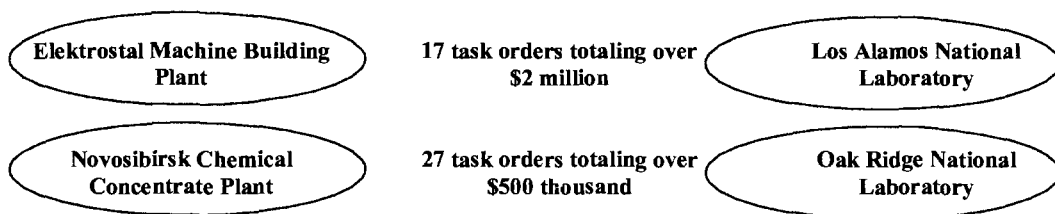
Fig. 4

at international conferences: the Gordon Research Conferences on Practical Applications (London, August 1997); and the Mine Removal Conference (Washington, November 1997). The main topics of discussion at these conferences were related to the improvement and creation of new technologies and, in particular, the use of neutron radiation (the APSTNG proposal from ANL and the Kurchatov Institute, and the *Zaslou* ("Shield") project from the International Science and Technology Center (INTC) at the National Research Institute for Automated Controls).

Russian-American collaboration on improvements to the second line of defense began in 1997.

**An important result of the scientific and technical collaboration between Minatom organizations and the U.S. Department of Energy on MPC&A has the establishment of mutual understanding and interests, trust, as well as professional and personal contacts among individual specialists.**

## US-RF COLLABORATION AT NUCLEAR FUEL FABRICATION FACILITIES



***Improving the Physical Protection, Control and Accounting Systems of High and Low Enriched Nuclear Fuel Fabrication Facilities:***

- Installing pedestrian, vehicle and hand-held radiation monitors
- Installing metal detectors
- Upgrading passageway video monitoring systems
- Installing access control systems
- Installing computer systems for nuclear material control and accounting

Fig. 5

### **Current Status and Future Prospects for Major Activities**

Minatom of Russia's priorities for 1998 are to develop and implement federal and ministry level regulatory documentation and to solve system-related problems. One of the most important tasks is to create an information system for the National MC&A System as quickly as possible.

#### ***The Development and Implementation of Regulatory Documentation***

As noted earlier, 1995-1997 saw the Russian Federation adopt major legislation and Government orders that defined the duties, authority and functions of various government bodies and autonomous republics within the National MC&A System. On the basis of these laws, development began on a set of federal and ministry level documents which contain standards, rules, measurement methodologies, MPC&A equipment requirements, data submission forms, information exchange procedures and decision making processes.

The experience of other nations was taken into account when conducting these activities, especially that of the U.S. In January 1998 Minatom of Russia and the U.S. Department of Energy reached an agreement on the joint development of 40 regulatory documents for the Russian National MC&A System, with the participation of Pacific Northwest National Laboratory (PNNL), IPPE, VNIIEF, the National Research and Design Institute for Power Engineering (RDIPE), and the Atomzaschitainform Center.

#### ***The Federal Information System for Nuclear Material Control and Accounting***

From 1995-1997, and in accordance with the Minatom plan for developing and implementing a National MC&A System, the Central Research Institute for Management, Economics and Information of Minatom (Atominform) developed components and a core version of a computerized information system for the National MC&A System. The Livermore National Laboratory and the Euratom Department of Safeguards provided additional financial and technical support for the project. There was an international demonstration of the core system Federal MC&A Information System in March of 1998.



**US-RF COLLABORATION ON PHYSICAL PROTECTION, CONTROL AND  
ACCOUNTING AT NUCLEAR RESEARCH ORGANIZATIONS**

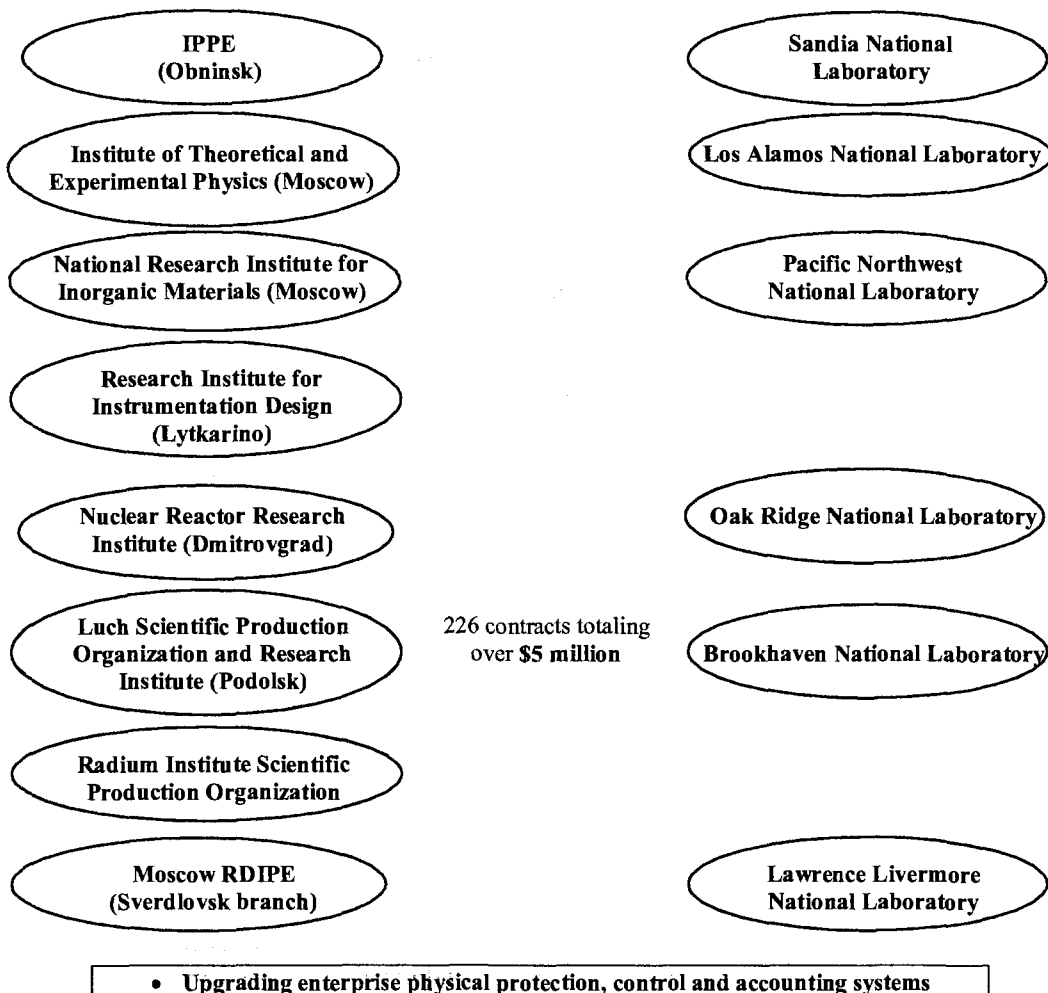


Fig. 6

The Expert Review Commission for Nuclear Material Control and Accounting reviewed the results of these activities on 14 April 1998 and confirmed that they may be used to create the Federal Information System (FIS). The significant assistance provided by the Livermore National Laboratory was noted, as was the personal contribution of Ms. Sandra Taylor, the joint project manager. The Expert Review Commission discussed and adopted the following conceptual design for the computerized FIS, which shall be created at Minatom of Russia in accordance with the order of the Government of the Russian Federation.

The Federal Information System for MC&A is a system of interconnected organizational units and equipment that gathers, processes and uses MC&A information in accordance with federal and ministry level rules. Specific MC&A tasks and functions, the required scope and form of the information to be submitted, and the procedures for exchanging this information shall be established by regulatory documents for all organizational units.

## Session 1

### **US-RF COLLABORATION ON INFORMATION AND TECHNICAL SUPPORT FOR THE NATIONAL NUCLEAR MATERIAL CONTROL AND ACCOUNTING SYSTEM AND PHYSICAL PROTECTION SYSTEMS**

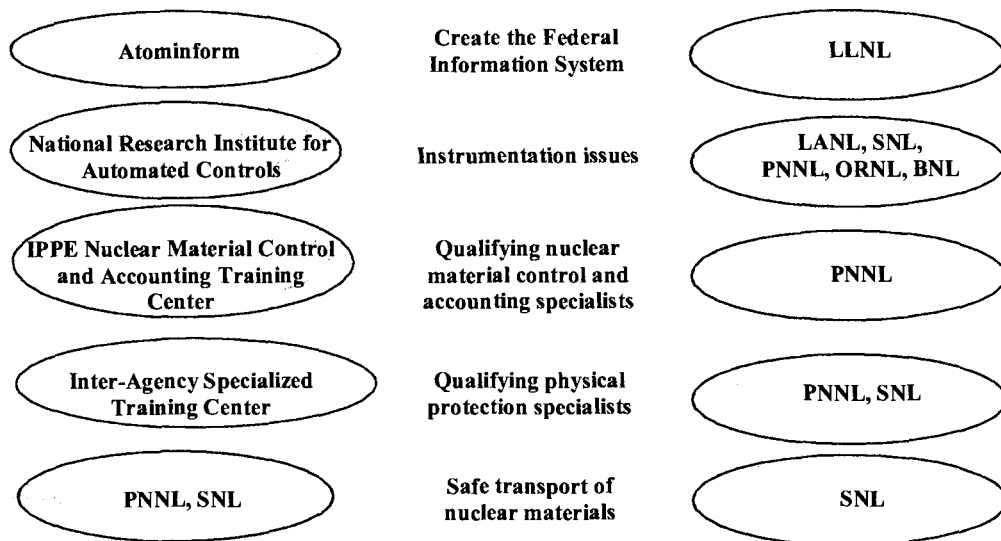


Fig. 7

The FIS will include:

- Information centers to collect and process information at the nuclear facility and NM storage area level;
- Information centers at the operating organization level;
- Information centers at the agency level;
- An information center at the federal level;
- The users of data provided by the various information centers.

The FIS will be equipped with modern computers and information processing software; electronic, magnetic and other information media; and modern communications and information protection equipment.

Standardized forms will be used to collect information and generate the following proposed databases:

- Organizational and technical information on nuclear facilities and NM storage areas with specification of the appropriate operating organization and/or agency;
- Quantitative characteristics of NM (including special non-nuclear materials) and information regarding their distribution at nuclear facilities and storage areas;
- Calibration support for determining quantitative characteristics of materials subject to accounting (existing, developed, future);
- Regulatory documentation on MPC&A and control and accounting system oversight;
- Organizational, technical and other information required to prevent illegal trafficking in NM.

**USA—RF COLLABORATION ON SECOND LINE OF DEFENSE**

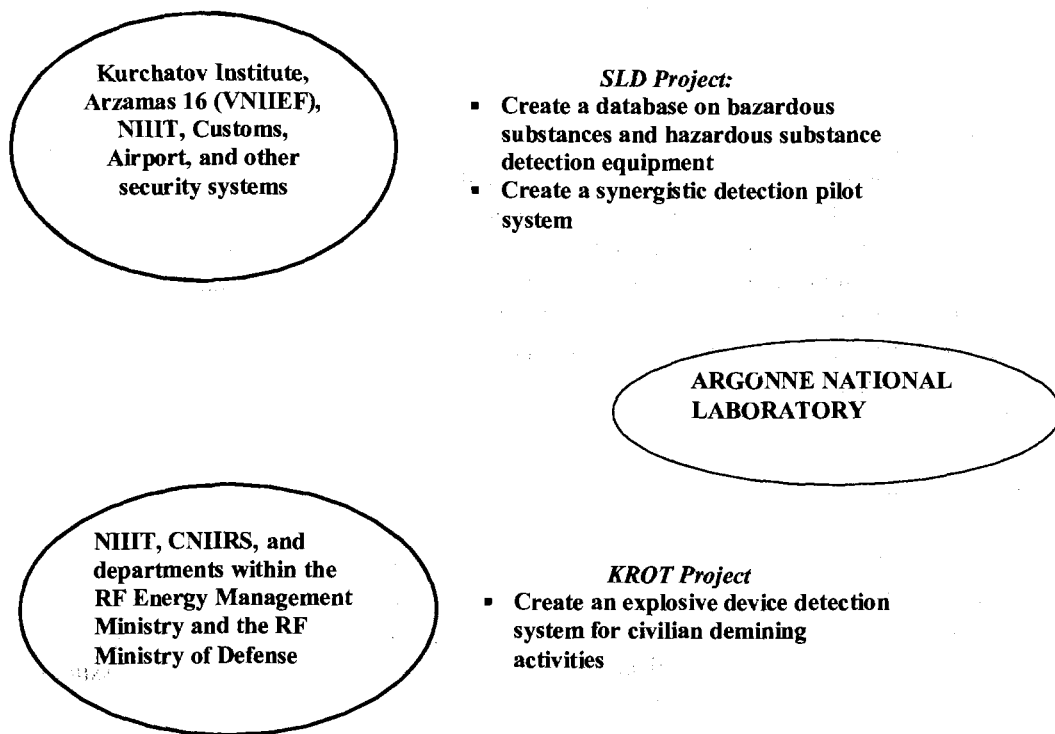


Fig.8

A number of databases will have international status. These databases include:

- Databases providing information on instances of illegal trafficking in NM and other radiation sources in order to assist law enforcement and international organizations, and to inform the public. The creation of these databases is fully in keeping with the efforts of the IAEA to develop an international database (DB) on illegal NM trafficking and with the tasks established for Minatom of Russia, which has been designated the central government agency and point of contact in accordance with the provisions of the International Convention on the Physical Protection of Nuclear Material. The Department of Information, Nuclear Material and Site Security of Minatom has already begun using this DB and, based on that experience, is working to upgrade it. The Department is also making proposals to other interested organizations for collaboration in its further development.
- In compliance with an order of the Ministry of Atomic Energy, the National Research Institute for Inorganic Materials created a laboratory in 1997 to identify NM of unknown origin. This laboratory will conduct analyses for Russia and other countries' law enforcement agencies. The Institute is working with the Institute of Transuranic Elements (Germany) to create a DB for inter-agency and international use which will identify the characteristics of NM used for peaceful purposes. The future and long-term goal of international cooperation is to provide scientific, technical and administrative support for this DB.

- Brookhaven National Laboratory and the Russian National Research Institute for Automated Controls created an electronic catalogue (DB) of MC&A instruments. The Los Alamos National Laboratory has created an encyclopedia of non-destructive assay methods. These activities have established a strong foundation for future collaboration in creating an international DB that will provide scientific and technical support and professional training for MPC&A personnel.
- The Ministry of Atomic Energy authorized IPPE to create a DB of plutonium used for peaceful purposes within the RF. This DB shall be used to implement the provisions of the "Guiding Principles for the Use of Plutonium," which was submitted by the Government of the Russian Federation to the International Atomic Energy Agency.

Organizational improvements have been proposed for the existing information structures in Minatom of Russia, such as vesting these structures with additional authority and duties.

Minatom of Russia is interested in studying the practical experience of the American company NAC International; of particular interest is the manner in which the collection and submission of NM accounting information is organized.

### ***Improving Methodologies and Upgrading Equipment***

Collaboration in the improvement of methodologies and equipment has been directed towards providing instrument and calibration support for MPC&A in the Russian Federation.

Minatom of Russia has focused largely on the following tasks: analyzing available instrumentation; developing a conceptual design and projecting its enhancement; developing and approving standardized specifications for instruments; and providing quality assurance, including calibration support and instrument certification.

Based on specific statements of work, Minatom of Russia is conducting research geared towards developing requirements, improving measurement methodologies, providing quality assurance for instruments, creating calibration standards, conducting calibration research and certification testing for instruments, and developing regulatory documentation to guide the use of these instruments.

The calibration support system encompasses the development, testing, manufacture and operation of instruments, and the standards and equipment used for attestation, verification and calibration. The set of calibration support measures are implemented by the calibration service agencies of Minatom, which consist of the Central Calibration Service, and the calibration departments of lead organizations and enterprises. The ministry has accumulated a great deal of experience in tracking and maintaining at a high level the calibration characteristics of different instruments during their development, manufacture and operation. Built-in self-monitoring and diagnostic devices are widely used, as are surveys and statistical monitoring of technical characteristics. These areas of calibration support are reflected in the general requirements and specifications for calibration instruments.

The current regulatory base includes standards, specifications, guidance documents, rules and regulations that establish progressive requirements for developing, producing and using instruments (including software, equipment and technologies), for quality assurance control and evaluation, for safety assurance, etc.

Legislation of the Russian Federation requires the certification of MPC&A system equipment. The general provisions of the certification system for equipment, devices and technologies for nuclear facilities, radiation sources, and storage areas were developed and subsequently

approved in 1998 by the following parties: the Russian Federation Ministry of Atomic Energy; the Chair of the State Committee of the Russian Federation on Standardization, Calibration and Certification; and the Head of the Federal Regulatory Agency of Russia for Nuclear Safety and Radiation Protection. Equipment, devices and technologies (including those that are imported) are certified for their compliance with established regulatory requirements.

Certification centers for Minatom of Russia include:

- The Sigma Specialized Research Institute for Instrumentation Design (Moscow), for radiation measurement instruments;
- The Eleron Specialized Research Organization, for intrusion detection equipment in physical protection systems;
- The National Research Institute for Inorganic Materials, for standard samples and sampling equipment used to measure NM characteristics.

The foregoing should be taken into account in the assessment of Russian-American collaboration on MPC&A instrumentation.

The current status of our collaboration in this area may be characterized by the following accomplishments:

Firstly, a significant number of American instruments (radiation monitors, neutron coincidence counters, radiation spectrometers, bar-coding equipment, computers, etc.) were delivered between 1995-1998. These instruments and equipment are being used successfully in MPC&A systems at Minatom production facilities and institutes. A good example of this is the 1997 U.S. shipment of 26 TSA pedestrian radiation monitors to the Siberian Chemical Complex. These monitors have been installed at the enterprise's entry control points.

Secondly, new instruments are being developed as a result of our collaborative efforts, and instrument methodologies are being updated at Russian research organizations. One example is the work of VNIIEF and Los Alamos National Laboratory on the Russian development of the new KPRM-01, and the decision to have VNIIEF mass produce this device for the physical protection systems at weapons facilities.

The improvements being made in methodologies and equipment as a result of Russian-American collaboration are leading to a new level in quality.

Activities are already underway for installing modern equipment in the first, second and third line defense systems which will be used to detect and identify NM, radioactive substances, explosives and other hazardous substances.

Joint research and development has also begun on new synergistic systems for comprehensive solutions to detection and identification problems. Proposals for collaborative efforts in this area are as follows:

- Create computer DBs on hazardous substance detection and identification, as well as expert recommendations for authorized government bodies and organizations under their jurisdiction regarding technologies to detect, identify and neutralize these substances;
- Develop and introduce new direct monitoring and measurement methodologies to be used to combat illegal trafficking in hazardous substances;
- Develop and implement comprehensive information and measurement systems to detect and identify hazardous substances. These systems shall be used for customs control, when monitoring the flow of freight and passengers, and when evacuating an area;
- Create a unified information space for regulatory documents of the RF, the U.S. and other countries, as well as for data on the results of joint activities.
- Train personnel for these activities.

## Session 1

At the initiative of Minatom of Russia, and with the support of the U.S. Department of Energy, the Moscow Engineering Physics Institute has organized student training in a new specialty, "Nuclear Physics Detection Methods for Hazardous Materials." The National Qualification Institute and the Minatom Nuclear Material Control and Accounting Training Center are working with the U.S. to qualify MPC&A specialists.

**Minatom of Russia places a high premium on the achievements, current status, and prospects for Russia and American collaboration in these endeavors.**

**The process of upgrading the MPC&A systems is an ongoing and long-term process that consists of modernizing measuring equipment and methodologies, improving data exchange and processing technologies, and improving administrative procedures.**

**The positive results that we have already achieved form a foundation upon which this collaboration may extend into other new and important areas, such as the "second and third lines of defense," which are directed toward countering illegal trafficking not only in nuclear materials, but in other hazardous substances that constitute a threat to the nuclear sites and national security of our country and of other countries.**



## **MAIN REQUIREMENTS AND CRITERIA FOR STATE NUCLEAR MATERIAL CONTROL AND ACCOUNTING**

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The paper presents comments and substantiation of the main requirements and criteria for the state NMC&A system in the draft of the federal "Main regulations of nuclear material control and accounting".

Purpose of the regulation development is a creation of a common system of organizational, methodological procedural and technical requirements to NMC&A at nuclear facilities, enterprises, utilities and governmental authorities, which the state-of-the art the current political and economical situation. The main purpose of the existing system modernization is putting into service the technical means aiming at the maximum possible reduction of the human factor influence on the system reliability.

Principal points of the Russian NMC&A system modernization are as follows:

- confirmation of actually available amounts of the nuclear materials under inventories and transfers based on relevant measurements,
- conclusions about the registered and actually available amounts of the nuclear materials on statistical base,
- rapid accounting achieved by computerization,
- rapid control achieved by measurements automation,
- use of state-of-the art tampering indicator devices (TID).

### **State NMC&A system structure and design principles**

Three level structure is laid down for the state NMC&A system by the Regulations with the purpose of ensuring the adequate reliability of the system:

- in material balance areas (MBA),
- at enterprises, utilities,
- at the authorities managing the nuclear power use (agency and federal levels).

In accordance with the federal law "On the use of nuclear power" a supervision of the state NMC&A system under the nuclear power use by state regulatory authorities – Ministry of Defense and Gosatomnadzor of Russia, is stipulated.

**List of nuclear and special non-nuclear materials which are subject to the control and accounting**

The following list of the materials is stipulated in the Regulations.

Nuclear materials:

- plutonium,
- plutonium with Pu-238 >20%,
- uranium,
- uranium-233,
- uranium-235,
- thorium,
- neptunium-237,
- americium-241,
- americium-243,
- californium-252.

Special non-nuclear materials:

- lithium-6,
- tritium,
- deuterium excepting deuterium contained in heavy water,
- heavy water.

The list of the nuclear and non-nuclear materials has been specified to develop the Conception of the state NMC&A system.

In addition the following materials have been included in it.

Nuclear materials:

- plutonium with Pu-238 >20%,
  - uranium-233,
  - uranium-235,
- and heavy water as a special non-nuclear material.

Plutonium with Pu-238 >20% is included into the list in order to simplify NMC&A procedures at nuclear facilities producing sources from that plutonium. As a rule these facilities don't possess plutonium suitable for production of nuclear explosive devices.

Uranium-233 and uranium-235 nuclides are included in order to ensure the differential approach to provision of safety measures for uranium of different percent enrichment in U-235 and an equivalency of containment measures as to plutonium and uranium-233 having comparable properties weapon-grade nuclear materials. The similar approach is used in other national systems, in particular in the US Department of Energy.

The heavy water is included in order to maintain the existing procedures of its control and accounting as a special material as well as with regard to its presence in the list of the materials which aren't subject to a free interstate distribution.

**NMC&A principles**

The basic principles are stated in the Conception of the state NMC&A system, the list of these principles supplemented and specified in the Regulations. They are as follows:

- Regulations are not used for NMC&A if the nuclear materials are part of the nuclear weapons, nuclear warheads and their components,
- Nuclear materials are subject to the state control and accounting beginning with the minimal amounts,



### General Aspects of the State System of NMC&A

- Nuclear materials are classified by categories depending on the amount, type and form of NM with the purpose of ensuring the differential approach to determination of the control and accounting procedures and methods,
- Material Balance Areas (MBA) are set up at the enterprises, they are the main structural element of the state NMC&A system,
- Key Measurement Points of (KMP) of the nuclear materials are fixed in every MBA,
- Tampering Indicator Devices (TID) are applied to the nuclear materials, they prolong the measurement confidence of the quantitative characteristics and attributes of the nuclear materials which have been done earlier,
- Nuclear material accounting is based on the results of NM quantitative characteristics measurements,
- It is admissible to use the results of the previous measurements of NM quantitative characteristics if their confidence is confirmed by the proper application of the TID and appropriate confirmatory measurements,
- Physical inventories are carried out to determine actual available amounts of nuclear materials in MBAs,
- Conclusions about a shortage/excess of the nuclear materials, an absence of anomalies/drawbacks in nuclear material use in the system of their control and accounting are made by means of comparing of the obtained value of the Inventory Difference (ID) with its admissible value for each MBA,
- If any anomaly in the control and accounting of nuclear material is not determined as a result of NM balance analysis, then the book inventory of the material balance in the MBA is used as a physical inventory of NM in the MBA at the beginning of the next material balance period,
- Common system of the report forms is established for all levels of the state NMC&A,
- System of NM measurements and Measurement Quality Assurance Program are established.

As the system of the state control and accounting of radioactive waste, materials and packaged sources of ionizing radiation (SIR) exists currently and continues to develop, the minimum quantities beginning with which the nuclear materials are subject to control and accounting within the state NMC&A system are fixed in the Regulations.

The minimum quantity values have been fixed based upon the small amounts of NM's in the following widely spread devices and instruments: packaged SIR, fire alarm detectors, neutron detectors, other instruments and devices.

The state accounting of NM's there is of no strategic importance because they are accounted in the state control and accounting system of radioactive waste, materials and packaged sources of ionizing radiation.

In the determination of minimum quantities of NM the threshold values of NM amounts were also taken into consideration. They are stipulated in the regulations for safe-secure transportation of radioactive and nuclear materials (see: Main regulations for Safety and Physical Protection under Nuclear Material Transportation – OPBZ-83, Regulations for Safe Transport of Radioactive Materials – Safety Series No. 6 of IAEA, Regulations for Safety under Radioactive Material Transportation – PBTRV-73).

In these Regulations the value of 15 g for uranium-233,-235, plutonium and other isotopes of the transuranium elements transported in a container is fixed as a boundary separating the nuclear materials from the radioactive ones. The threshold approach has been accepted in other

**TABLE 1. Minimum amounts of nuclear materials beginning with which they are subject to the state control and accounting**

#	NUCLEAR MATERIAL	MINIMUM QUANTITY	Last significant digit of NM mass in reporting documents
1.	Plutonium	15 g	1 g
2.	Plutonium with Pu-238 > 20%	15 g	1 g
3.	Uranium-233	15 g	1 g
4.	Uranium >10% enriched in <i>U-235</i>	15 g in <i>U-235</i> isotope	1 g
5.	Uranium no less than 10% but more than naturally enriched in <i>U-235</i>	15 g in <i>U-235</i> isotope	0.1 kg
6.	Neptunium-237	15 g	1 g
7.	Totality of NM listed in items 1-6 of the Table	15 g from sum of masses of <b>Pu</b> , <b>U-233</b> , <b>Np-237</b> and <b>U-235</b>	1 g
8.	Americium-241	1,0 g	0.1 g
9.	Americium-243	1,0 g	0.1 g
10.	Californium-252	0,1 g	0.001 g
11.	Uranium no more than naturally enriched in <i>U-235</i>	500 kg	1 kg
12.	Thorium	500 kg	1 kg
13.	Lithium-6	1,0 kg	0.1 kg
14.	Tritium	0,2 g	0.01 g
15.	Deuterium excepting deuterium in heavy water	2 g	0.1 g
16.	Heavy water	200 kg	1 kg

national systems, in particular in the national NMC&A system in France. Decree No. 81-512 of May 12, 1981 determines the main requirements to NMC&A and fixes the following threshold values: plutonium and uranium-233 – 3 grams, uranium >20% enriched in *U-235* – 250 grams in *U-235*, natural and depleted uranium, thorium – 500 kg, deuterium – 200 kg.

The approach to the minimal amount determination adopted in the Regulations enables an essential number of facilities to be excepted from the terms of reference of the Regulations (those facilities working with rather small amounts of NM with the purpose of producing or using detectors, instruments and devices of various types) and to concentrate efforts on NMC&A improvement at the facilities possessing significant amounts of NM.

On the basis of the existing practice of the state control and accounting of the nuclear materials as well as radioactive waste, materials and sources packaged SIR, uranium and thorium contained in the ore and in interim products processed at mining and smelting enterprises are not included in the terms of reference of the Regulations.

Americium-241, lithium-6 and deuterium are excepted from the terms of reference of the Regulations with the purpose to maintain the existing practice of the control and accounting of these materials, if:

- americium-241 is in plutonium-containing products,
- content of lithium-6 doesn't exceed 8 at. % in lithium,
- relative isotopic content of deuterium in hydrogen-containing materials doesn't exceed 50 at. %.

These threshold values have been fixed based upon the existing capabilities of americium-241, lithium-6 and deuterium extraction from the above mentioned compounds.

The scope of control and accounting measures application is determined to depend on NM categories. They in turn are determined depending on the properties and masses of NM in MBAs.

This principle has already been exercised in Russia but the MBA boundaries were fixed proceeding from the administrative structures with no regard to NM characteristics and masses.

NM categories and category criteria have been established based upon the technological process parameters existing at the Russian facilities processing NM compounds so that to connect NM category determination with these processes as much as possible.

The requirements to MBA structure of the nuclear facilities are determined based upon the foreign practice and existing administrative structures at the Russian enterprises so that to maintain the boundaries of the material accountability for NM and confine the anomalies in NM use within technically founded limits.

#### **Measurement system for the values for NMC&A and Measurement Quality Assurance Program**

The accepted system and program are practically the same as for the process parameters measurements. A possibility to use the samples for certification of procedures and measures developed and certified by an enterprise is provided (if it is stipulated in the state standards or those for the branch of industry).

#### **NM transfer procedures**

In addition to the existing requirements to transfer procedures there is a necessity to measure the characteristics of NM both under the NM delivery and receiving of the NM.

The criteria for determining the significant discrepancies in Shipper/Receiver data have been established. The period during which the Receiver has to perform the necessary checks and measurements and to place the NM under accounting has been established. The confidence coefficient 0,99 has been chosen as a numerical criteria, a 10-day period has been taken for the receipt procedure fulfillment. These requirements are implemented on the majority of enterprises under NM shipment/receipt.

#### **Physical Inventory Taking, Closing a Material Balance and Evaluation of Inventory Difference**

The nuclear material physical inventory are defined as the *main measure* to confirm the inventory of nuclear materials and to detect of the nuclear material NM shortage/excess, anomaly/drawback in the use of nuclear materials, their control and accounting system.

The procedures of inventory taking have been essentially modified and complicated especially for MBA with the nuclear materials of the first and the second categories. The material balances period for them has been assigned 1 and 3 months respectively. At a number of facilities, research ones in particular, inventories of these nuclear material are taken now once a year. The nuclear safety practice shows that such a long term does not allow the adequate efficiency of detecting of the accounting drawbacks, losses, violations of nuclear material handling to be ensured. In particular the results of the nuclear accidents investigations at the Novosibirsk Plant of Chemical Concentrates and at RFRC Research Institute of Experimental Physics which took place in 1997 confirmed these facts.

In the process of the physical inventory performance in MBAs the following procedures have to be carried out for every nuclear material:

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- accounting data checks,
- attribute checks,
- quantitative measurements,
- closing of a balance,
- determination of the Inventory Differences (ID) and their errors.

The inventory frequency is:

- 1 calendar month for MBAs with materials of the 1<sup>st</sup> category,
- 3 calendar months for MBAs with materials of the 2<sup>nd</sup> category,
- 6 calendar months for MBAs with materials of the 3<sup>d</sup> category,
- 12 calendar months for MBAs with materials of the 4<sup>th</sup> category,
- 12 calendar months for all the nuclear materials of the enterprise.

For nuclear materials in the *bulk form* the inventory difference is chosen as a quantitative measure according to which the values of the nuclear material shortage/excess, anomaly/drawback are defined. The numerical criteria have been established for them in the Regulations and the statistic of procedures and assessment parameters have been defined.

In contrast to the existing order of inventory taking the Regulations allow the use of the results of previous measurements of NM quantitative characteristics only in the cases when the confidence of the results has been confirmed by the proper application of the tampering indicator devices (TID) and relevant *confirmatory* measurements. This requirement provides the assurance of the previous measurement data (extends their confidence) used in closing the balance. In order to fulfil that requirement the additional expenses are needed for inventory taking and TID's application as compared to the existing ones.

### **Balance closure of Bulk Form NM**

When closing a balance of bulk form NM the values of every component of the balance equation for each nuclear material for every MBA have been determined (not for the facility as a whole) and the values of these component errors (the masses of NM obtained, shipped, available, lost forever).

These data have to be used in order to prove that the inventory difference every nuclear material type for the MBA is really accounted for by the measurement errors of the nuclear material characteristics and rather than results from the nuclear material shortage/excess and anomaly/drawback during their use or in the system of their control and accounting.

The conclusions about the nuclear material shortage/excess and the absence of anomaly/drawback in the use of nuclear materials within the control and accounting system are made on the base of the results of the nuclear material inventory taking including the statistical rules of decision making by comparing of the inventory difference value obtained with its admissible value for every MBA.

If the nuclear material control and accounting anomaly has not been stated as the result of nuclear material balance then the nuclear material amount documentary recorded in the MBA is used as a nuclear material inventory in the MBA at the beginning of the next material-balance period.

The condition of the inventory difference (ID) module excess of the threshold values of three types set by the Regulations is adopted as a criterion for detecting the above mentioned cases. Each of three threshold values is important for MBAs, for different types of nuclear facilities.

ID error value increased threefold is chosen as a threshold value of the first type. If it is exceeded it can be confirmed that ID really differs from zero and it is necessary to determine the

cause of that difference. First of all that value is important for a MBA in which the nuclear material accounting measurements are carried out in the material-balance period when receiving shipping, or identifying the inventory.

The existing rate of irretrievable losses which is different for the different types of facilities is adopted as a threshold of the second type. If ID exceeds that rate then the drawbacks in the technology and in the control and accounting may be the possible reasons of it. This threshold is important for MBAs when great amounts of the nuclear material are handled in the bulk form. The MBAs where the measurements are made with large errors have the greatest difficulties. In these cases ID statistic fluctuations can often exceed that threshold and it is necessary to improve the methods and measures or to take inventories more often, possibly depending on the nuclear material flow.

The nuclear material mass, which is different for different categories is the third type of the threshold value. This threshold also follows the administrative purpose: the managing bodies and persons have to be sure that there are no losses, excess of significant amount of the nuclear material in a MBA.

Owing to the above reasons the following threshold values are set for the inventory difference (ID) module (with 0.99 confidence probability):

1. ID error confidence limit (criterion of that type is used in all national and international nuclear material control and accounting systems). For example, according to the standards of the US Nuclear Regulatory Commission the warning limit is on reaching the confidence interval corresponding to the confidence level of 0.95, and the emergency limit corresponds to 0.99 level).
2. 0.2% of the sum of the recorded amount of the given nuclear material and its amount increasing during the material-balance period – for commercial nuclear facilities (the criterion of that type is used in the foreign nuclear material control and accounting systems, but with different values of the criterion, for example, for the US NRC it is 1%, and for Japan it is 0.25%).  
3% of the same value is for experimental commercial and research nuclear facilities.
3. 3 kg of plutonium, uranium-233 for MBA containing the nuclear material of the first and the second categories; 8 kg of uranium-235 for MBA containing the nuclear material of the 1, 2 and 3 categories; 70 kg of uranium-235 for uranium of less than 20% enrichment.

The first value (criterion number 1) defines the limit of the inventory difference of statistical significance (3 errors of its definition). The similar limit is two or three errors of the ID definition depending on the system. It is used in all national accounting systems. In the US Department of Energy it is the main criterion due to the fact that the ID definition error obtained at the enterprises is small owing to the high quality of the accounting measurement reached during the half century of its existence.

The second and third values (criterion number 2) limit the ID by the rates of irretrievable losses (these rate values and even smaller ones are valid today for the most part of the facilities). Besides, it is an important criterion for the facilities operating with small amount of the nuclear materials close to the threshold amounts, for example, 3 kg of plutonium.

The last three values (criterion number 3) have been stated originating from safeguards IAEA requirements (the similar values were stated in IAEA recommendations, see, for example, the IAEA/SG/INF/4, Vienna, 1983). If the limitations on the nuclear material amount are not introduced in absolute values then the enterprises dealing with large amounts of the nuclear materials meet all the requirements and the uncertainty in the nuclear material accounting can be tens of kilograms of the highest category nuclear materials.

The given threshold values require that the MBA structure at nuclear facilities be determined in such a way that it is possible to confine the losses/excess of the nuclear material of significant masses.

### Accounting Units in the Inventory

The Regulations stated the necessity of carrying out *only optional confirmatory measurements* for the accounting units in which the true information about the nuclear material had been confirmed by tampering indicator device (TID) or by the special structure of the item (for example, the one-piece products in the given nuclear facility). These measurements confirm the passport data of the item characteristics at the moment of inventory taking.

The statistical significance of the difference between the results of the accounting and confirmatory measurements of the NM quantitative parameters, items, products is determined from 0.99 confidence level. If the difference is not significant, then the "passport" value is accepted (obtained in the accounting measurements).

The sample size is determined taking into account two parameters: the nuclear material threshold amounts for detection of their shortage/excess and the probability values for the detection of shortage/excess of these threshold amounts:

The threshold amounts for the nuclear materials of the 1,2 and 3 categories are:

- 3 kg – for plutonium, uranium-233,
- 8 kg – for uranium-235.

For uranium of less than 20% enrichment (4 category) the threshold amount is

- 70 kg of uranium-235.

**TABLE 2. Probabilities of Detecting Nuclear Material Threshold Amounts Shortage/Excess for the Confirmatory Measurement Sample Size Calculation**

TID application area	Detection probability, %	
	1,2,3 categories	4 category
Only TIDs (seals, stamps, etc.) were applied to the nuclear material	50	30
Only monitoring devices (tele-, radio-monitoring, etc.) were applied to the nuclear material	50	30
Two types of TID simultaneously applied to the nuclear material	25	10
More than two types of TID simultaneously applied to the nuclear material	10	5

The stated detection probabilities encourage the TID application and are based on the assumption that any of TID type possesses enough and approximately similar reliability and allows (several times or even tens and hundreds times – for the items with the nuclear materials of small masses) the scope of confirmatory measurements to be significantly reduced. For nuclear materials of the 4<sup>th</sup> category, the probabilities are reduced taking into account the principle of difference of the control and accounting measures.

However, even if there are several types of TID it is necessary to check the attributes at least of a small part of the items. This check is required for confirmation of the availability of the items with the declared masses and parameters of nuclear materials. The similar approach is used in the nuclear material control and accounting system of the US DOE and during the inspections of Euroatom and IAEA.

### **The System of Accounting Report Documentation and Preliminary Notifications**

The Regulations do not require essential modification in the content and procedure of the reports presentation at the enterprise level.

The main modification and complications will be required for the documents prepared in MBAs (by the nuclear facility departments). These documents have to be presented to the nuclear material accounting service at the enterprise. The most essential modification is required for the procedures of **Material Balance Reports (MBR)** preparation from MBA. The values of the inventory differences and standard errors of their definition according to each nuclear material have also to be included into these reports.

The modification and additions will also be required for the accounting and operation and technical documentation in MBAs, where the information about control and measurement results for the nuclear material batches is to be recorded.

The order of notifications about nuclear material transfers has not been modified in practice. Major attention is paid to the export-import operations and the nuclear material transfer between the enterprises.

### **The Nuclear Material Control and Accounting Arrangement**

In practice all the aspects of the upper level are defined by the "Regulations of the State Nuclear Material Control and Accounting System Arrangement". The given Regulations define the structure and requirements of the control and accounting system arrangement at the enterprises, which have to be recorded in the corresponding provisions of the enterprises and MBA instructions and of the enterprise departments.

### **The Federal and Departmental Control in the State Nuclear Material Control and Accounting System**

The scope and types of the control at the federal and departmental levels have been defined in the Regulations. The federal control carried out by Minatom of Russia is directed to the systems directly providing the state bodies with the necessary data, first of all the federal information center. They are the systems of the enterprises, utilities and departments. The departmental control carried out by the Ministries and utilities is directed to MBA systems, to the procedures and data, which are carried out and generated in them.

### **The State Nuclear Material Control and Accounting System Supervision**

In accordance with the "Regulations of the State Nuclear Material Control and Accounting System Arrangement" in force the bodies of the state safety regulatory authorities in the frameworks of their authority provide the inspections under the use of atomic energy. These inspections goal is checking the state of the nuclear material control and accounting and the nuclear material availability within the territory of the Russian Federation. The Regulations stipulate that the procedure of inspection activity depends on the category of NMs kept at the enterprise, in the MBA, the intensity of nuclear activity there, TID reliability and is defined by the state safety regulatory authority in accordance with the administrative body for the use of atomic energy. It has been dictated by the necessity of the safety assurance and the technological modes observation during the inspections.

### **Requirements to the Personnel Carrying out the Nuclear Material Control and Accounting**

The requirements stated are similar to the requirements to the personnel carrying out the responsible and dangerous work (training and testing the knowledge).



## **NUCLEAR FUEL CYCLE IN RUSSIA FLOWS AND PARAMETERS OF NUCLEAR MATERIALS REPROCESSED AND PRODUCED AT RADIOCHEMICAL PLANTS**

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The structure of nuclear fuel cycle in Russia and nuclear material flows between radiochemical plants and reactors, as well as nuclear facilities using plutonium and regenerated uranium as input materials are reported. The properties and parameters of nuclear materials received by radiochemical nuclear facilities and shipped therefrom are specially considered.

### **Introduction**

For the state system of nuclear material accounting and control (NMC&A), the most important information items include the reception, shipment, and transfers between nuclear facilities. Reported are major data needed for the analysis of flows of nuclear materials produced and processed at radiochemical plants. One of the objectives of upgrading the system for state accounting and control of nuclear materials both at federal level and in operating organizations is the improvement of prompt information transfer and reliability of the data on nuclear material flows between facilities and plants.

It is an important factor facilitating the upgrading of the state NMC&A system that all nuclear fuel cycle facilities of Russia are operated by one governmental body – RF Minatom. Therefore, all information on nuclear materials is available at its departments, whatever the data form; the nuclear material flows are regulated by its departments, and safety and technology-related issues are also their domain.

If the RF GAN authorities for the supervision of MC&A systems and nuclear and radiation safety are taken into account, it becomes clear that safety assurance for all nuclear facilities subordinated to the Russian Minatom, including the radiochemical plants of "Mayak", Siberian Chemical Combine, SChK (radiochemical plant) and Mining Chemical Combine, GChC (RT-2 and radiochemical plant) appear to be under strict supervision of two governmental bodies.

Because of the USSR disintegration, the percentage of export-import operations with nuclear materials has drastically increased. Many NPPs appeared to be operated beyond the USSR borders, their spent fuel being stored and reprocessed at Russian radiochemical plants. This number includes the Ukrainian NPPs with VVER-1000 reactors, Mangyshlak facility with BN-350.

As a result, work scope has increased for NM accounting and control in the import- and export operations with foreign countries, their nuclear facilities being under IAEA supervision.



**TABLE 1. Major parameters of spent nuclear fuel reprocessed at radiochemical plants**

Type of reactor, shippers	Type of fuel loaded, nuclear materials	Parameters of nuclear materials in spent fuel	Receiver
<b>VVER-1000</b> Kalinin NPP, Novovoronezh NPP, Ukrainian NPPs	LEU dioxide, U enrichment from 3% to 4.4%	Plutonium (up to 25% Pu-240) Uranium, enrichment 1.4 – 2.0 %	<b>RT-2 Plant, GChK</b> (Spent fuel assemblies are stored in special canisters and are not reprocessed )
<b>VVER-440</b> Novovoronezh NPP, Kola NPP	Dioxide of low enrichment U Uranium enrichment from 1.6% to 4.4%	Plutonium (up to 25% Pu-240) Uranium, enrichment of 1.4 – 2.0 %	<b>RT-1 plant, “Mayak”</b>
<b>BN-600</b> (Beloyarsk NPP), <b>BN-350</b> (Kazakhstan)	HEU dioxide Uranium enrichment from 17% to 33%	Plutonium (from 0.8 to 10% Pu-240) Irradiated HEU	<b>RT-1 plant, “Mayak”</b>
<b>BOR-60</b> SSC RF-NIIAR	HEU dioxide Uranium enrichment 90%	Plutonium (from 0.8 to 10% Pu-240) Irradiated HEU	<b>RT-1 plant, “Mayak”</b>
<b>MIR, SM-2, IRT, IVV, MR, VVR</b> NIIAR, TPI, BNPP, KI	Alloys of dioxide, metallic HEU with Al Uranium enrichment from 10% to 90%	Plutonium (from 0.8 to 10% Pu-240) Irradiated HEU	<b>RT-1 plant, “Mayak”</b>
<b>Reactors for vehicles, Coast</b> spent Fuel storage facilities of Navy and Murmansk Shipping Company	Alloy of metallic HEU	Plutonium (from 0.8 to 10% Pu-240) Irradiated HEU	<b>RT-1 plant, “Mayak”</b>
Commercial reactors (IUE) of GChK and SChK	Metallic natural uranium	Weapons-grade plutonium, irradiated natural uranium	Radiochemical plants of GChK and SChK

#### **Nuclear material flows in nuclear fuel cycle recycled and produced at radiochemical plants**

The input information for the analysis of flows in fuel cycle of nuclear materials reprocessed and produced at radiochemical plants include the data:

- On the burn-up of spent fuel subassemblies and standard irradiated uranium elements (IUE) received from national and foreign reactors;
- Composition of end products obtained at radiochemical plants;
- Containers used for transportation of both input products (spent subassemblies – SA; IUE), and end products (uranium and plutonium oxides, uranium nitrates)

Also, data are necessary on the properties and quantities of radioactive wastes (RW) produced at radiochemical plants, on the content of nuclear materials in the RW.

Table 1 lists research, power, and commercial reactors, spent fuel subassemblies, the IUE being reprocessed at Minatom radiochemical plants; major properties of reprocessed materials [1] important for nuclear material accounting and control.

The Russian nuclear fuel cycle is partially closed in its uranium branch [2], which is an important feature:

- First, the uranium extracted in the reprocessing of spent SA at radiochemical plant RT-1 of "Mayak", is prepared and transported as uranyl nitrate at PA "UMZ" (Ulba Machine-building plant); protoxide of HEU is sent to the Machine-Building plant at Electrostal
- Second, regenerated uranium extracted in the reprocessing of IUE from commercial reactors at radiochemical plants of GChK and SChK is prepared and transported to the sublimation plants, being added there to natural uranium in the production of uranium hexafluoride – source material for uranium enrichment plants.

TABLE 2. Parameters of NM flows at spent fuel reprocessing (cont.)

Receipt		Shipments		Waste
Product, NM	Parameters of containers	Product, NM	Parameters of containers	
RT-1 "Mayak"				
SPENT ASSEMBLIES OF 1) VVER-440	Transportation package containers TK-6 SA per TK: < 30 items	Civil Pu dioxide PuO <sub>2</sub>	Tight containers: H=260 mm, D=148 mm, PuO <sub>2</sub> mass up to 3,5 kg	Are reprocessed and stored on site. NMC&A for liquid waste and washed-off shells only. The other solid waster are washed and contains negligible quantity of NM
2) BN-350, BN-600	TK-11 SA per TK: <30 items	Irradiated HEU Oxide	5 or 8,3 k vessels in TK-24 containers Or TK-42 (steel container with up to 230 mm thick walls)	
3) Transport Reactors	3) TK-18 SA per TK: 49	Nitrate of Irradiated LEU	Stainless steel annular vessel: V=5101, H=1,8 m, D=0,8 m. Wall thickness 3 mm, UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> mass up to 1,2 t	
4) Industrial Reactors	Internal cask (container-wagon for transportation inside facilities)			
5) Research Reactors	TK-5			
RChP RT-2 of GChK				
IUE of commercial reactors	Internal cask	Nitrate of Irradiated NU (natural recovered U)	Cylinders: D=800mm, H=1750 mm, t=3 mm	Are reprocessed and stored on site. NMC&A for liquid waste and washed-off shells only. The other solid waster are washed and contains negligible quantity of NM.
Spent assemblies of VVER-1000		Spent assemblies are stopped, no reprocessing		Wastes do not contain NM

These facts should be taken into account when developing and using methods and tools for measuring the nuclear material masses, contents, and isotopics, both at radiochemical plants and at other nuclear facilities.

As for plutonium, it is separated from spent SAs of power and research reactors, IUE from commercial reactors, and is stored in dioxide form at radiochemical plants of PA "Mayak", GChK, and SChK. Only insignificant part of it is used for the fabrication of test fuel assemblies with mixed uranium-plutonium fuel of fast power reactors (BN-350, BN-600) and research ones (BOR-60). Fuel and fuel elements for BN-350 and BN-600 are fabricated at facilities of "Mayak". Fuel elements with mixed and vibro-packed fuel for BOR-60 reactor are fabricated at NIIAR, Dimitrovgrad.

Figure 1 shows schematically the flows of nuclear materials reprocessed and produced at radiochemical plants. Flows and major parameters of products, nuclear materials shipped and received by/at radiochemical plants, as well as some parameters of containers essential from the standpoint of NMC&A are shown in Table 2.

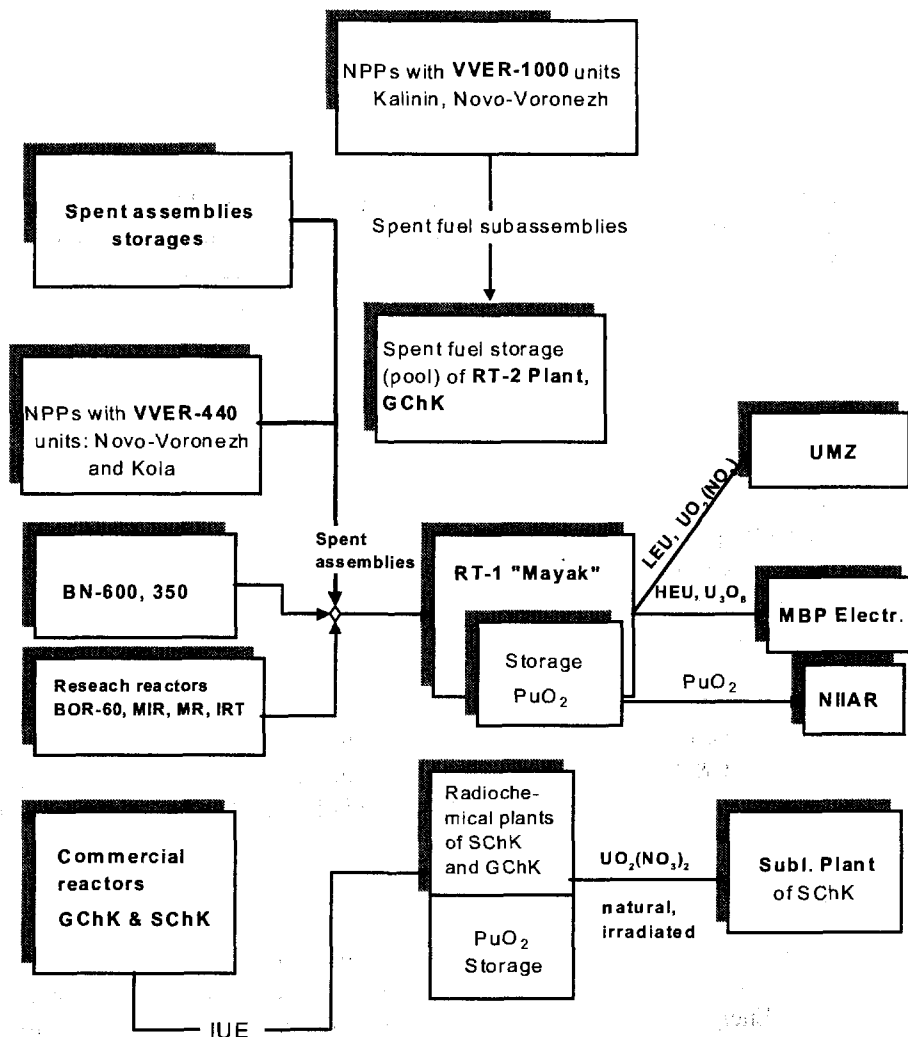


Fig. 1. Scheme of flows of nuclear materials reprocessed and produced at RF radiochemical plants

**REGULATIONS FOR RADIOCHEMICAL FACILITIES IN THE UNITED STATES**

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**Introduction**

The nuclear enterprise as we know it came into being during World War II. Radiochemical facilities involve complex chemical processes that make it difficult for material control and accounting (MC&A) to close material balances with a high degree of precision. A principle tenant in a material protection control and accounting (MPC&A) system is that physical protection provides the protection of the material. Material control and accounting (MC&A) is implemented to provide assurance that physical protection measures have been effective, and in case they fail, to provide delayed detection of the loss. These systems must provide protection against both the external and internal threats.

Material accounting can be based on traditional physical inventories and closing a material balance, process monitoring to provide more frequent tests, or a combination of these two approaches. Typically, process monitoring techniques are used to monitor and track material flows, and limits on campaign uncertainties establish measurement/accounting accuracy requirements. Measurement programs must address both accurate measurements of inputs and outputs, as well as estimation/measurement of in-process inventories and equipment holdup.

Regulations in the United States have been developed that include both performance and compliance requirements to provide a defense in depth approach addressing the unique characteristics of each facility. Regulations address administrative controls, material control and material accounting. MC&A plans are negotiated between regulatory agencies and facilities to develop a site-specific approach. An overview of the regulations and their implementation in the United States is provided.

**Atomic Energy Act**

The Atomic Energy Act was first issued in 1946 to create the Atomic Energy Commission and provide for the control of nuclear energy. The Atomic Energy Act of 1954, as amended, directed the Atomic Energy Commission (AEC) to regulate the receipt, manufacture, production, transfer, possession, use, import, and export of special nuclear material (SNM) in order to protect the public health and safety, and to provide for the common defense and security.

## General Aspects of the State System of NMC&A

The Energy Reorganization Act of 1974 separated functions for the oversight and regulation of the licensed facilities and provided a distinction between government owned nuclear materials and private owned nuclear materials. The licensing and related regulatory functions were transferred to the Nuclear Regulatory Commission (NRC). The NRC regulations are issued as law in the Code of Federal Regulations. Other functions were assigned to the Department of Energy, as well as oversight of facilities operated for the its defense missions for nuclear material production, processing, and use. The DOE requirements are issued as DOE Orders.

### **DOE Orders**

The DOE requirements for Safeguards and Security are contained in the 5630 series of the DOE the Orders. DOE Order 5633.3B contains the primary requirements for material control and accounting. Other Orders in this series address such topics as inspections and assessments and reporting to the national accounting data base. The requirements in DOE Order 5633.3B are subdivided into three functional areas: 1) Program Administration, 2) Material Accounting, and 3) Material Control. Each section contains a combination of compliance and performance requirements.

The Program Administration element of MC&A includes those functions that, while not a direct element of protecting and accounting for nuclear materials, are essential for the MC&A program to perform well. The elements include having documented plans and procedures, training, and a system of internal reviews and assessments to ensure that the program performs as intended.

The Material Accounting element includes those functions necessary to account for the nuclear material and report on its status. The functions are: an accounting system, a physical inventory program, measurements and an associated measurement control program, material transfers, and documentation and reporting.

The Material Control element includes those functions to provide for timely detection. Those functions are: access control, material surveillance, material containment, and detection elements.

DOE has developed a wide variety of guidance documents and training programs to assist sites in developing programs to meet the individual elements of the requirements in the DOE Orders. These included such document are standard formats for MC&A Plans, technical procedures, and technology evaluations, such as for measurements or tamper-indicating devices.

### **NRC Regulations**

The principle requirements for the material control and accounting at licensed facilities are found in Title 10, Code of Federal Regulations, Part 70 (10 CFR 70), "Special Nuclear Material" and Part 74 (10 CFR Part 74), "Material Control and Accounting of Special Nuclear Material. "The requirement in 10 CFR Part 70 were promulgated by the NRC following the Energy Reorganization Act of 1974. These regulations focus on periodic physical inventories and closing a material balance to determine a plant-wide inventory difference (ID) and its associated uncertainty. Some facilities with complex chemical processes were having difficulties in conclusively resolving large inventory differences. A Task Force was established to review MC&A performance at the various facilities and make recommendations to provide greater assurance that nuclear materials have not been diverted (USNRC 1978). Feasibility studies were conducted at several facilities to gain experience and identify specific procedures that might be used. A set of reform amendments were drafted for facilities processing significant quantities of plutonium and highly enriched uranium and issued for review and comment.

The reform amendments were issued for implementation in 1987 as 10 CFR Part 74 Subpart E, along with guidance documents to assist licensees in complying with the new requirements.

These Reform Amendments are a performance-oriented regulation that emphasizes timely detection of nuclear material losses and provides more conclusive resolution of discrepancies. This is accomplished by taking advantage of process controls, production controls, and quality controls already in place. The regulations in Subpart E were divided into process monitoring, item monitoring, alarm resolution, and quality assurance and accounting. Goals and system capabilities were specified, and licensees then developed specific methods to achieve them.

The requirements separate items facilities from bulk handling facilities. Item monitoring defines units with unique characteristics such that removal of nuclear material would be apparent. The intent of this requirement is to ensure timely plant-wide detection of the loss of items that amount to a goal quantity of nuclear material. To achieve this capability, the licensee is expected to verify the presence and integrity of selected SNM items on a periodic basis. The required frequency of tests for missing items is graded according to the relative attractiveness of the material type in the item, the ease with which the item could be diverted without being observed, and the degree of surveillance and containment provided for by the physical security. If SNM is not tamper-saved, stored in a vault, or permanently CAA that provides protection at least equivalent to tamper-saving, encapsulated, or in samples containing less than a threshold amount, it is not considered an item and the SNM is subject to the in-process control requirements for bulk material.

The goal of the quality assurance and accounting requirements in 10CFR Part 74 is to assure that the MC&A system is performing as intended. Because the requirements in Part 74 are performance oriented, each facility must implement a management structure that provide clear lines of responsibilities and a system of checks and balances. Quality assurance requirements address management structure, personnel qualification and training, measurement control, physical inventories, internal controls, and accounting.

### **NRC Guidance Documents**

The principal requirements with respect to SNM licensing are found in Title 10, Code of Federal Regulations, Part 70 (10 CFR Part 70), "Special Nuclear Material" and Part 74 (10 CFR Part 74), "Material Control and Accounting of Special Nuclear Material." Paragraph (b) of §70.22 of 10 CFR Part 70 specifies that special nuclear material control and accounting (MC&A) information must be provided in a license application to show how compliance with the fundamental nuclear material control requirements of §70.58, §74.31, §74.33 or §74.51 will be accomplished. The NRC issued guidance documents to suggest a standard format and content for use in preparing material control and accounting plans. One example is NUREG-1280, Standard Format and Content Acceptance Criteria for the Material Control and Accounting (MC&A) Reform Amendment, issued for the requirements for facilities with significant quantities of Strategic Special Nuclear Material in Subpart E of 10 CFR Part 74. These guidance documents typically provide an intent and scope statement for each requirement and the acceptance criteria that will be used by the NRC in evaluating the adequacy of submitted plans.

The NRC has also issued other guidance and reference documents focusing on specific aspects of MC&A. For example, *Statistical Methods for Nuclear Materials Management* (1988) was issued to assist licensees in choosing statistical methods for specific applications and address both the general background and specific examples.

### **International Safeguards**

The United States is party to the Nonproliferation Treaty. The United States negotiated an agreement with International Atomic Energy Agency (IAEA) for the application of international safeguards in the US, which was signed in 1980. The requirements for international safeguards

at DOE facilities are contained in DOE Order 1270.2B and the NRC requirements are contained in 10 CFR Part 75. The IAEA began implementing safeguards at licensed facilities in 1980. On September 27, 1993, President Clinton offered to make nuclear materials excess to national security needs available for safeguards. In 1994, the IAEA began implementing safeguards at facilities that previously supported the nuclear weapons complex.

### **Summary**

The United States has a well developed regulatory system that has been evolving since the inception of the nuclear enterprise over 50 years ago. The regulations have been developed in a deliberate and process, that included public input as well as that of regulators and regulated. Separate regulations have been developed in the United States for licensed and DOE defense facilities. These regulations have been published and are available to the public. The regulations have evolved over the years as facilities changed, external conditions changed, and experience was gained. Over the history of this regulatory development, radiochemical facilities have presented some unique challenges. These regulations now contain a combination of performance and compliance requirements to allow facilities to develop a site-specific approach to material control and accounting.

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## **EURATOM SAFEGUARDS. SAFEGUARDS VERIFICATIONS IN REPROCESSING PLANTS**

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### **Abstract**

The "Treaty establishing the European Atomic Energy Community" established Euratom Safeguards in 1957. This paper provides a brief historical view of the legal basis for Euratom. It then goes on to look at the specific application of safeguards to large scale reprocessing plants, from a theoretical "model" to the practical application of inspections.

### **History of Euratom Safeguards**

The "Treaty establishing the European Atomic Energy Community", (Euratom Treaty) defines the provisions for the establishment of a safeguards system, in Chapter VII in Articles 77 to 85. The treaty was signed in Rome in 1957. (Ref. 1).

Chapter VII requires the Commission to:

- Satisfy itself that nuclear material is not clandestinely misused, that the provisions of the Euratom Treaty related to supplies (Chapter VI) are complied with and that obligations to external suppliers are respected (Art. 77).
- To be informed of the design of nuclear plants. (Art. 78).
- To approve the techniques to be used for the chemical processing of irradiated materials, to the extent necessary to attain the objectives set out in Article 77.
- Require accounts of nuclear material to be held and reported. (Art. 79).
- Have full access to and be able to make inspections of nuclear material (Art. 81).
- Have the right to impose sanctions in cases of non-compliance. (Art 83).
- Excludes nuclear material used for military purposes. (Art. 84).

In order to fulfil these obligations, the Commission set up an inspectorate, the Euratom Safeguards Directorate (ESD), to deal with the numerous operational aspects involved.

The Directorate is part of Directorate-General XVII (Directorate E) and is based in Luxembourg. It has about 260 staff of whom approximately 200 are authorised safeguards inspectors, having access to any civil nuclear installation in the European Union (EU).

The Euratom Safeguards Directorate has grown with the growth of nuclear material under safeguards. Now, although the nuclear industry is not expanding much, safeguards activities are still expanding due to the presence of large amounts of plutonium throughout the fuel cycle. The challenge to Euratom safeguards results from the large scale reprocessing installations (La Hague in France and Sellafield in the United Kingdom) and the Mixed Oxide (MOX) fabrication plants in operation, or under construction.



## **Legal Basis**

The Euratom safeguards system is founded in European Law and includes as an ultimate step strong sanctions for infringements. It applies to all civil nuclear material from the moment it is mined on EU territory or arrives in any form from outside the EU. Whilst principally concerned with detecting diversion from peaceful to non-peaceful use it also checks that declarations of specific use are correct and that obligations imposed by material suppliers are respected.

To translate the requirements of the Treaty into an applicable scheme of safeguard measures Euratom Regulation Number 3227/76 (Ref. 2) defines the obligations of nuclear plants, the most important being:

The operator has to declare the "Basic Technical Characteristics" (BTC) in the form of a detailed questionnaire. The information required includes an account of the arrangements for handling nuclear material, a description of the nuclear material, a description of the system of nuclear material accountancy control. The precision and accuracy of all determinations and measurements must be established. Any changes in the BTC must be communicated to Euratom. On receipt of the BTC, Euratom can send inspectors to verify the information.

The operator must establish and maintain a system of nuclear material accounts when he starts to handle such material. The part of the facility where nuclear material is to be found is divided into Material Balance Areas (MBA). For each MBA the accounts must record the details of all material which enters and leaves the area. The account must record the quantity, type, composition, obligation etc. of the material. Other inventory changes, e.g. nuclear transformation by irradiation or change of category must also be reported. Separate accounts must be maintained for plutonium, highly enriched uranium depleted uranium and thorium. Within pre-defined limits a physical inventory must be taken and reported to Euratom.

An outline programme of activities and the intention to carry out a physical inventory must be notified.

Certain transfers, e.g. imports and exports of nuclear material must be notified in advance.

Details of the above obligations, specific to each installation under safeguards, are laid down in the "Particular Safeguards Provisions" (PSPs), which is a legally binding document issued by the Commission after consultation with the Member State and the operator concerned.

## **Safeguards at a reprocessing facility**

In the early days of the commercial nuclear era the field of nuclear reprocessing was on a much smaller scale, within the European Communities plants such as Eurochemic, in Belgium and UP1 in France were capable of reprocessing 60 and 300 tonnes of LWR fuel per annum respectively.

As the energy requirements of a nation grew, strategic decisions were made as to the energy source profile for each state. France in particular favoured the nuclear route. This led to an increase in reprocessing capacity at La Hague with the commissioning of UP2 in 1966 which was capable of reprocessing 400 tonnes of fuel. A demonstration facility was also commissioned at Karlsruhe in West Germany in 1971; this plant was capable of reprocessing 35 tonnes of LWR fuel per annum.

With the accession of the United Kingdom to the European Communities in 1974, a large scale reprocessing plant at Sellafield came under Euratom safeguards. This plant is capable of reprocessing 1200 tonnes of Magnox fuel per annum. A further smaller reprocessing facility in Dounreay capable of reprocessing 50 tonnes of specialist fuel was added at the same time.

## Session 1

The global market for reprocessing services has consolidated in the last 20 years, there are currently within the European Union, two large scale commercial reprocessing facilities, located at La Hague and Sellafield.

Modifications at La Hague led to the design throughput of the UP2 facility being increased to 800 tonnes per annum. Additional reprocessing capability in the form of UP3 was commissioned in 1989 again this plant is capable of reprocessing 800 tonnes per annum. At Sellafield the THORP facility was commissioned in 1994 this plant has a throughput of 1200 tonnes per annum and is capable of recovering 8 tonnes of plutonium per annum.

The general characteristics of such a facility i.e.

- Large flow and hold up of nuclear material.
- Inaccessibility of the nuclear material in large areas of the plant.
- The complexity and high degree of automation of the operation within the facility.
- The complexity of the plant nuclear material accountancy system.

All lead to the general conclusion that the implementation of an efficient safeguards system in such an installation presents a significant challenge.

### **Safeguards Approach**

The development of a safeguards approach for a reprocessing facility has to take into account the following:

- The Basic Technical Characteristics provided by the operator.
- This defines the specific measures, which will need to be applied in order to safeguard to the facility
- The goal quantities and timeliness defined by Euratom.

The political goal is to follow continuously each fissile atom, i.e. "gram by gram second by second". Clearly a more practical approach needs to be developed which takes into account, limited resources (financial and human), non-perfect measurement systems, local agreements (e.g. PSPs) and facility conditions (24-hour operations).

The fact that modern plants are computer controlled and that all operating data is generated, transmitted and processed automatically allows Euratom to capture data at strategic points. The authentication / verification systems are based on the transfer and analysis of selected operating data in near real time and in an unobtrusive manner.

This allows the fundamental objective of safeguards to be met practically. That is to say the development and maintenance of a high degree of assurance to the effect that what the operator declares is happening (in operating records, accounting records and reports) is an accurate reflection of what is actually happening.

### **Practical Application of Inspection Activities**

The first stage in defining the Inspection Activities is to consider the physical form of nuclear material within the process, in order to develop the most practicable points for data capture. Euratom has adopted the following model for MBA structure and Key Measurement Points (KMP).

#### ***Receipt and Storage Area. (MBA 1)***

In this area the basic approach is to measure all irradiated fuel before storage. A Containment / Surveillance (C/S) system for maintaining continuity of knowledge before transfer is employed. The C/S system includes optical surveillance, monitoring systems for movements within the MBA, ultrasonic boltseals, which are applied to the container lid and a measurement device for consistency checks on storage containers.

**Head End Area. (MBA 2)**

Material is transferred from MBA 1 into this area where the material is sheared and then dissolved, prior to collection in the accountancy tank. The control is maintained by C/S methods.

In addition to the primary accountancy tool of weight measurement, the output from the MBA is monitored on specific flow routes to determine the volume, density and chemical composition of nuclear material transferred. Other methods such as neutron determination to follow waste routes and C/S on other potential diversion routes are also employed.

**Chemical Process Area. (MBA 3)**

From the previous point where accurate material accounting data is established and verified, the approach is based on material accounting, verification of flows and inventories. The approach is based on independent verification of the input from the accountancy tank, verification of the output at the nitrate stage, a minimum of one verified physical stock-take per year combined with frequent in process stock-taking.

**Plutonium Product Store. (MBA 4)**

All plutonium dioxide product is fully independently verified before entering the store. Continuity of knowledge is maintained by C/S methods, these include optical surveillance, seals and radiation detectors.

**Verification and assurance of nuclear material control**

The hierarchy of Euratom control can be defined as follows.

1. Independent verification of key elements (measurement of irradiated fuel assemblies, calibration of all process vessels, check weights on weighing systems, taking of and analysing samples.
2. Branching and logging of key operator generated data at defined points, (differential pressure signals for tank levels, weighing signals etc).
3. Containment / surveillance measures including for example on operator's equipment after a calibration has been established to increase the difficulty in the production of false operating data.
4. Data transfer, which in this context means the direct transfer of operating data for all external, key internal and inventory points in near real time for subsequent analysis.

**Key implementation features**

A number of novel features have been incorporated in the implementation of the safeguards system, these include.

**Design verification**

The importance of prioritising verification activities with the resources available was recognised at an early stage. As such a classification system for plant components and pipe-work was devised. The classification was based on the safeguards relevance of the particular item, in terms of the quantities of nuclear materials present under normal operating conditions.

### ***Use of Data Loggers***

The operator's raw data signals are being branched and logged at several key points for flow and inventory measurement. The results are then compared with operator declarations to give a valuable validation of the data for the following points.

- Level, weight and density on the input accountancy tanks.
- Level and density on the buffer tanks fed by the accountancy tanks.
- Level weight and density for the plutonium nitrate accountancy and buffer storage vessels.
- Level and density for the recycled liquor from the plutonium conversion line.

The data is collected in a quasi-continuous fashion and is processed in such a way so as to allow transfer's to be detected by a computer algorithm specially developed to detect material movements. In this way a material transfer calculated from the operators branched raw data signal can be compared with the operators accountancy declaration. The signal processing also looks for the corresponding receipt, this allows the detection of diversion or inadvertent transfers (such as splash over in divertors).

This signal processing capability has provided the on-site inspectors with a very sensitive method for the following of nuclear material.

### ***In process inventory taking***

When the operator takes an in-process inventory, samples will be taken and analysed independently, and the inventory itself verified on classical lines making use of the logged operator signals for tank levels etc. and the operators data transmitted on a near-real-time basis. Each inventory is analysed for trends and sequential statistical analytical techniques will be applied to the results of a series of in-process inventories.

### ***Sampling and Analysis***

The inspector using authenticated sample lines witnesses a sample taking. These are placed in tamper-proof sample transport vessels, which are then transferred to the On Site Laboratory (OSL) where Euratom inspector analysts provide a fully independent analytical capability. The provision of an OSL was deemed necessary due to the large number of samples generated at the facility, the difficulty of conditioning samples for transport and the high cost of these transports. Results will be available on a much shorter timescale than if they were transported to an independent laboratory. This of course allows anomalies to be followed up closer to the event.

### ***Operating data generation, transmission and treatment***

A key part of the safeguards implementation scheme is the transfer in near-real-time to the Euratom computer of an extensive set of operating data relating to all external / internal flows. This operating data set will be validated for consistency and will be verified at different levels at chosen points. In this way a high degree of assurance can be built up with regard to plant operations.

### ***Quality Control / Authentication***

The extensive amount of data which is transferred in near-real time to Euratom in itself provides considerable assurance against data corruption at key points as the data sets must in themselves present a consistent picture. The diversity in the different levels of Euratom's independent data also provides additional assurance on the veracity of the data set.

In addition the following possibilities have been foreseen for quality control, authentication and anomaly resolution.

- The possibility to inject known electrical signals into the data loggers to test response.
- The provision of radioactive sources of known strength to various instruments to establish their calibration.
- Classical re-calibration of tanks for weight or volumetric determination.
- Sending test samples to other laboratories to control the performance of on-site analysis.
- The use of rare earth (lutetium, erbium) tracers to resolve anomalies and to recalibrate the accountancy system.

### **The Lessons for Safeguards**

The project to safeguard reprocessing facilities has yielded many important lessons for both Euratom and the operator.

Early consultation is invaluable in building up knowledge of the plant construction, commissioning and operating phases. Additional safeguard features could be built in at the design stage leading to efficient, effective and unobtrusive safeguards under operational conditions.

Extensive use has been made of computers to analyse data. The value of the plant knowledge has become apparent in the interpretation of the analysis provided from these data sets. This requires inspectors to become familiar not only with the specific instrument types, but also be able to adapt this knowledge to plant specific issues.

### **Conclusions**

The challenge to adequately safeguard major commercial reprocessing facilities has led to many novel approaches being developed. These lessons will also benefit other safeguard projects as a result.

Good co-operation between the operator and regulator is essential for the satisfactory installation of adequate safeguard controls.

The use of modern data processing technology combined with other diverse monitoring techniques has shown that a major industrial scale reprocessing plant can be controlled under international safeguards to provide a high level of assurance.

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## REACTOR-GRADE PLUTONIUM INVENTORY TAKING IN THE RT-1 STORAGE PRODUCTION ASSOCIATION 'MAYAK SITE'

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General characterization of Pu physical inventory taking (PIT) system at the RT-1 plant is given in the paper.

### Input Data and Restrictions

1. The Pu PIT must deal with mass, not with items (in this case - containers)
2. The balance estimation quality in the course of PIT must be the same as that of the accounting records when receiving Pu into the storage.
3. The PIT procedure must be without any opening of containers.
4. The PIT procedure must last not more than 3 days.
5. The personnel irradiation from the containers in the course of PIT must not exceed the preset limits.

All these requirements are contradictory and it is impossible to meet them with direct methods. The principal efforts are directed towards the possible solution when these requirements can be maximum satisfied.

The reactor-grade Pu storage facility on the 'Mayak' site consists of two temporary buildings, numbers 104 and 142, built somewhere in the end of 50s. When the RT-1 plant was designed there was no assumption about any long-term Pu storage due to the potential use of closed fuel cycle in which reactor-grade plutonium was planned to be used for MOX fuel production for fast neutron reactors. However after the events happened in 1986 the construction of MOX fuel fabrication plant was suspended and on the 'Mayak' site more than 10 thousand containers (items) with Pu have been accumulated. Probably, it is the biggest Pu storage in Russia.

The containers with plutonium dioxide are designed in such a way that they are hermetically sealed. In order to locate these containers in the above-mentioned buildings special concrete trenches lined with steel were constructed. Each trench consists of several dozens of sockets into which 1-2 containers are installed. The sockets are closed with light steel covers. These covers have staples with steel wire going through them and sealed on the ends. These wire and seals serve as a TID (tamper-indicating device). The current PIT procedure does not require any direct or indirect measurements of Pu if the TID is intact. The PIT procedure is carried out every three months. In the course of PIT all the TIDs are checked and all the data taken at the stage of putting Pu into containers are put down in the inventory summary.

A great scope of work on upgrading Pu physical protection, control and accountability in storages 104, 142 has been performed during the recent two years.

The work is under way with the assistance of US National Labs. The issues of Pu inventory taking are of the highest priority.

## PIT Concept

One of the principal restrictions in the course of Pu PIT in buildings 104, 142 is the requirement not to open containers which makes it impossible to take samples.

So the Pu type and mass can be determined only by indirect measurements. For instance, isotopic composition which characterizes the reactor-grade plutonium is determined by measuring gamma spectrum of Pu natural radiation and Pu mass in the container is calculated based on measuring Pu gamma spectrum and neutron flux with the help of a neutron coincidence counter.

The accuracy of these measurements does not meet the requirements to balance calculation accuracy in the course of Pu PIT.

So the results of above-mentioned measurements cannot be used for the balance calculations but they can serve as identifiers of:

- isotopic composition which allows Pu in the container to be categorized as reactor-grade Pu;
- Pu mass close to the average (the most probable) value of Pu mass put into the container.

The concept of Pu PIT in the storage consists in the following: based on the definite values or identifiers' states a certain conclusion must be made during the period between two PIT's.

The principal item is Pu mass inside the container.

The conditions required for conclusion made about invariability of Pu characteristics to be made are as follows:

The identifiers must be determined and analyzed. The analysis must show that:

1. The item under account (the container with Pu) is still in the storage;

2. TID is intact (there was no attempt to open the container);

3. Pu mass in the container is within the allowable range. This range is determined by the process regulations and statistical data on Pu mass put into the container.

Statistical processing of random samples taken in 1990-1991 showed that the root-mean-square deviation ( $\delta$ ) from the average value of Pu mass in the container ( $M_{av.}$ ) does not exceed 5%, so it is possible to assume that all the containers represent a single Pu item category. And in order to identify a separate container in terms of Pu mass it is sufficient to get such a mass value ( $M_{measured}$ ) which is within the range of acceptable values.

The data of statistical processing showed that this range covers the values from 2800 g to 3100 g.

4. Pu isotopic composition in the container is also within the range of acceptable values. According to the requirements  $^{239}\text{Pu}$  mass fraction must be lower than 90%. Statistical processing of random samples taken in 1990-91 showed that  $^{239}\text{Pu}$  mass fraction ( $M_{239}$ ) in the reactor-grade plutonium dioxide is within the range:

$$64\% < M_{239} < 90\%.$$

In order to determine identifier parameters the following instrumentation is used:

1. a scanner for the container bar code;
2. a scanner for the TID and bar code on the container caps;
3. a gamma spectrometer to measure Pu isotopic composition;
4. a neutron coincidence counter to measure Pu mass in the container or scales to weigh the container with Pu.

The results of measurements obtained with these instruments further on will be called Pu item identifiers.

If the item identifier values in the beginning and ending inventories coincide (in view of acceptable limits) this item is considered intact for the ending inventory.

As the time allowed for the work inside the storage building and with some containers outside the building is limited and the number of items in the storage is too large, not every item is going to be checked with measurements. We are going to deal with item samples.

This is a known equation which is used in order to determine the probability of defected sample detection:

$$P = 1 - e^{-nx/N},$$

where N is a number of containers, x is a number of defected containers, n is a number of containers in the sample under measurement

In view of the fact that in buildings 104, 142 there will be applied several means of access control (video monitors, TIDs for baffles, for containers, etc.) the defected sample detection probability equal to 20% is assumed sufficient.

With this approach the sample of 60 items (all the items are intact) will indicate within the mentioned probability that in the storage the amount of defected items does not exceed 0,4%. The confidence interval in this case will be equal to several dozens of kilograms of Pu.

The attempt to improve the reliability of PIT results in the large Pu storage will require the construction of special installations, introduction of various changes as well as a lot of additional financial and material resources.

So in the course of each PIT we are planning to measure 60 containers.

If all the 60 chosen containers are intact it will be quite sufficient to make the conclusion that all the items in the storage are in the same state as before. And it will be enough just to rewrite the previous records as the data on ending inventory.

If in the sample certain defected items are detected the investigation of possible reasons will be started. The inventory commission will make their decision based on the results of this investigation. It might be the decision about the necessity to perform additional checking with random samples or to change records made in the previous PIT.

### **PIT Procedure and Its Required Frequency**

Currently the PIT in the storage is performed every quarter. In the period between them there is certain checking in order to confirm the identical records in account books and accounting documents.

Additional means of access control (more than 3), computerization and random Pu item sampling will improve the value and efficiency of periodical PITs.

We think that these additional measures will make it possible to increase the period between PITs to 12 months. It will not contradict the principal rules of control and accountancy.

It is planned:

- to check TIDs on the trench covers (wire and seals on caps);
- to measure and read automatically:
  - container bar codes;
  - bar codes on the container cap TIDs;
  - container temperature;
  - Pu mass in the container with a gamma spectrum and neutron coincidence counter or with a piezometric device for weighing;
  - Pu gamma spectrum in the container.

The measurement results will be compared with the ones obtained earlier (comparison between bar codes and with acceptable range of variations in Pu mass and its isotopic composition).

If the measured parameters (identifiers) coincide with the previous records or are within the acceptable range the measurement of random samples will be considered sufficient for the conclusion to be made that all the items are intact and the beginning inventory data can be used as the ending inventory records.



The following measures are planned to be taken:

A random number generator must choose 60 samples (numbers of containers) out of all the containers numbers introduced into the data base. These 60 items are to be measured.

Measurements of each identifier (bar codes and their numbers on the containers' caps, Pu mass and isotopic composition in containers) for 60 items chosen must be processed and tabulated.

In this table the agreement between identifiers and the required values registered in the computer data base, as well as the information that there are no defected items in the samples under measurement are given. This table is a part of inventory report.

The measurement results (all the items are intact) are transferred to the inventory difference computation code.

In the computer data base 4 files are developed.

- $F_1(t_i)$  the file which describes Pu mass in the storage as the beginning inventory.
- $F_2(\Delta t)$  the file which describes containers with Pu received in the period between two PIT's. The data are put into the computer by the operational staff of the RT-1 back-end sector (bar codes, container numbers, TID numbers, Pu mass and isotopic composition).
- $F_3(t_2) - F_1(t_i)$  rewriting as the ending inventory
- $F_4(t_2)$  the file which describes containers with Pu received in the period between two PITs (this file is developed in order to check data in the  $F_2(\Delta t)$  file).

The data are introduced by the responsible custodian as ending inventory.

Then two equations are compared:  $F_1(t) + F_2(\Delta t) = F_3(t_2) + F_4(t_2)$ .

If the equation is true, the data from  $F_3(t_2) + F_4(t_2)$  are rewritten into the  $F_1$  file. These data will serve as the beginning inventory for the next PIT.

## Conclusion

So the proposed PIT procedure is based on measuring indirect parameters of inventory Pu, the so called attributes of items, i.e. containers with Pu.

The method how to assess quality of item attribute measurements is proposed.

If any defect is detected, the container is sent back to the place where it has been filled and packed in order to perform direct measurements of Pu mass.

Taking into account the above-mentioned techniques and the availability of several access control means we propose the period between two PIT procedures be equal to 12 months.



## ON THE ACTIVITIES IN BUILDING A COMPUTERIZED SYSTEM OF NMC&A AT THE SCHK RADIOCHEMICAL PLANT

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### 1. Introduction

One of the most crucial issues for the nuclear states and world community is the assurance of safeguards on non-proliferation and containment of nuclear materials (NM), primarily those which can be used for nuclear explosives.

This problem has been getting particularly urgent lately with respect to the scales of nuclear weapons reduction and involvement in production or transfer to storage of significant quantities of surplus NM.

The key issues in the resolution of this problem turn out NM accountancy, control and physical protection, which meet the state-of-the-art requirements.

Recently these requirements have essentially grown. The project "Development of the NM control and accountancy system model on the example of the SCP Radiochemical Plant (#40 ISTC)" has been fulfilled by the Siberian Chemical Plant in collaboration with a number of organizations since October 1992 through October 1996.

The chief goal of the project was creation of the basis for improving the NMC&A system at the SCP radiochemical plant.

One of the key goals of Project was the use of new criteria and approaches to NM control and accounting, including step-by-step implementation for all the NM flows measurement principles.

The work on Project has resulted in the development of the model for NM C&A system at RCP. When designing the model, the single RCP balance area on Uranium and Plutonium was broken down to four NM balance areas.

The model developed within the Project is being implemented in a few ways. They are introduction of innovative NM measurement techniques, working out regulatory documents, adaptation of computers for control and accountancy etc.

With an aim to secure safety in the most problematic area MBA-2 (Plutonium dioxide production) transition to the real-time NMC&A will be required. These problems cannot be resolved without implementation of computerized system of NM control and accountancy (NMC&A system).

The SCP-LANL Joint Contract signed in August 1997 served the starting point of NM C&ACS at SCP RCP. The contract itself appears sequential to the ISTC Project-40 "Development of the NMC&A System – A complex process vessel on the example of SCP RCP".

## **2. Brief Description of RCP Mission**

The Radiochemical plant has been primarily designed for reprocessing of nuclear materials in the form of irradiated standard uranium blocks of industrial reactors (ISUB) with an aim to separate plutonium and uranium. Plutonium is separated in the form of dioxide, uranium – in the form of uranyl nitrate.

Owing to the recent conversion in the defense industry and broadening of SCP international links the RCP tasks have expanded. The activities in purification of recovered uranium oxide supplied from foreign companies came into being.

The Radiochemical plant comprises a few sections where nuclear materials are kept in process vessels, tanks etc. Nuclear materials available at the plant are in different states of aggregation, the predominant state being liquid – uranium and plutonium solutions of various concentrations. The sections and vessels are interconnected with communication lines. Most vessels and the communication lines connecting them are assembled below the zero datum in the canyons protected by concrete ceilings and separated by concrete bulkheads. At the plant output as well as in the storages plutonium dioxide is kept in canisters. Therefore, nuclear materials at the plant appear both in the predominant bulk-form and in the form of items.

## **3. Description of NM C&A System Currently in Force at RCP**

The C&A system operates on the basis of the Russian Federation law in force (The law of the Russian Federation on “ the use of atomic energy” of 20.10.95, the decree of the President of the Russian Federation of 15.04.94 on “ the Priority Measures on the improvement of nuclear material surveillance and containment system” approved by the resolution of the Russian Federation government #1205 of 14.06.96) and the requirements of the guidance and regulatory documents, which are in force at the facilities and institutions of the Ministry of the Russian Federation for Atomic Energy. Actual work in NM C&A at the plant has been performed in accordance with the internal compliance regulations and rules worked out on their basis . The system of NM control and accounting at the plant aims to secure continuous and systematic control of receipt, use, storing, consumption of NM at all stages of reprocessing, transfer to other SCP subdivisions, timely revelation and prevention of losses, facts of unauthorized uses of NM and/or attempted theft.

### **3.1 *Expeditious accounting and storage of nuclear materials at RCP***

With an aim to conduct expeditious accounting, storage, movement of special products (NM) at RCP the following work is performed:

- registration of special products receipt at RCP;
- account of special products and execution of accounting-technical documents;
- preparation and release of Plutonium dioxide to the storage;
- shipment of special products to SCP and intervening organizations;
- storage of special products, assurance of containment in operations with them;
- personnel clearance for work with special products;
- performance of inter settlements on special products between the plant subdivisions;
- inventory preparation and taking;
- special products book – keeping;
- securing safety in storing, transportation and work with special products.

Expeditious – technical accounting of special products on technological subsets (sections) of the plant carries out a multi staged (about ten sections and one and a half hundred process vessels) NM accounting in the process of Uranium and Plutonium separation from spent reactor fuel. The details of expeditious-technical account are articulated in technological regulations.

The data on NM expeditious accounting, storage and transportation are entered into the expeditious documentation logbook by the process personnel and RCP specialists in special accounting.

### **3.2 Physical inventory taking for nuclear materials**

Physical inventory will be taken every month. The scheme of physical inventory taking is as follows: The procedures of feed receipt and release of products are terminated, part of NM-containing vessels are cleared and washed. In other vessels and tanks the volume of solutions is measured. Before the inventory is taken samples are taken of some products, and analysis is made to determine Uranium and Plutonium concentration in the chemical laboratory, or concentration is determined by the indications of remote analytical surveillance devices.

Determination of accountable (book) ending at the moment of inventory taking will be done as follows:

- Amount of NM received at RCP over the material balance period is counted;
- Amount of NM shipped from RCP during the material balance period is counted.

The data from account books are used for determination of the amount of materials received at RCP and shipped therefrom within the material balance period.

Inventory difference is obtained via subtracting the book ending from the physical (measured) ending. The resulting value of inventory difference is statistically processed in order to assess the results of physical inventory taking with the standard existing at the plant.

Currently NM accounting is being performed without the use of computers. The information featuring arrival of the materials, their movement following the plant scheme, quantity and quality of end products are entered into operational logbooks by hand, the data are copied from the remote surveillance measuring devices. The data on NM concentrations are received over the phone from the lab located in a different building or readings are taken from the remote analytical control devices.

Calculations in NM accounting in the course of inventory taking are also made manually by special accounting engineers. The calculations consist in conducting NM balance calculation over the material balance period and statistical analysis of the inventory difference value.

All documents on accounting and book-keeping are executed manually. Most documents are presented on out-dated forms.

## **4. Nuclear Material Balance Areas**

Currently there are two material balance areas (MBA) at RCP – in Uranium and Plutonium, each actually encompasses the entire plant. Physical inventory according to the regulatory documents shall be taken in-process once a month.

By Project #040 ISTC as a result of the analysis of NM movement scheme, instruments and methods of MN control, as well as NM significance, inventory taking procedures and assurance of more effective safeguards measures, it was offered that the plant should be broken down to four MBAs:

## Session 2

- MBA-01 Pu dioxide storage;
- MBA-02 Section of sorption purification, precipitation and calcination of Pu
- MBA-03 Section of irradiated uranium slug dissolution, processing U and Pu solutions including radioactive liquid waste;
- MBA-04 Storage facility for foreign companies-derived low-enrichment Uranium compounds.

Breaking a single balance area of the plant (RCP) down to a number of areas was aimed to assign process lines to individual balance areas that belong to the same type of composition and arrangement of process vessels, accessibility and attractiveness of MN.

- MBA-01 Plutonium dioxide storage vault and MBA-04, storage facility for foreign companies-derived low-enrichment Uranium compounds, appear the areas where NM undergo no operations on redistribution of quantity, packaging etc. Therefore, these MBAs incorporate items only. In MBA-02 and MBA-03 virtually all NM are in the bulk-form.

### **5. Analogue Selection**

In the course of developing a computer-aided system of nuclear materials control and accounting (NMC&A) for the radiochemical plant, the SCP have contributed significant efforts to studying CSC&A developed both in the RF and U.S.A. These systems were being studied primarily with an aim to select a prototype to be used in implementation of CSC&A at RCP.

Consideration was given to the following systems – COREMAS and EZ MAS LANL, VNIITF CSC&A (Snezhinsk), VNIIEF CSC&A (Sarov), “KI-MAX” RSC Kurchatov Institute.

The following basic conclusions can be drawn:

- In the RF the work on this topic has been pursued by many organizations, but under weak general coordination of these efforts;
- All computerized systems of NMC&A developed (both in the RF and U.S.A.) differ in user's interface, structure of data bases, methods of statistical processing, quantity and types of potential transactions etc., but they have one thing in common. They all have been developed virtually for the items only (canister, batch, fuel elements,...);
- None of the CSC&A analyzed by us can be directly implemented on a facility where NM in the bulk-form are available which is basically the point for MN at SCP RCP;
- The present lack of approved “Basic Rules on MN control and accounting” (BRCA NM) overburdens the development of CSC&A for balance areas of NM in the bulk-form. Implementation of approved quantitative and time criteria by the results of physical inventory taking eventually may result in the need to alter the developed system to bring it into line with BRCA NM;
- The scope of labor and time consumption for the creation of CSC&A for RCP (adaptation of CSC&A SOFTWARE analogues for the RCP needs, development of blocks for bulk-form NM control and accounting) may essentially overrun the time stipulated in the contract;
- Participation of the specialists from the organization whose CSC&A has been taken as an analogue would be preferable in order to improve productivity of work and reduce the time for CSC&A for RCP;
- RCP NM balance areas are equipped with expeditious monitoring devices to various extents. Thus, process tanks hosting instruments for on-line (non-destructive) assay of Uranium and Plutonium concentration are virtually lacking in MBA-03. Therefore account is taken by the results of chemical analyses of the samples. On the contrary in MBA-02 virtually all process tanks carry this outfit;

## NMC&A Overview for Radiochemical Plants

- The early efforts in adaptation of the selected CSC&A analogues for the solution of bulk-form NM accountability problems have indicated that software and database management system applied in these analogues were insufficient. Complex algorithms of NM movement, availability of numerous process situation in the same tanks, unwillingness to employ such conceptions as virtual container (due to computational operation being ponderous and complicated) have led the SCP specialists to the conclusion on the need to additionally apply intelligent means of development (expert systems).

With a view to the abovementioned the SCP specialists offer the following solutions:

1. Within time and finance framework of the contract in place CSC&A shall be worked out and implemented for an individual MBA with NM of high degree of attractiveness and featuring all specialties of NM reprocessing at RCP.

2. He systems developed in VNIIEF (Sarov) and E/Z MAS 1.0 LANL using intelligent environment of SOFTWARE development (expert system) for the tasks of expeditious accounting of nuclear materials in the bulk-form will be selected as analogue for NM C&A at RCP.

### **6. Selection and Validation of the Balance Area for Implementation of the First Phase NMC&ACS at RCP**

The SCP offer MBA-02 as a balance area to introduce the first phase of RCP NM CSC&A. Below comes the outline of the main cases for the selection of this very section of technological process:

- Availability of only one nuclear material on the area (Plutonium);
- In this area the material is available in the most attractive forms for potential adversaries. Attractiveness of NM in MBA-02 is due to:
  - processing equipment is predominantly at "polar" marks with potential access of personnel;
  - NM is available in different forms ( concentrated solutions of plutonium, sludges, plutonium dioxide in the form of green and calcined powder);
  - Interim control of NM inside the balance area is carried out with an error higher than an error of weight-taking method at the MBA-02 outlet;
  - This area encompasses the final processing section where gathering, weighing and interim storing ( as long as 5 days with account for week-ends and holidays) plutonium dioxide casks takes place;
- By contract with LANL the RCP has designed, produced and assembled a gamma-densitometer with a small error of plutonium concentration measurement in a solution ( $<2+3\%$  with  $P=0.95$ ) at the entrance to the MBA-02, which will allow a more accurate determination of a book ending in material balance closure;
- Virtually all process tanks of this MBA are equipped with NDA devices for plutonium concentration and liquid volume, which allows implementation of computer-aided data input into CSC&A;
- In the section of gathering and interim storage of casks according to contracts with LANL and BNL the equipment for more accurate NM accounting and control - electronic platform balance and neutron coincidence counter – has been installed. This allows the measurement results to be obtained in the sections with a computer-aided input to CSC&A.;
- In this MBA periodical shut-down of the technological process is possible to be followed by equipment washing for physical inventory taking;

- The possibility of periodic equipment washing allows the amounts of NM hold-up in the equipment to be specified, as well as the charts of hold-up in time. This will enable the hold-up determination errors to be assessed and the volume of their accumulation to be predicted in case of in-process physical inventory taking and equipment washing.
  - For expeditious control and accounting of NM during the material balance period there is a possibility to set up operation-by-operation accounting of NM.
- Therefore taking into consideration the above arguments MBA-02 was selected by us as the first priority processing section for NM CSC&A implementation at RCP.

## **7. Functional Tasks for the First Phase of C&ACS**

**MBA-02** was selected as the first phase. We have outlined the functional tasks for the first phase of NM CSC&A to be resolved by April 1999:

- Physical inventory taking in MBA-02 as frequent as once a month;
- Data input on the NM quantity of the material received at MBA-02 and shipped therefrom for the calculation of book ending should be performed on-line;
- To calculate the ending inventory the data input on the availability of nuclear material in process tanks (indications by concentration meters, level gauges and results of chemical analyses) have been performed manually;
- To implement statistical analysis of the inventory difference obtained as a result of physical inventory taking.

## **8. Features of Software Designing for the Tasks of In-Process Expeditious Accounting of Bulk-Form Nuclear Materials**

As a result of analyzing the requirements to NM CSC&A it has been found out that the book-keeping methods do not resolve all problems because of non-determination of objects for the tasks of expeditious accounting at RCP. There may be a few reasons for that, they all being interrelated:

1. Most RCP NM are in the bulk-form represented as solutions with different isotopics and concentrations (special products).
2. Special products are kept in numerous tanks interconnected with a system of pipelines through which they are being continuously transferred. As a rule a few special products are fed and released in one tank.
3. In the course of transfer special products are being transformed.
4. Clear geographic boundaries of special products' localization are lacking;
5. Irretrievable losses of special products are normalized (evaporations from the tank system, laboratory measurement losses,...).
6. The regulatory documents take no account of NM hold-up in the equipment in the beginning of cycle (after being washed) as well as in the course of cycle.
7. The regulatory documents identify various methods of measuring physical values required for NM control and accounting with the selection technique lacking.
8. Incoming control of special products as supplied to RCP is not conducted. The contractor's data are taken as the basis.

The above challenges as to posing problems of RCP NM CSC&A appear an evidence of their non-formalization. The tasks of this class are insoluble within the framework of mathematical models. For their solution the models are needed which could be capable of reproducing the

contents of syntactic marker with the use of explicitly indicated semantic means. The like models have been called informational ones, the most widely spread being BD database models (relational, network, hierarchical) and KB database models (production, frame, semantic networks).

In relational DBMSs like Microsoft SQL Server, Oracle Server, the languages of data processing (Transact-SQL, PL/SQL) have been realized in the conventional way, namely, the program modules (scripts, stored procedures) are called in by name. The programmer while designing and debugging a data processing program should reveal a great many all imaginable situations which might pop up in the course of running a common program with different input data. In each point where the operation of one module is completed a single-value transition to another module has to be programmed in the explicit form with an indication of the name of module and the list of the data used therein.

The data management language Transact-SQL constituting an integral part of DBMS Microsoft SQL Server, as well as any other data management language for relational BD does not ensure the call of some modules (rules) on the description of the situations, formation of the method of interrelating the modules (rules) in the process of task solution, which is the requirement for the solutions of the tasks in providing control and accounting of nuclear materials at RCP. These tasks can be solved within the framework of the knowledge base model (KB). This informational model is supported by the expert systems (ES).

At the stage of software development for the first phase with an aim to structure the experts' knowledge (operators, process engineers, safeguards group officers) in the area of NM management in MBA-02, we have been using a system of applied codes "EXPERT-ASU". This system was designed by the RCP programmers, but did not undergo all requisite tests and is unfinished. To solve all problems of expeditious accounting of bulk-form NM using this system essential efforts should be contributed to update it. To satisfy the deadlines stipulated in the contract a commercial product should be purchased.

As an intelligent environment of the development we propose that an instrumental complex G2 of the U.S. Gensym Corporation be taken. G2 represents a graphical object-oriented environment for the creation of intelligent applications of monitoring, diagnostics and management of dynamic media in real and simulated situations. A natural language of designing rules, procedures and models will constitute the basis of all problem-oriented G2-based applied systems. G2 secures parallel execution of rules and procedures, and a logical conclusion on the basis of variable data. The principal advantage of G2 is a possibility to apply it as an integrating component, which due to openness of interfaces and maintaining a vast spectrum of computational platforms makes it possible to easily integrate the currently existing disembodied automation means into a unified management system covering all aspects of production activity.

## **9. Software Technical Solutions for NMC&ACS at RCP**

Structure of network-client-server

Software environment:

Microsoft Windows NT version 5.0 and over – as an operational system of local network.

Microsoft Studio or Visual C++, and Borland Delphi 3 Client/Server – as an environment for designing applied software.

G2 or other expert system for the solution of problems related to expeditious accounting of bulk-form nuclear materials in the course of technological processes.





## REQUIREMENTS AND PROCEDURES OF INSTRUMENTED INSPECTION IN THE SCP RCP MATERIAL BALANCE AREAS

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### Introduction

This paper outlines one of the potential approaches to setting up the procedures of inspection for nuclear material (NM) control and accounting (C&A) on a radiochemical production site using technical facilities, on the basis of the results of expeditious and custom-dedicated inspections with allowance for the system of NM C&A existing at that moment.

At the initial stage the analysis was made of NM flow movement at the RCP process lines, applied devices and procedures to measure material quantity and flows, categories of nuclear materials at various stages of reprocessing.

The preliminary assessment underpinned the proposal to set up NM Balance Areas within the plant:

- MBA-01      Pu dioxide storage;
- MBA-02      Section of sorption purification, precipitation and calcination of Pu;
- MBA-03      Section of irradiated U slug dissolution, processing of U and Pu solutions including liquid radioactive waste;
- MBA-04      Storage facility and facility for dissolution of compounds of low-enriched (up to 1% in  $U^{235}$ ) U.

Over the subsequent period the NM flow in each MBA was analyzed, the key measurement point were established. Material balance and operational documents which currently constitute the basis of the accountancy system at RCP were studied as relating to the development of inspection data acquisition procedure. The material balance documents are presented in the form of "Incomplete Production Inventory-Taking Logs" on all process lines at the plant, which are kept by the authorized personnel involved in accountancy at the plant. These documents contain accounting and actually available (by the results of inventory sample measurements) data on the amount of NMs available in the production line (incomplete production residuals) at the front and back end of the inventory taking process within one month as is customary at RCP. The operational accounting documents are presented by "Logs of Senior Operators' Reports" of the process engineering section and "Process Operations Charts" for each of the process engineering departments on the site.

### Objectives and Goals of Inspection

In the course of inspection of the C&A system the GAN inspectors should focus their efforts on the following objectives:

- Review of compliance of NM C&A status with the requirements of federal rules and regulations, guidance documents of the GAN of the Russian Federation, regulatory documents of the Atomic Energy Use Administration.

- Disclosure of inconsistency of the data contained in the reports to be forwarded to RF GAN on the regular basis with data of internal accounting at the plant;
- Revelation of potential unauthorized uses or diversion of NM significant quantity;
- Disclosure of inconsistency between the stated principal specifications (Design-related Information) and actual specifications and operation of the plant, particularly measuring systems aimed for control and accounting;
- Finding weak points in the system of NM C&A;
- Establishing the cause of the incident in case of conducting inspections on the event of theft, loss or unauthorized use of NM.

Reaching the goals of inspection is based on the review of accounting and operational data submitted by the operator, control of the actual inventory of nuclear materials and maintaining continuity of knowledge of NMs using the methods and TID and surveillance devices.

In the course of inspection complete or random checks of the following should be performed:

- quality and completeness of material balance and accounting/record documents, operational records;
- data on actually available NM inventory taking;
- quality and completeness of institutional measures including personnel training and clearance to work with NMs;
- quality and proper use of NM TIDs;
- quality of measuring systems applied by an operator for NM control.

This paper virtually neglects the issues of inspector's work with material balance and operational records on the facility, which currently appears to be his major activity at the time of inspection, whereas proposed are practical procedures of inspection using diverse hardware, which will allow the oversight efficiency to be upgraded.

Inspection procedures in each MBA are qualified by their own features.

#### **Inspection Procedures in MBA-04**

The input material for MBA-04 is oxides of reprocessed uranium with enrichment 1% in  $U^{235}$  contained in canisters, i.e. this balance area accommodates the 4 - class material in the form of items. Currently oxides of non-irradiated uranium with enrichment up to 5% in  $U^{235}$  in canisters are being interim-stored in this MBA. The uranium compounds with enrichment up to 1% in  $U^{235}$  only undergo processing in MBA-03 at RCP.

C&A inspection is conducted once a year and includes identification and counting of items, checking TIDs on 1% enrichment uranium oxide package. Besides measurements are taken of uranium isotopics and weighing the in-storage packages of up to 5% enrichment in packages by way of random check using statistical plans;

Application of inspection-related surveillance instruments in this balance area is deemed inadvisable.

#### **Inspection Procedures in MBA-03**

The incoming nuclear materials are shipped to the MBA-03 from the reactor plant in the form of standard irradiated uranium slugs (SIUS) fabricated of natural uranium and assembled in batches using a special technique. The amount of uranium and plutonium in a batch is determined through calculations using an appropriate technique on the reactor plant. The outgoing materials for this MBA are nitrate solutions of U and Pu. The MBA outgoing amount of uranium is determined through calculations by the results of volume and concentration measurements.

The on-going technology of SIUS reprocessing ensures Pu extraction factor over 99%, hence efforts should be focused on high - accuracy measurement of Pu amount in the outgoing solution of this MBA represents the input product for MBA-02. This enables the information on hand to be sufficient for real-time balance assessment in Pu without any stop in the plant operation.

Nuclear materials in this balance area can be considered as protected because they are actually in acceptable for the following reasons:

- the products located in MBA-03 have a high radioactivity;
- the operational staff is responsible for the continuous monitoring of nuclear materials being stored in the form of a diluted solution in tanks of a big volume.

Due to that any loss of a significant amount of NM can hardly escape notice. Therefore one inspection of NM inventory a year is considered sufficient for this MBA. But this inspection should be performed simultaneously with the inspections of other plant MBAs which have process connections with MBA-03.

Updating of instruments and procedures aimed to measure the quantity of NMs in the products obtained after SIUS dissolving is viewed necessary for the following reasons:

NM measurements constitute the basis for verification of the results of material-balance records, as well as determination and estimation of the difference between NM amount in slugs as stated by the shipper and that measured in the accounting vessel. The use of these measures allows Pu to be counted in the balance area as frequently as once a month without any stop in the plant operation. Inspection may be carried out based on the data on the initial physical inventory of NM, input and output measurements.

Application of inspection – designed TID and surveillance instruments in this balance area is viewed inadvisable.

### **Inspection Procedures in MBA-02**

Incoming NMs are fed to MBA-02 in the form of nitric solutions with relatively high concentration of Pu. The amount of Pu is taken through calculations by the values of concentration measurements in the solution and solution volume in the tank. The MBA output is Pu dioxide powder in 3l canisters. The Pu amount is taken by weighing measurements with an accuracy 0.1g, isotopics is not determined. NM in this MBA is presented in the bulk form actually in all key measurement points.

C&A system is inspected once a month, and as a rule concurs with dates of physical inventory taking. The inspection operation conditions are as follows:

- movements of nuclear material are not carried out either from or into the MBA, and within it;
  - equipment and tanks whose contents cannot be directly measured or assessed should be emptied and conditioned in advance;
  - NM has to preconverted to a more appropriate form to make measurements;
- In addition the inspection procedures include:
- supervision over sample taking procedures;
  - supervision over sample measurement procedures;
  - quality control and testing of the measurement system performance via application of inspection standards and samples including, if need be, measurements on stand- alone equipment.

The RCP A&C service shall submit the following documents to the inspector immediately before the inspection onset:

## NMC&A Overview for Radiochemical Plants

- report on total ending of NM in MBA on the beginning of the month;
- report on variation of material inventory (receipt/shipment) over a month;
- list of KMP containing NMs at the moment of inspection.

Preparatory measures:

- preparation of a working records form;
- input of end number from the previous inspection on NM in the MBA into the working records;
- sample of KMPs out of the presented list in order to exercise supervision over the procedure of taking samples and over measurement of inventory samples (at least 10% a month);

Sequence of inspection procedures:

- checking of initial values (end results of the previous inspection) together with the A&C service based on the report on NM total ending on the beginning of the month;
- examination of material-balance and operational accounting documents (KMP of NM shipment and receipt in MBA) for proper keeping and agreement of the records therein with the data of NM inventory change submitted to the inspector and the results of continuous inspection on MBA-01;
- supervision over taking and measuring inventory samples in the selected KMPs;
- introduction of all NM inventory changes and adjusted initial values into the working records;
- calculation of material balance book inventory on the day of inspection via the formula:  
$$B = TE + TR - TS;$$
where: TE = Total Ending of NM in MBA on the beginning of the month;  
TR = Total Receipt of NM in MBA; TS = Total Shipment of NM from MBA;
- Introduction of the results of material physical inventory taking (PI) in KMPs containing NM at the moment of inspection into the working records;
- Calculation of inventory difference:  
$$ID = PI - B$$
- evaluation of the results obtained, their consideration together with the A&C service, and resolution of disagreements;
- drafting a monthly report on inspection.

### **Procedures of TID and Surveillance System Application in MBA-02**

As a surveillance system it is sufficient to install into the premises of filtration and calcination section a system with two video cameras of MIVS type which operate in the alternate survey mode with an interval of 5 min.

The procedures of surveillance system application in MBA-02 include:

- checking the system performance once a week;
- scanning videomaterials once a month;
- verification of conformity between the facts of access to NM recorded by the system and the activity stated by the plant operator (by accounting logs), and measures taken on finding the cause of discrepancy.
- finding the causes of short-duration cutoffs detected by the system;
- verification of data on operator's adoption of NM access control measures within the period of the system's being cut off;

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- if necessary, carrying out inspection procedures to determine NM physical inventory (inventory outside the plan).

The procedures of TID application in MBA-02 include:

- application of paper seals on uncoverable components of technological equipment;
- operator's advance notice of the need to open the sealed equipment for the GAN inspector;
- examination of the seal integrity by the inspector prior to opening the equipment and applying a new seal upon the completion of work;
- random check of seal integrity at least of 10% of their total quantity by the inspector once a month.

### **Inspection Procedures in MBA-01**

The Pu dioxide storage facility is located in the isolated part of the technological building and represents a number of isolated compartments.

Incoming Pu dioxide powders fed to MBA-01 in canisters of 3 liter capacity, where Pu amount is determined by the scales.

Impurities are determined by the data on the analysis for the samples taken at the MBA-02 outlet. Isotopics is not determined. No measurements are made with an aim to recheck the presence and amount of Pu at the moment of inventory taking, account number of each canister and total quantity of canisters are checked. The facility operator in this balance area applies technical and organizational TIDs like:

Application of locking and warning devices, seals, label, restriction of access to a minimum number of workers, observation of the "two persons rule" etc. Any book – physical inventory difference calls for appropriate measures to be taken in order to reveal the causes. NM in the MBA is available in the form of items and is classified as Category 1. Therefore continuous inspection is envisaged. Application of inspection – designed TID and surveillance system in the storage section is deemed necessary.

The following measures have to be taken in the course of inspection:

- NDA measurements of isotopics (InSpector devices), as well as measurements of the total NM quantity (by neutron coincidence counters) shipped to this balance area, application of TIDs (seals) and identification tools (bar codes);
- Identification and measurement of items by way of random check using statistical plans;
- Examination of surveillance system materials adopted for inspection purposes and TIDs applied by the inspector beforehand, identification of items by bar codes.

At the initial stage, if need be, the initial inspection of in-storage NM's is conducted by GAN inspectors using "InSpector" – type NDA devices and a passive neutron coincidence counter.

### **Requirements and Procedures to Initial Inspection**

In the course of initial inspection the physical inventory of NM should be checked completely (measured and identified – 100% NM), which are available in the MBA at the moment of inspection.

The inspection program and its date have to be defined by the GAN regional office at least 10 days before the inspection date and brought to the notice of the RCP administration. The RCP administration determines the place and the required personnel for the inspection procedures and gives this information to the regional GAN inspectorate.

## NMC&A Overview for Radiochemical Plants

Two days before the inspection date based on the accounting records the (RCP) C&A service draws up a list of NM physical inventory (list of inventory items) with an indication of an inventory numbers for the canisters, NM mass in each canister, and it is forwarded to the GAN territorial office, after that NM movements to and from the balance areas are blocked.

In the event of NM inventory change within two days after submitting the documents, the administration submits a report on NM inventory change on the day of inspection.

The operations of canisters movement to the measurement bay and back to the storage facility are carried out by the plant personnel.

Prior to the measurements the inspector verifies calibration and performance of the applied instruments by the adequate documents.

Measurements of each canister with NMs are made directly by the GAN inspector using an appropriate technique.

The measurement results including the canister number are recorded in the working records in the computerized form and in the form of print-out, each canister is sealed up with a seal, and a bar code is applied. The canister is sent back to the storage.

The measurement values of NM mass in canister are checked for the conformity with the plant account data.

In the event of difference in NM mass measurement values obtained by the operator or inspector for each canister over the allowable error, the calibration quality and performance of measuring systems used by the operator and inspector is controlled, and the operator and inspector repeat their measurements, find out the causes of discrepancy.

If the discrepancies cannot be resolved in the due course in the process of inspection, the inspector interrupts the inspection and reports in the situation to the management.

Then the regime of continuous inspection is carried out in MBA-01.

### **Requirements and Procedures of Continuous Inspection**

The plant administration informs the GAN inspectorate at least at three days' notice about the planned schedule of NM receipt in the MBA for the forthcoming week.

Each canister with NMs to be received in the MBA is measured by the GAN inspector using NDA instruments, to be followed by application of seals and bar codes.

In the event of difference in the NM mass values measured by the operator and inspector by a value over the admissible error, the calculation quality and performance of the measuring system are checked, the operator and inspector repeat their measurements, the causes of discrepancy are found out.

The GAN inspector counts the items (canisters) kept in the storage facility and identifies them by the bar codes monthly, normally during physical inventory taking conducted by the operator.

Every month the inspector examines the application of TID for NM by the plant administration in the following way:

- analysis of records on the time and nature of access control measures which have been taken;
- analysis of records on periodical check-out of NM TIDs;
- control of the state and serviceability of NM TIDs, check-out of the state of the labels, seals, locking devices used as technical and TIDs for NM, can be conducted selectively on the basis of random samples.

## **Procedures of Inspection-Designed ITD and Surveillance Devices**

Video surveillance systems should be adopted in every room to ensure continuity of information on NM storage and movements, and revelation of unstated movements, thus essentially diminishing the need to recheck the material inventory.

To reach this goal it is sufficient to install a MIVS two-camera system in every room, these cameras operating in the alternate survey mode with a period of 1 min.

The procedures of surveillance devices application in MBA-01 include:

- testing serviceability of the system once a week;
- scanning videomaterials once a week;
- verifying conformity of the facts of access to NM recorded by the system with the activity stated by the plant operator (by registration logs);
- taking measures to find out the causes of discrepancies;
- finding out the causes of short-duration cutoffs detected by the system;
- checking the data on operator's application of NM access control measures within the period of surveillance system being cut off;
- if necessary, performance of inspection procedures aimed to determining NM physical inventory.

Metallic (brass) seals will be sufficient as TIDs.

The procedures of TID application in MBA-01 include:

- application of metallic seals on canisters;
- replacement of seals once a month on 8% of the total number of canisters in storage;
- examination of the removed seals for radioactive contamination;
- sending the seals for identification;

The report on application of TID and surveillance system is drawn up monthly.

Quality control of the measurement system is performed by the inspector once every three months.

In the event of NM shipment from the storage facility the GAN inspector's presence is required in order to replace the seals at the storage position.

The report on inspection in this balance area is drawn up monthly.

In conclusion it should be mentioned that most of the proposed inspection procedures for C&A system, application of inspection-designed TID and surveillance tools are not realized now for a few reasons:

1. Unavailability of requisite equipment – surveillance system, that for item identification, TID, etc.
2. Absence of procedure and inspection standards, and samples for measure quality control of the system employed at the plant;
3. Incapability to take and independently measure inspection samples.
4. NM balance at RCP SCP is closed over the entire plant, without assigning MBAs.
5. Insufficiency of federal-level regulatory documentation base on NM C&A
6. And for a number of other reasons as well.



## PRACTICAL EXPERIENCE WITH NUCLEAR MATERIAL CONTROL AND ACCOUNTANCY IN A LARGE REPROCESSING PLANT

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### Abstract

The first large unit of La Hague reprocessing site, UP3, has been commissioned in 1989, its large uranium and plutonium throughputs made compulsory an effective and efficient nuclear material accountancy and control.

The system used in the old facility commissioned in 1966 was effective but the flows of nuclear material were relatively low and did not need the use of a computerised system for data collection. Although the basic system was sound, it implied the involvement of the operator and that an organisation was dedicated to the accounting tasks. It was not adapted to the large throughputs of the new facility.

The system designed in 1988 was about the same as the previous one, with respect to the organisational structure; the aim was to capture all the data and events related to nuclear material in order to satisfy safety, security and commercial constraints. The facility having been designed with the objective of determining as good as possible nuclear material throughputs and inventories, a computerised system permits accurate and timely data collection, following up of throughputs and inventories, establishment of reports and records requested by national and international authorities.

This paper describes the system implemented in UP3 and provides the results of the 9-year operation experience. It will be insisted on the necessity to perform measurements as accurately as possible in order to have an effective system.

### 1) General

Several reprocessing facilities have been built in France since 1958, the first one located in the South East of France at Marcoule was dedicated to non commercial objectives and reprocess low burn-up spent fuels. The second one named UP2 has been erected on the Cap de La Hague site in Normandy in order to reprocess Magnox fuels arising from the first EDF nuclear reactors. It was completed in 1975 by an head end dedicated to the shearing and dissolution of LWRs fuel when Electricity de France (EDF) decided to abandon the Magnox reactors and to build PWRs. Theses facilities could be called first generation facilities with respect to the handling of nuclear material and to the processing of data. They have provided invaluable information related to the accounting and control of nuclear material.



The following elements of the La Hague reprocessing complex have been built in 1989 (UP3) and 1994 (UP2 800 now named more simply UP2). The handling of nuclear materials, the data processing are based on the best knowledge acquired in the previous facilities and use the best computerised systems.

## **2) Nuclear materials management objectives**

In the first facilities:

The main objective was safety and security, nuclear material accounting was implemented firstly in order to satisfy this objective. For example, if the balance between two workshops of the facilities was assessed too large, the operations could be stopped, the facility emptied and investigations pursued up to the solution of the problem. Nevertheless a centralised organisation was dedicated to the follow up and accountancy of nuclear material. In every workshop, data related to nuclear material were manually recorded and transmitted to the personnel of the above organisation. Commercial objectives were not stringent, nor at the beginning international safeguards constraints.

In the new elements of the complex, the safety objectives are still more stringent than in the old ones but reached by means other than nuclear accounting, nuclear material is inaccessible and the operations are remotely ordered from a central control room. Computers are widely used in particular to collect and process accounting data.

The nuclear material management has the following objectives:

- to meet the national regulations;
- to meet the international regulations, treaties, agreements;
- to satisfy our customers,
- to meet the internal needs of COGEMA.

The national regulations are the most stringent with respect to the accuracy of the nuclear materials determination, to the recording conditions and to the reporting to the State authority of each inventory change. This condition implies that the nuclear materials in waste must also be accurately determined for our authorities, for the organisation in charge of the management of nuclear waste in France and also for our customers.

The facility has also to ensure the following functions:

- arrangement of the production operations as they are planned;
- end product quality control.

## **3) Short description of one unit of the complex and of the process**

Each unit can be divided into areas, each of them related to a specific processing phase.

### **3-1) Reception and storage of spent fuel**

The transport casks containing the spent fuels are unloaded and the assemblies are stored under water in baskets, no measurement is performed at this stage for the operation purpose but the identification number declared by the reactor is verified and logged. For international safeguards purpose, the spent fuels are checked for irradiation by neutron monitors that provide a semi-quantitative determination of plutonium and the ponds as well as the transfers to the further area or out of the facility are under surveillance.

### **3-2) Head end area**

The spent fuel is directed to the head end area where chopping of the assemblies and dissolution of the nuclear material are carried out, the assemblies identification number is verified prior to the transfer to the shearing machine and this number is recorded.

Up to this phase, nuclear material is itemised, after the dissolution the main quantity of nuclear material is in a bulk liquid form while minor quantities are in the hulls and end pieces or in centrifuge sludges.

The nuclear material in solution is directed to the input accountancy tank, in this tank the volume of solution is accurately measured and samples are taken for plutonium and uranium determination.

The hulls and end pieces are rinsed and collected in drums transferred to Non Destructive Assay equipment for plutonium and uranium measurement.

The sludges are rinsed and a sample is taken for plutonium and uranium measurement.

The above described operations aim to determine:

- the quantity of nuclear material actually received from the reactors that is the sum of nuclear material measured in the input accountancy tank, in the hulls and end pieces and in the sludges, it represents the acknowledged quantity, the quantity declared by the reactor being only the result of a calculation more or less accurate, depending upon many factors. The difference between the above quantity and the reactor's declaration is the Shipper Receiver Difference (SRD)
- to know the masses of nuclear material transferred to the process area.

### **3-3) Process area**

This area comprises the uranium and plutonium purification cycles, the uranium-plutonium partition, the uranium concentration and storage, the plutonium conversion and the plutonium oxide conditioning.

The large quantities of nuclear materials have made it necessary to subdivide this area into sectors, this subdivision has been done in such a manner that the material transferred from one area to the following one and to facilities/workshops outside the reprocessing facility (such as the vitrification workshop) is accounted for.

The end products are concentrated uranyl nitrate and plutonium dioxide. Uranyl nitrate is shipped in mobile tanks that are weighed after solution sampling and analysing.

Plutonium is converted into oxalate transformed into oxide by calcination, plutonium dioxide is, after sampling and analysing, conditioned in small cans weighed on accurate scales under the control of the Inspectorate. The final packing is a canister that can contain up to five cans. The canisters are stored in the plutonium storage up to the shipment.

Other end products are liquid and solid wastes, liquid wastes are collected in tanks, nuclear material is measured prior to the transfer to the appropriate conditioning workshop. Nuclear material in solid wastes is measured either at the shipping or at the receiving workshop.

### **3-4) Plutonium storage**

No accounting operation is carried out in this area where the plutonium is only stored under severe containment/surveillance measures. The safeguards Inspectorate measure the plutonium masses in canisters by NDA at the entrance in the storage and before shipment.

#### **4) Implementation of the management system**

The system comprises the organisational structure concerning the material management and the means needed to carry out the related tasks, broadly, the structure is the same as in the old facilities but the means have been considerably increased and improved through the wide use of computers in order to face the difficulties linked to the much larger quantities of nuclear material and to the increased number of tasks.

##### **4-1) Responsibilities**

The first level of responsibility is at the operator level, only the operator can order the movements of nuclear materials and decide of the tasks associated with these movements and with the in process inventory. His decisions are only possible within the limits stated by the management of COGEMA and of the site. As an example, the operator does not select the spent fuels to be dissolved but he decides the time of the dissolution and every operation related to. Each working step of the operating level has his own tasks, e.g. checking and validation to carry out.

The second level of responsibility lies with the analytical services: routine analysis requested by the operator and needed for the day to day operations are carried out by a service that depends upon the manager of each unit; analysis related to accounting operations, more accurate and needing more time, are carried out by an independent analytical service.

The third level of responsibility is the general management of nuclear materials and is carried on by a service (GMN) depending of the director of the La Hague complex.

##### **4-2) How the system works?**

The core of the system is made of a data processing system named "Follow up of Nuclear Materials" (SMBD is the French acronym), every data related to nuclear material, from or to a facility or subfacility or laboratories is processed through this system. The SMBD is integrated in the La Hague data processing network named HAGUENET.

The process area has been subdivided into smaller units called Material Management Unit (MMU), this subdivision makes easier the follow up and inventory verifications of nuclear material because each transfer from one MMU to the following one can be accurately measured.

Before a transfer, the shift operator issues a "transfer slip", in this slip are indicated the starting point, arriving point and volume of the transfer, the number of the analytical request, the concentrations and masses of nuclear material and more if there is a need. The transfer must be accepted by the operator of the receiving unit.

The operator who requests the transfer issues also an analytical request to the concerned service when an analysis is planned or needed for operational reasons, samples are taken by automated sampling benches and directed to the laboratory.

The data and orders related to the analysis are processed through a computer subsystem named ANAH.

In the same manner an "inventory slip" is produced when there is a need, in the slip are indicated: the tank identification number, the volume of solution, the number of the analytical request, the concentrations and masses of nuclear material and more if there is a need.

Each transfer slip or inventory slip has two successive levels, a provisional one as long as every information has not been completed and checked and a final one when the slip has been completed and checked at the second level of responsibility.

Through SMBD, the transfer, inventory slips, analytical bulletins and other elements of information are dispatched to the services/persons who have the need to know and particularly to the GMN service (Nuclear material Management Service).

This service has a wider range of tasks as:

- to decide the assemblies to be reprocessed, to establish the transports orders, to set up reports for the managers and specific reports related to commercial and other activities.
- to set up reports for the national authorities and for the Safeguards Inspectorate and to keep in touch with the Safeguards Inspectorate.
- to check the transfer and inventory slips as well as the analytical bulletins in order to detect inconsistencies or clerical mistakes.

#### **4-3) Examples**

The GMN sets up the following reports in using the SMBD:

##### **Shipper-Receiver Difference**

At La Hague, a commercial batch is made of spent fuels belonging to a given customer; the shipper receiver difference is evaluated, after dissolution of the assemblies and emptying the tanks as far as practicable. It takes due consideration of the quantities remaining in the tanks, the quantities in the secondary outputs such as the hulls and end pieces and the sludges and the nuclear transformation. The decay of plutonium 241 into americium from the unloading date to the chopping date is evaluated. The difference between the quantities declared by the reactor and the quantities measured is the "shipper-receiver difference" as already mentioned in chapter "Head end area".

##### **Accountability reports**

The data collected, validated and processed is transferred in forms related to a given period of time, for example, one month; the form named "Inventory Change Report" reports the inventory changes of the Material Balance Area of the facility. It is electronically transferred to a terminal connected to the centralised accounting office of COGEMA. This office despatches the forms to the National Accounting Service.

By using the transfer slips and the inventory slips, the SMBD permits a more accurate and timely control of nuclear material illustrated as follows in the case of the Dynamic Monthly Inventory (named Dynamic because it is taken without operations interruption) :

- a physical inventory consisting of the sum of the nuclear material in the tanks or equipment of the MMU or MBA;
- a theoretical inventory consisting of the sum of every transfer, in or out of the MMU.

Both are matched and action is taken if justified by the difference.

#### **5) Quality of measurements**

The best "nuclear materials follow up system" serves no purpose if the measurements are not carried out with the best accuracy and reproducibility available; for the beginning, the operator has striven to do the utmost in this direction.

##### **5-1) Measurements in the facility**

The most important accounting element is the input accountancy tank because it determines the quality of the material balance and it is the first time that an accounting operation has been able

to be carried out since the fuel fabrication; the input accountancy tank has been designed, constructed and controlled with the utmost care. The calibration of the tank has been carried out with the assistance of the best experts of France and with the assistance and under control of experts of Euratom and of the IAEA; the calibration results have been verified at several levels. It is possible to state that the volume uncertainty is of a few litres, to be compared to the useful volume of 16m<sup>3</sup>. Verifications are performed during operations to ensure that the quality of measurements remains constant or to correct the deviations.

End products. They are contained in items that are weighed, the accuracy of scales is very high and the scales are verified in using certified weights.

Secondary inputs and outputs. All secondary flows are measured. They are usually of minor importance compared to the main flows but far from negligible with respect to the inventory difference, measurements devices are periodically checked and recalibrated if there is a need.

### **5-2) Analytical determinations**

Analytical determinations deal with numerous determinations on the flows and inventory of nuclear materials. There are about 400 main input and output batches and several thousand of secondary inventory changes and inventory takings per year.

As already mentioned, routine analysis have the sole objective to provide data needed for the day to day operations; they must be carried out rapidly and high accuracy is not requested.

On the contrary, precision and accuracy of the analysis requested for nuclear material accounting is the major objective and timeliness if not a real concern; in addition, precision and accuracy of the analytical techniques must be tailored to the quantity of nuclear material in the concerned flow. As an example, input batches have a paramount impact on the nuclear material balance and must consequently be determined with the higher available accuracy. Analytical strategies have been designed in order to apply techniques that are the best appropriate in our context for dealing with the constraints we have to face: quality of measurements, wide variety of flows, low operating costs, minimisation of liquid and solid wastes.

The analytical techniques result from more than 30 years of experience; from this long and wide experience and the adaptation to technical constraints such as waste reduction, automation, development of physical rather than chemical analytical techniques, have resulted in a "Quality Control Strategy".

The quality control techniques can be divided into two groups:

- a priori verifications which are performed independently from the current analysis;
- a posteriori verifications which are performed in using data from sample analysis.

A priori verifications consist in:

- checking of instruments capabilities with reference materials;
- checking of whole analytical procedures by inter-laboratory round robin.

A posteriori verifications are applied on data which are gathered along the daily practice of analysis.

The analytical schemes are designed to collect as much information as possible:

- two independent methods as often as possible;
- several determinations carried out with each method.

The multiple determinations on a given sample allow evaluation of the quality of results; the great deal of samples gives the opportunity for trend analysis. For instance, drifts or sudden

shifts between two methods can be detected by the means of very straightforward graphical examination of results.

## **6) Conclusions**

The Nuclear Material Control and Accountancy implemented at La Hague has proven to be an effective and efficient tool for the management of the facility. The experience acquired in the previous facilities has been invaluable although the amounts of nuclear materials are hardly comparable. In particular it has shown the necessity to determine as accurately as possible every transfer of nuclear material out of or into the facility or area of the facility, whatever is concerned, in order to establish the best possible balance of nuclear material.

Reprocessing is a subject of concern for the public and is also strongly attacked by the environmentalists; it is an additional reason to be as irreproachable as attainable in order to get the confidence of our authorities and to keep their support.

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**MC&A IN A RADIOCHEMICAL PLANT**

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**Abstract**

MC&A in reprocessing plant in the United States is based on solution measurements of the dissolved fuel assemblies, periodic inventories, and solution measurements of product and waste streams. At the Savannah River Site, SRS, considerable effort has been involved in obtaining accurate measurements of the input and product output streams including both volume analytical measurements. In general, volume measurements are based on differential pressure measurements of level and solution density. Topics being addressed are:

- the calibration of tanks in a reprocessing plant,
- the instrumentation used for the input and product volume measurements,
- the analytical measurement capabilities used for input, output and waste streams,
- the measurement uncertainty of these measurements,
- the inventory measurement techniques used at SRS,
- the frequency of material balances,
- a summary of typical measurement system uncertainties, and
- typical material balance uncertainties achieved at SRS.

An overview of these topics with examples in each of these areas based on SRS experience will be provided.

**Introduction**

The Savannah River Site (SRS) was established in 1950. Its primary mission was to provide strategic isotopes (plutonium-239 and tritium) used in the development and production of nuclear weapons for national defense. The historical production cycle at SRS involved fabrication of metal fuel and target assemblies for irradiation in the site reactors, followed by chemical dissolution, separations, and conversion into solid forms. The current mission is to store, treat, stabilize, and dispose of waste material; manage nuclear materials and facilities; and restore and manage the site's natural resources.

The Savannah River Site is located in southwestern South Carolina. It occupies approximately 198,000 acres (310 square miles), and it is 25 miles from Augusta, Georgia. The site is composed of forest, undeveloped land, wetlands, streams, and lakes. The facilities at SRS include 16 major production, service, and research and development areas. Some of these facilities are not currently operating.

The separation process consists of dissolution of fuel, separation and purification of actinides to a nitrate product. The measurement systems in use at SRS to determine material bal-

ances around this process will be described. The measurement capabilities achieved in a production plant are not always the same as those achieved in a research or a laboratory environment. It is expected that SRS experience parallel much of the experiences in Russia. In general, the error estimates provided are obtained from measurement control programs in place at SRS to control, monitor and estimate measurement error and from tests performed to estimate measurement errors.

A simplified material balance flow chart for the separations material balance is provided as Figure 1.

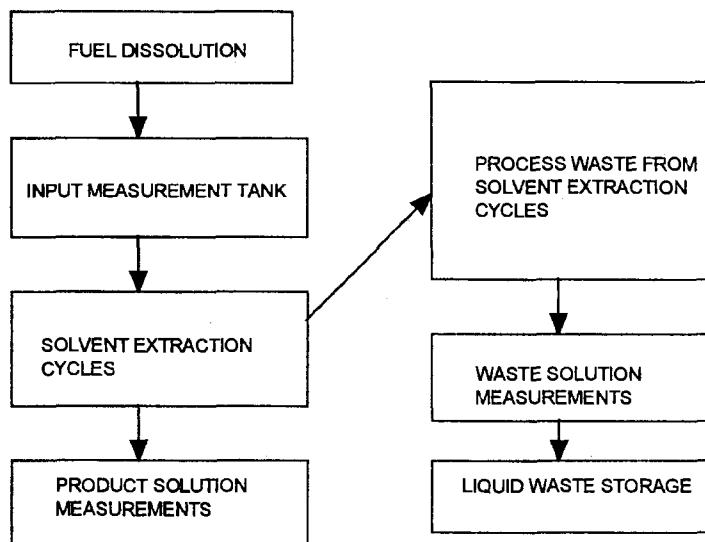


Figure 1. Simplified MC&A Material

### Volume Measurements

The separation plants at SRS use bubbler probes to determine tank volumes. There is a long history of using this technique for measuring solutions in US separations (radiochemical) plants. Obviously, the quality of these measurements is dependent on the measuring instrument and the tank calibration. In the late 1970s to early 1980s, SRS started to use more accurate differential pressure instruments together with extensive tank calibrations to reduce the volume measurement error. Typical volume uncertainties were about 0.5 to 2% for input and output product tanks before this change. Currently all of the input and product tanks in SRS separation plants use the more accurate differential pressure measurements and have detailed calibration equations. The tank volume errors are consistent with the maximum target error limits of uncertainty for primary accountability tanks of 0.3% random and 0.2% systematic (IAEA/ANS/INMM-INMM 1994) <sup>10</sup>In fact, volume measurement errors for input and product measurements were reduced to less than 0.2% at one sigma for both random and systematic errors. For our best calibrations uncertainties of less than 0.1% at one sigma for both random and systematic errors were achieved.

Most of SRS tanks are equipped with the less accurate instrumentation and have volume measurement uncertainties from 1 to 5% random and systematic errors. The calibrations for these tanks are considered adequate based on the amount of material measured. The larger uncertainties do not significantly increase the overall balance uncertainty.



To ensure that volume accuracy's are as expected, SRS has implemented a volumetric measurement control program to control errors and to detect anomalies. The program consists of periodic instrument calibrations, comparison of redundant instruments when possible, comparison of volume transferred from tank to tank and comparison of in-tank density measurements. These types of checks have proven beneficial in improving the reliability of volume measurements and in reducing the number of anomalies.

### **Analytical Measurements**

One of the separation plants at SRS has processed primarily Highly Enriched Uranium (HEU) while the other has primarily processed Plutonium and depleted Uranium. Therefore, analytical measurement methods provided are from analysis of current analytical measurement control data.

The uranium measurement methods in use at SRS are the standard methods in use in the U.S. These are:

- Isotope dilution mass spec for input, impure inventory and waste solutions. The uncertainty of this method as routinely applied at SRS is about 0.5% random and 0.2% systematic.
- Modified Davies Grey (potentiometric titration) method for lower activity uranium solution. This method is used for product solutions and has an uncertainty estimate of about 0.2% random and 0.1% systematic.
- Chemchek (phosphorescence) for low concentration levels. The uncertainty of this method as used at SRS is about 10.0% random and 6% systematic.
- The plutonium measurement methods in use at SRS are the standard methods in use in the US. These are:
  - Coulometry is used for product measurements and as the method of choice to characterize solutions for other methods. The uncertainty of this method as used at SRS is about 0.2% random and 0.1% systematic.
  - Diode Array Spectrometer (DAS) is used to determine plutonium concentration in input solution as well as for many tanks on inventory. The uncertainty of this method as used at SRS ranges from 1 to 5% random and 1 to 5% systematic depending on the concentration and control used.
  - Alpha counting techniques are used to determine plutonium concentration for waste output and inventory. This method includes several adaptations such as gross alpha without corrections, alpha count after removing impurities, or gross alpha with corrections for alpha activity from interfering isotopes (Am and Pu-238). The uncertainty of this method as used at SRS varies from 1 to 5% random and 3 to 5% systematic.

An analytical quality control program is in place at SRS to provide assurances that analyses are reliable and to estimate and monitor method performance. Checks include the analysis of standards prior to the use of each method each shift the method is used. The program uses standards that match the material being analyzed in both matrix and concentration.

As an additional control check on our analytical measurements, SRS participates in a sample exchange program administered by the New Brunswick Laboratory (NBL). Material is sub-sampled with these samples sent to different laboratories in the U.S. NBL collects the data, analyses the results, and issues reports on the performance of laboratories.

## Inventory Measurements

Currently inventories are conducted every two months at SRS separation plants. The inventory steps are:

- cutoff of material flows into and out of process,
- ensure that the mixer-settler systems are brought to standard or known condition for inventory,
- transfer and collect solution in measurable locations,
- mix and sample tanks,
- monitor sample quality from replicate samples,
- analyze and report sample results,
- calculate inventory quantity, and
- compare book value to inventory results

At SRS for several years, each inventory difference is compared to the combined errors for the balance. That is the error estimates for input, output including waste, and beginning and ending inventory are combined by variance propagation techniques to determine total uncertainty on the balance. Care must be taken to ensure that the measurement correlation is correctly included in the analysis. Any inventory difference that exceeds the calculated 99% uncertainty limits is investigated. This is one of the primary ways used to judge the adequacy of the inventory.

## Overall Balance

For many years the inventory differences were tracked and compared to historical limits. The results of some of these inventory differences are plotted in Figure 2 for High Enriched Uranium (HEU). The equivalent data for plutonium is provided in Figure 3. These show that there were long-term measurement biases in one or more of the measurement systems. These plots are for the entire site and reflect more than just the separation balances. However, separation processes were contributors to the cumulative balances. Trends are apparent in much of this historical data.

At SRS for several years, cumulative inventory difference is compared to the combined errors for the balance. That is the error estimates for input, output including waste, and beginning and ending inventory are combined by variance propagation techniques to determine total

### CUSUM for Total Plant Pu239

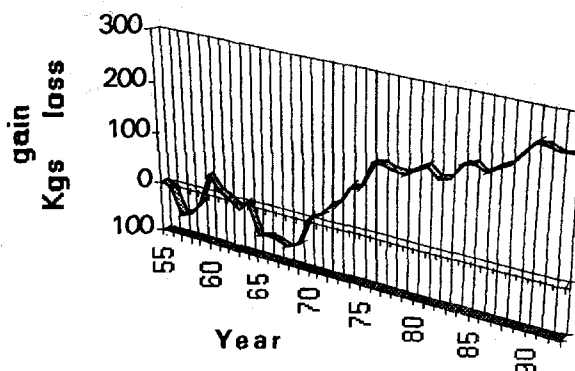


Figure 2

## Session 2

### CUSUM for Total Plant U235 > 20% Enriched

CUSUM = -392.4 kgs U235

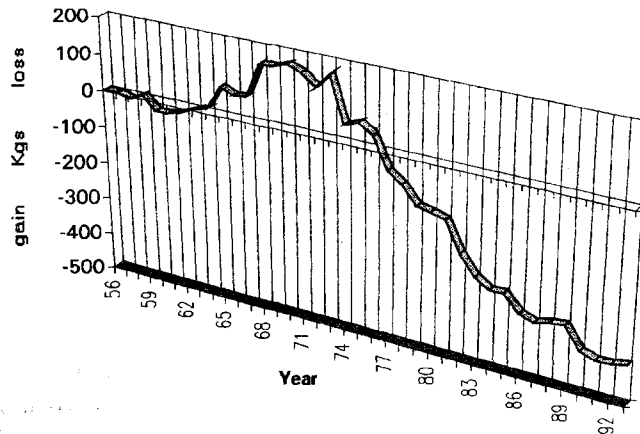


Figure 3

uncertainty on the balance. Care must be taken to ensure that the measurement correlation is correctly included in the analysis. The results from current evaluations are presented in Figure 4 for uranium balances and in Figure 5 for plutonium balances. These are considered typical of performance for the last several years. This represents the magnitude of errors experienced at SRS and what can be achieved from propagating the measurement errors described in the above paragraphs.

To put these data into a meaningful perspective, the cumulative balance data for HEU is about 2% of the active inventory. These numbers are from a time period with relatively small throughput compared to the historical plots.

Similarly, the Pu cumulative balance data is based on relatively low throughput values with decreasing inventory values. These values vary between 2 and 3% of the active inventory.

H-Canyon Total U Cumulative ID Trend  
August 1997 through June 1998

Cumulative ID with 99% Limits

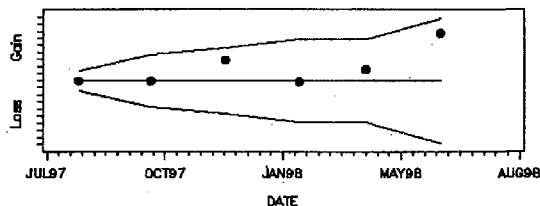


Figure 4

F-Canyon Total U Cumulative ID Trend  
January 1996 through September 1998

Cumulative ID with 99% Limits

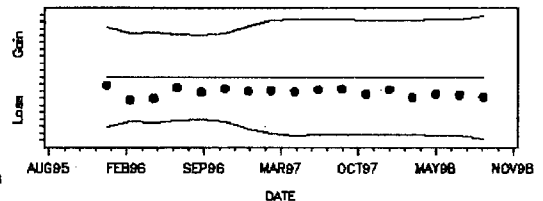


Figure 5

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## EXPERIENCE GAINED WITH NM MC&A IN STORAGE FACILITY FOR PLUTONIUM DIOXIDE OF SChK RADIOCHEMICAL PLANT

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### 1. Introduction

The works for the upgrading of nuclear material accounting and control (NM MC&A) at the storage facility for  $\text{PuO}_2$  were started after concluding the contract with US national laboratories: Los-Alamos, Oak-Ridge, and Brookhaven "Development of express inventory taking of special nuclear materials using barcodes". The storage facility for plutonium dioxide operated at Radiochemical plant of SChK was selected as a demonstration facility for the introduction of bar-coding methods.

### 2. Brief description of storage facility for $\text{PuO}_2$

Plutonium dioxide is received at the storage facility in sealed steel containers from a department of radiochemical plant. The storage facility is subdivided into compartments with locked doors. In the compartments, the containers are arranged in groups, placed in spacing cells. All works for containers transportation and emplacement at the storage are mechanized. Quantification of  $\text{PuO}_2$ , its isotopic, impurities present, etc, is provided at the radiochemical plant department, before the dioxide is placed into containers.

### 3. Equipment used

The following equipment is used at the storage facility for  $\text{PuO}_2$ : personal computer P/133 16 Mb RAM with a source of continuity of service, portable barcode scanner Janus 2010, laser scanners of barcodes 1545E, network controller 9154, laser printer Laserjet 6L, and barcode printer Hercules 200. All barcoding equipment manufactured by Intermec company was shipped to the SChK by Oak-Ridge national laboratory (ORNL), USA. Computer equipment was provided by Steck company, Ekaterinburg; however, when operated, the latter proved to be of low grade, there were problems with repairing needed even before the end of its warranty period.

ORNL experts convened a workshop at SChK and provided valuable help for the application and use of barcoding equipment and arranging it into a network. The SChK specialists were informed on the modules of software transferring the barcodes scanned by portable scanner to personal computer.

#### **4. Computerized system for accounting containers**

The computerized system for container accounting is operated by Windows NT 4.0 system. The user-related part is developed with MS ACCESS 97 (the first version of the client part was written in Visual Basic 4.0). MDB bases are used as the database management system; the inclusion of MSA SQL server is feasible.

The system's functions include:

- Password access to the system;
- Reception of containers at the storage facility with simultaneous application of barcodes;
- Transfer of containers from the storage;
- Availability of containers at the storage with a possibility of correcting the accountancy data;
- Transferring the barcodes scanned from the portable scanner to the computer;
- Comparison of barcodes scanned with those in the database with issuing of reconciliation protocol;
- Preparation and printing of the inventory taking act;
- Printing barcodes;
- Scanning the history of transfers and correcting the data on containers according to sections of reports; all receipts of containers; all shipments; histories for individual containers; deleted entries; all transaction record book.

Any alteration of accounting data on container is registered in the transaction record book with the operation data and operator indicated. The operator has "read-only" access to the transaction record book. The system administrator can access all system tools only.

Databases are archived to optical disks of 640 Mb.

#### **5. Procedures for container accounting at the storage facility**

To date, the following system has been realized for containers accounting at the storage facility:

- All measurements of nuclear materials are performed in the department, before the dioxide is loaded into containers;
- Sealed containers are shipped to the storage;
- Information on the container received is introduced into database, simultaneously corresponding barcodes are printed;
- Barcodes are glued on containers;
- Containers are emplaced into the storage facility compartments
- Monthly inventory taking procedures are implemented in the storage compartments with barcode verification with portable barcode scanner;
- The inventory taking is carried out by a commission;
- Barcodes scanned are compared with those in the database;
- The inventory taking act is printed.

After the computerized system for NM MC&A is put into operation for NM of the whole Radiochemical plant, the workstation installed at the storage facility will be included into the network of the plant. Barcode labels will be printed and applied on items in the department. No barcode printer will be available at the storage facility.

User's guide has been developed for the operation of the system. Since the operator work place includes not only computer, but also barcode scanner and barcode printer, the instruction has the following parts:

## NMC&A at Storage Facilities

- Description of the menu, displays, and functions of the software;
- Algorithms for the following conventional transactions: receipt of containers, shipment thereof, inventory taking;
- Charging the accumulator cells of barcode scanners;
- Actions of operator in abnormal situations in the comparison of barcodes and handling equipment/hardware.

### **6. Further upgrading the MC&A system**

In addition to the planned work for the inclusion of the workstation currently operated into the future computer network of the radiochemical plant, the following is done now for the development of MC&A system at the storage:

- Software testing and design-based assembling for the installation of the system for continuous video-monitoring in the storage compartments are carried out (contract with the LANL, USA)
- Techniques are developed and reference materials prepared for the confirmatory measurements of containers with plutonium dioxide by non-destructive assay with passive neutron coincidence counter JCC-41 (contracts with LANL)
- Works have been started for the procedures of physical inventory taking including the statistical sampling, confirmatory measurements, etc. (contract with BNL).

### **7. Conclusions**

The task for the computerized accounting of containers at the storage with barcoding equipment for inventory taking has been performed to achieve the pre-commissioning phase. This gave the following upgrades:

- Decrease of the time spent by the personnel in storage compartments with plutonium dioxide during inventory taking, thus diminishing the dose for personnel;
- Changeover from the traditional record book to computerized accounting of nuclear materials at the storage, which will make it possible to include the local workstation of the storage into computer network for NM accounting at the Radiochemical plant.
- Test and improve technique for the use of barcoding equipment for further introduction at plants and storage facilities of the SChK.

Works are underway for a further improvement of the NM accounting at the storage for plutonium dioxide.

**SURVEILLANCE SYSTEMS FOR NUCLEAR MATERIALS IN STORAGE**

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As nuclear weapon reductions are occurring in Russia and the United States, large excesses of weapons usable materials are being created and placed into storage. These stored materials, along with materials in storage at existing facilities and sites, pose a challenge for the materials protection, control and accounting (MPC&A) system. Monitoring surveillance systems can be used in a variety of ways to enhance MPC&A system performance and to support the reduction of the physical inventory requirements for frequency and level of assurance. The goal for the use of surveillance systems is the real time event detection and subsequent anomaly identification/investigation. The objectives to be addressed by surveillance systems include:

- Timely and localized detection of changes in the status of nuclear materials whether from normal operations, unauthorized/unplanned incidents, or diversion of materials, or providing assurance that no changes have occurred,
- Verifying that operations involving SNM were conducted in accordance with established procedures and assuring the quality of MC&A data.

There are two general approaches to establishing a monitoring and surveillance system. In one case, containment of the materials is based on boundary and pathway controls. This strategy generally relies on mechanisms to detect any breaches of the containment boundary and provides assurance that all transfers were carried out through defined pathways as authorized. This approach typically relies on area integrity, utilizes video area surveillance, ingress detection through volumetric motion sensors, surveillance of portals/access points, securing/controlling penetrations, etc.

The other approach is material/container oriented as opposed to boundary/pathway containment. The material oriented system focuses on detection of abnormal situations involving unauthorized material movement whether it involves an inadvertent or erroneous transfer, an operational mishap, or a diversion of material. This approach is based on providing positive assurance of material status, i.e., that no changes have occurred in the contained material, and utilizes techniques such as tags and seals, attribute monitoring, comparative video images, etc.

### NMC&A at Storage Facilities

Some simple systems only detect the unauthorized opening of doors, while sophisticated systems can provide continuous monitoring and control of stored items. The enhanced capability of sophisticated surveillance systems allows a facility to compensate for material control and accounting systems that do not provide an adequate level of safeguards protection or reduce some of the inventory requirements as a result of the added detection capability. Program flexibility is based on the ability of surveillance systems to detect unauthorized access and activities. The capability of surveillance systems to compensate for the reduced effectiveness of other protection systems is documented in national and international regulations. The US DOE MC&A order allows for a reduction in the frequency of physical inventories and the draft Russian Federal Regulations allow for a reduced sample size and confidence levels when surveillance and access control systems provide compensatory safeguards.

In the US, a wide variety of surveillance systems are utilized. At one site, a typical storage vault for nuclear materials employs surveillance systems to monitor individual items. This system monitors container identification, presence, shape of the containers, and the heat attribute. At other sites, digital imaging is used to detect the movement of stored items and to detect unauthorized access to nuclear materials in storage. Yet another site uses a load cell based weighing system to detect changes in the weight of each containers residing in a storage rack.

To enhance the capability domestically, automated containment/surveillance and measurement systems are being designed to augment the level of safeguards and to provide continuous inventory verification. In this case, a combination of boundary/pathway controls and material/container approaches are being developed. Container control is based on unique identification of each container, container barcode as an additional identification measure, and tamperproof integrity checks. Transfers are subject to video surveillance monitoring, detection of radiation, movement, and movement direction, and checking of item identification. Containment and surveillance is based on container identification and integrity as well as vault integrity based on application of containment measures and video surveillance. In addition, measurement verification/confirmation is based on isotopic and calorimeter measurements for vault input and transfers are monitored using isotopic and neutron multiplicity counter measurements.

Another site is developing and deploying surveillance systems that have the capability to perform verification measurements of the nuclear material content of stored items. Optical fibers are used for weight measurements and radiation detectors are used for quantification of the nuclear material content. The combination of weight and radiation are sensitive to movement of the container and changes in nuclear material mass.

These techniques generally permit detection of abnormal situations within established sensitivities (many on a real time basis) and can create alarms for investigation. It should be noted that these methods generally do not provide real-time accountability; rather, they utilize real-time surveillance of materials, boundaries, and transfers. (In actual practice, the system may provide real-time monitoring/surveillance and recording of this information, but this does not always provide for real-time assessment of the information.) These mechanisms, in conjunction with the accountability system, provide assurance that unauthorized material removals would be detected or that none have occurred.





## **ENSURING PRESERVATION OF NUCLEAR MATERIALS AT RT PLANT STORAGE FACILITIES BY UPGRADING PHYSICAL PROTECTION, CONTAINMENT (TID) AND TV SURVEILLANCE MEASURES**

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### **1. Introduction**

Storage for civil plutonium in buildings 104, 142 of PA "Mayak" belongs to the most important objects from the standpoint of NM protection. A description of the storage characteristics has been reported previously.

The upgrading of nuclear material physical protection system (NM PhPS) of storage facilities for plutonium dioxide has been carried out at PA "Mayak" in the frame of contract A-0251 between Sandia National Laboratories and PA "Mayak". PA "Eleron" is involved as a subcontractor for designing the TV-monitoring system and system for the detection devices' data acquisition and processing. We consider it necessary to point out very intensive participation of the US specialists at all stages of works.

### **2. Measures for upgrading NM PhPS**

Upgrading physical protection implies the following elements (measures):

- Organizing the local security area;
- Enforcing doors and liquidation of windows in the buildings;
- Preservation of engineering security means available;
- Upgrading physical protection of containers in the buildings listed;
- Sealing the containers' elements for physical protection;
- TV-surveillance;
- Upgrading the building security teams' engineering support
- Re-construction of the system for detection device data acquisition and processing;
- Organization of backup control room for building security.

### **3. Description of technical solutions accepted in the project**

#### **3.1. Local perimeter around storage buildings**

Actual arrangement of the elements of local perimeter is defined taking into account the technology of storage facilities, layout of the surrounding production and auxiliary buildings, plant service lines, provisions for vehicles access, etc.

Total length of the perimeter is 500 m. Many service lines appear under the perimeter, therefore, its structures are made lighter, which will allow subsequently to minimize the expenses for possible emergency works and maintenance at these service lines.

The perimeter is equipped with the following elements for physical protection:

- External and basic fences;
- Anti-ram device made from ropes with breaking load 27 000 kg each. The device must sustain strike of a car up to 15 t mass at 20 km/h velocity;
- The internal limiting barrier from reinforced concrete blocks;
- Complementary detection means arranged within the zone between the main fence and internal limiting barrier;
- Systems for TV-surveillance and electrical lighting, also arranged in the zone between the main fence and internal limiting barrier.

### **3.2. Access control point (ACP) for vehicles**

The ACP is equipped with electrically driven one-wing opening gate;

The external side of the passage for vehicles and gate are equipped with anti-ram means based on metallic rope. The passage for vehicles is also equipped with detectors for smuggled goods possibly hidden in tyre casings of cars (wheel check) and uplifted passage for inspection of vehicles.

As at the main fence of the perimeter, the passage for vehicles is equipped with complementary detection devices.

One video camera is installed at the passage for vehicles. Its picture is displayed not only in the main security post, but on the ACP monitor as well.

### **3.3. ACP building**

The ACP building has two rooms: for people passing and that for security personnel (post). Doors and windows are bulletproof.

The room for people passing are also equipped with enforced glass doors (triplex glass blocks); electrical locks; a cabin is arranged, where detector for radioactive substances is installed. Doors are controlled from the post. There is special inspection window with a unit for providing access cards and documents, made from enforced glass.

The post has windows with protection: one – in front of the entrance door, the other – at the metal detector, to inspect the people passing.

Thus, the sentry can survey those entering, remaining inside the post, and also survey the access to the perimeter, including the control for radioactive substances and metal-containing objects. Besides, there are three video cameras installed in the ACP building: one – in front of the door; the second – for inspecting the vehicles passing; the third – at the inspection window. The picture is displayed to the monitor installed in the post room. The latter is equipped with prompt telephone communication with the main guard force and emergency signaling device, as well as intercommunication systems.

### **3.4. The main and stand-by security buildings of the ACP**

Perimeters of the buildings are equipped with additional engineering devices for restricting access, detecting devices, TV, lighting. The doors in ACP buildings are made from bullet-proof materials, secured against automatic guns.

### **3.5. Engineered devices for protecting nuclear materials in storage facilities**

The storage floor is made from reinforced concrete with channels with nuclear material stored.

A peculiar feature of this design is that reinforced concrete structure of 305 mm thick covers the whole surfaces of the room and provides physical protection of the channels for NM storage.

The permanent part of the building of this design is made from 94 units of 67 fillings, forming a carcass for installation and fixing the elements of removable part of the mass. The blocks are fixed by welding to the filling elements of the floor.

Permanent protection of ventilation boxes of the channels is provided at the level of the mass.

The necessarily large number of units is due to the need to represent the complex layout of the building rooms in the mass design.

The removable part is made of 117 plates of four modifications and 18 lattices. The plates make it possible to access the containers placed in the channels, whereas the lattices are elements of physical protection at the points where filters of the forced ventilation are installed in the channels.

Designs of the units and plates are similar: they have weld frames from reinforcement elements used for construction with facing of black steel sheets covered with concrete. The design ensures a possibility for TID applications.

The lattices are cell-type weld structure from black steel sheets.

The plates and lattices have lock devices with retractable rods of 45 mm diameter from stainless steel for the fixation in grooves of the non-removable part of the mass.

The plates installed at the edges of channels have 4 lock devices. The internal plates and lattices have 3 lock devices each. The lock devices are controlled ("open" – "closed" positions) manually with a special key. Besides, the lock device design includes adapters for installing lock of Mult-Lok brand (for fixing the rod position), or ring bolt (to ensure the possibility of attaching to a hook).

To ensure a possibility to withdraw/install the plates and lattices, the design includes movable cranes A1T12-A10J of 2 tons' load-carrying capacity. Beyond the zone of cranes' reach, manual movable hoists of 1.5 t load-carrying capacity are used. These mechanisms are installed in the room for the reloading operations.

### **3.6. Technical tools for physical protection system (PhPS)**

The design includes upgrading the system for physical protection through the introduction of modern, efficient engineering devices for security (TSO), integrated system for access control and security (SUDOS) "ZIRCONIUM", and the system for TV surveillance (STN) manufactured by "Grundig".

According to the design, the complex of engineering tools is to perform the following functions:

- Displaying and registration of any attempts of adversaries to overcome the TSO-equipped barriers and other actions concerning the object:
  - within the local zone specified;
  - in rooms of building attributed special categories;
  - in the areas of the main and reserve guard teams;
- Automated accounting, control, and management of access of authorized personnel to the local zones and rooms of the buildings;
- Detecting the attempts of unauthorized carrying metal objects into the local area through the ACP and carrying nuclear materials out of these areas;
- Centralized and de-centralized control of peripheral TSO devices operation;
- Remote TV surveillance at perimeters of the local zones, including those inside the storage rooms and guard teams' rooms, including the option of management in automatic regime by signals developed by the ZIRCONIUM system.

We considered it necessary to describe the measures for upgrading the physical protection of storage facility buildings for the following two reasons:

- The implementation of this project is of a great importance for ensuring the preservation of nuclear materials in storage.
- The project envisages a complex approach to the solution of the task: the use of engineered fences, detection means, observation devices, access and control thereof for the personnel; the organization of security force in such way that to give a possibility to take adequate and purposeful actions towards possible adversaries.

The project, in our opinion, can be of interest for specialists. The works for the project implementation have been started, to be completed according to the plan in 2000.

#### **4. Tasks for upgrading physical protection in the frame of laboratory-to-laboratory collaboration between US and PA "Mayak"**

In addition to what has been described above, the facility is working for the solution of several other tasks for upgrading physical protection in the frame of laboratory-to-laboratory collaboration of US national laboratories and PA "Mayak".

4.1. Upgrading physical protection of the RT plant facility for temporary storage of NM. According to the plan, elements of the building structures are to be enforced; special protection device for the nuclear material will be fabricated, and the system for personnel access and TV surveillance will be introduced.

This work will be completed in 1999.

4.2. Equipment of RT plant ACP, the personnel passing these buildings, with devices for detecting nuclear materials and metal objects.

The project was accomplished in 1998.

4.3. Upgrading the system for physical protection of ACP (for vehicles and railway cars) at RT plant

Working documents have been developed.

4.4. Upgrading the radio communication system.

According to the Project, a five-channel tracking system of radio communication for security forces of RT plant is to be deployed.

The equipment has been obtained. The basic system has been initiated, and it is at the experimental stage (operation).

4.5. Upgrading the access control (badge) system of RT plant.

The project is at the phase of engineering detail development.



## **NUCLEAR MATERIAL CONTROL AND ACCOUNTANCY IN A SPENT FUEL STORAGE PONDS**

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### **1. Introduction**

In order to achieve the objective of safeguards, the international control inspectorate provides assurance that the nuclear material under safeguards remains in a peaceful use. The approach to implementing a safeguards system is based on a spectrum of safeguards relevant information concerning facility design, operation and material accounting that is provided to the inspectorate by state authorities and the facility operator, and then verified according to agreed methods and procedures.

The spent fuel storage ponds of a large reprocessing plant such as La Hague in France are under safeguards by means of a wide range of techniques currently used. These techniques include the nuclear material accountancy and containment/surveillance (C/S). Non Destructive Assay (NDA), Design Information Verification (DIV), and authentication of equipment provided by the operator are also implemented. Specific C/S equipment including video surveillance and unattended radiation monitoring have been developed and implemented in a spent fuel pond of La Hague.

These C/S systems named EMOSS and CONSULHA with a high degree of reliability and conclusiveness provide the opportunity to improve the efficiency of safeguards, particularly as related to spent fuel storage areas where the accountancy is verified by item counting.

### **2. Operations in a spent fuel storage ponds of la Hague reprocessing plants**

The scope of the operations considered in the spent fuel storage area is ranged from handling shipping casks containing a number of spent fuel assemblies to transferring individual spent fuel to the mechanical cell. The spent fuel assemblies are issued from Boiling Water Reactors (BWR) and Pressurised Water Reactors (PWR).

The spent fuel transferred to the mechanical cell are assumed to be integral units without prior disassembly and packaging as consolidated fuel rods, but with prior removal of such hardware as channel boxes (BWRs) and burnable-poison 'spider' (PWRs). Damaged assemblies are assumed to be few of numbers and enclosed into a special container.

The storage is underwater in open multi-assembly baskets placed in the pond.

The spent fuel assemblies are unloaded from the shipping casks in a relatively short time after receipt from the nuclear power station and placed in storage baskets in the pond.

The Spent Fuel Assemblies (SFAs) are removed either underwater or directly from the cask into a transfer cell.

Underwater unloading takes advantage of water's shielding and cooling properties, using cranes to place the cask suitably prepared deep in a receiving pond and take off the lid. The SFAs are removed from the cask and placed in a basket for transfer to the storage area in the pond.

Dry unloading involves coupling the cask directly to a transfer cell and removing the SFAs one by one by means of automated transfer system. In either case, empty casks are removed from the unloading station and prepared for return shipment.

Operational control in the storage area is based on identifying and monitoring all transfers of the accountability items being handled and recording the item locations in a computer-based storage map. Most such operation are conducted from a central control room.

### **3. Nuclear Material Accountability**

The control of nuclear materials accountability in the spent fuel storage area of a reprocessing plant is based on item accountability techniques for the individual items identified as containing discrete amounts of nuclear material. Conventional accountability includes verification (by item counting, identification and NDA) of all the items transferred into and out of the storage area (i.e. inventory changes), coupled with verification of the physical inventory (PIV) taken annually of the items within the area. Operational control and safeguards verification methods is needed for both casks and spent fuel assemblies.

#### **Cask Items**

A shipping cask would be loaded with spent fuel assemblies at the reactor site. Verification of the SFA contents is performed at the reprocessing plant during the unloading operations. Casks are intended to be unloaded shortly after receipt at the reprocessing plant, a seal may or not be attached at the reactor site.

The content of the cask is currently verified by safeguards inspectors against the operator's declaration when the cask is placed in the receiving pond or coupled to a transfer cell, and the SFAs are removed, counted and identified for transfer to storage.

#### **SFA Items**

Spent fuel assemblies are handled as individual items in the storage facility upon unloading shipping cask in the receiving pond or in a hot cell for transfer to the storage area.

During unloading the identity of the individual items could be verified for comparison with the reactor operator's declaration by reading the assembly serial numbers using CCTV cameras. This operation cannot be assured for safeguards inspectors in unattended mode. In a pond system, the cameras are underwater units mounted in the wall of transfer channel and in the hot cell, they are radiation-resistant units mounted on the cell wall and fitted with mirrors. Monitoring and attribute testing with system measuring the neutron and gamma emissions could serve to detect gross defects in the individual SFAs.

This system permits unattended verification, by item counting and NDA testing of items transferred into storage area.

During storage, the inventory of SFAs is verified at the annual PIV by:

- Examination of the inventory records maintained by the operator's computer system for nuclear materials accounting, which could provide to the safeguards inspector up-dated information based on records of all SFA movements into and out the storage area.
- Visual counting of fuel assemblies in the storage baskets with Cerenkov Viewing Device (CVD).

The inventories of SFAs in the receiving pond and hot cell are expected to be low or zero at the annual PIV.

SFAs transferred out of the storage area to the mechanical cell are counted and identified on an individual basis. Identification of the assemblies in a transfer basket in the pond at the transfer points is confirmed by CCTV cameras.

### **Containment and Surveillance Measures**

Containment and surveillance measures serve to a major roles in safeguarding the spent fuel storage and handling areas:

- To ensure the completeness of accountancy data on inventory changes, such as spent fuel receipts and transfers;
- To maintain continuity of knowledge for verified items during storage, and thereby reduce the need for item accountancy measures and the frequency of inspections.

The extensive shielding required for safe handling of irradiated fuel provides inherent containment features that facilitate safeguards. Large and heavy casks are necessary for the transport of spent fuel outside a shielded facility. The heavy-duty equipment required for handling such casks limits the number of access ways to a storage facility and the places in a facility where the casks could be unloaded.

Dry unloading of casks requires special coupling to a transfer cell where the spent fuel can be handled remotely within heavily shielded cells. Transfers outside a transfer cell to a storage vault position would require large and highly specialised equipment. Similarly unloading a cask underwater requires a heavy-duty crane and a deep pond. In a pond with a shallow storage area equipped with only medium duty fuel crane, the system would not permit loading SFAs into a normal shipping cask. The containment features limit the types and locations of spent fuel handling operations and could thus contribute to the effective use of surveillance measures.

### **Cask Items**

Surveillance measures are used in cask transfer operations for both dry and underwater unloading.

In the case of dry unloading, optical surveillance is carried out by specially designed radiation resistant cameras located in a port of the transfer cell. The cameras, fitted with mirrors have an optimised field of view enabling the monitoring of the unloading cask area and the basket loading zone before transfer to the storage pond. Underwater CCTV cameras in a storage pond are used to verify basket movement in and out of the hot cell.

These cameras are linked by means of fibre optic network to a recording system called Euratom Multi Optical Surveillance System (EMOSS).

The presence of nuclear materials is determined by gamma and neutron detectors placed above the basket.

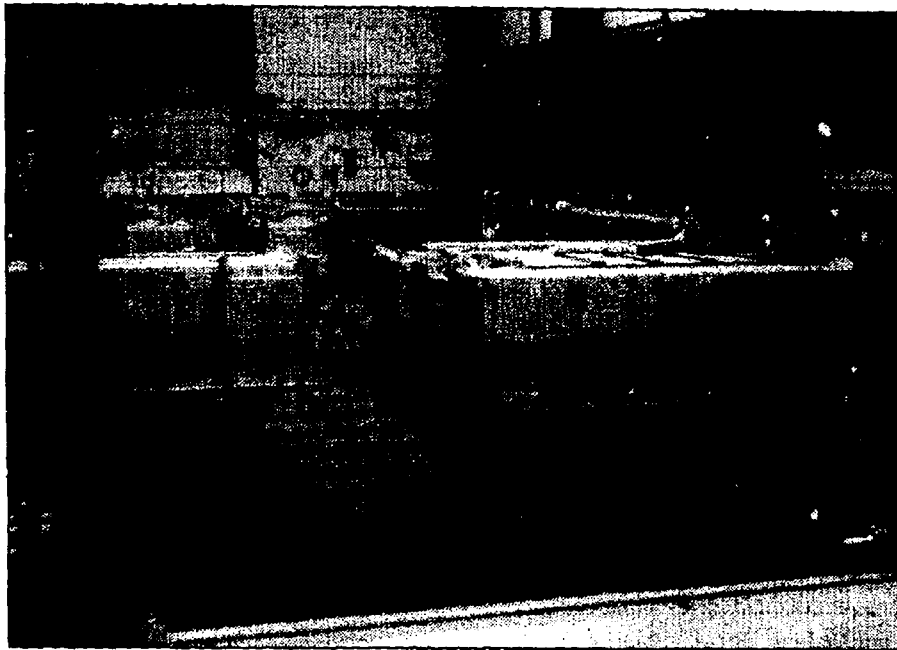
### **Wet unloading**

At the storage pond NPH2 of La Hague an unattended monitoring system, named CONSULHA, and a video surveillance, named EMOSS, have been implemented.

NPH2 is an wet unloading workshop of UP2-800 plant.

This system meets the Agency requirements regarding verification of spent fuel assemblies.

## NMC&A at Storage Facilities



*View of the storage pond of La Hague*

This system includes:

### ***Nuclear radiation detectors***

There are two fission chambers, one acting as a back-up to the other and two gamma detectors (silicon detectors). The silicon detectors are spaced in such manner that the direction of the spent fuel movement can be seen.

A cabinet near the pond contains the pre-amplifier electronics and provides high voltage to the detectors as well as provides amplification and signal conditioning circuitry before transmission to a specific "inspectorate office" dedicated to the Agency.

### ***Video system***

A controller unit handles 2 cameras and provides video compression, front-end motion detection and adequate data storage and retrieval. The EMOSS system provides alarm tampering capabilities for the cabling from the cameras. These cameras are placed in a water-tight box.

In addition a separate EMOSS is located in the "inspectorate office" as the review station.

### ***SFAs Items***

Surveillance measures are used to maintain continuity of knowledge of the verified SFAs in storage during the time intervals between PIVs. All movements of SFAs within the area are recorded by the surveillance system of the safeguards inspectors and in the operator's computer system. The baskets are used directly for storage or as transfer unit that could be emptied by fuel handling machine in the mechanical cell. The outgoing transfer station before the mechanical cell is observed by underwater CCTV camera.

### **The unattended monitoring system CONSULHA**

The CONSULHA (CONTainment SURveillance for Low or High Activity) is an unattended monitoring system for the C/S of spent fuel assemblies.



### Session 3

The main philosophy of this system is to detect any movements of SFA, coming "in or out" of the area detection, in a storage pond, a hatch or a hot cell.

In addition, the systems must be left unattended for a minimum 3 months, and therefore has enough storage capacity to maintain an archive history of the movements (gamma and neutron spectrum) for the defined period.

Components:

In any case, CONSULHA includes:

- Neutron detectors that are fission chambers when the gamma dose rate is high (more than 0.5 Gy/h) or if not, proportional counters.
- Gamma detectors that are chosen according to the sensitivity. They can be either scintillator detectors either silicon detectors.

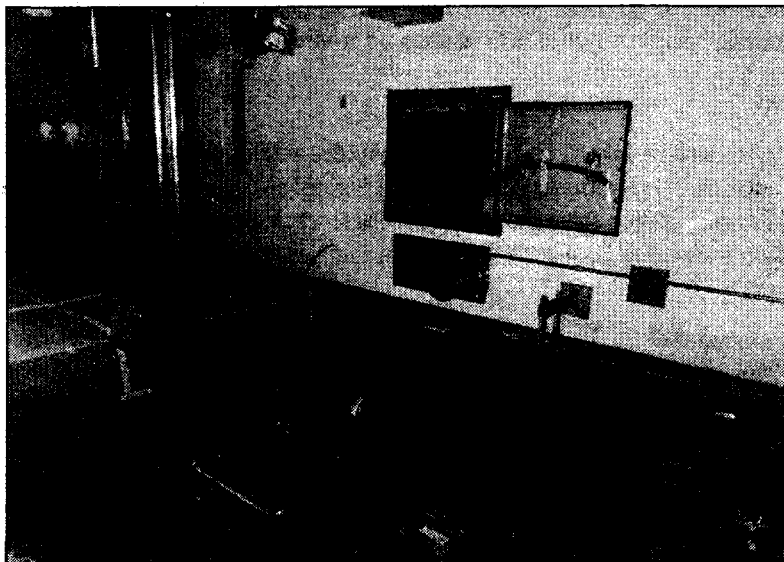
Generally, gamma and neutron detectors are sealed in one heavy stainless steel enclosure. The design of these detectors depends on characteristics of each site.

- Electronics: high voltage, amplifier, an acquisition module,...
- Buffer which allows communication between a computer and the acquisition system.
- Analog data recording device (data logger), for redundant capability.
- Digital recording system (PC data processing computer)

Sometimes, the Consulha system is powered by an uninterruptible power supply which provides back up power in case of main power failure.

In addition, Consulha systems provide authentication and tamper resistance such as:

- Door opening alarm,
- Possibility of the use of passwords by inspectors,
- Date-stamped gamma and neutron radiation data,
- Locked and booby-trapped acquisition software,
- cables protected by stainless steel conduits,



*View 1 of the hot cell R1*

- Physical seals on cabinet and doors.
- 11 CONSULHA systems have been installed:
- 4 in the reprocessing plant of La Hague (in France),
- 4 in the plant Superphenix (in France),
- 2 in the Chernobyl Npp (in Ukraine),
- 1 in the Paks Npp (in Hungary).

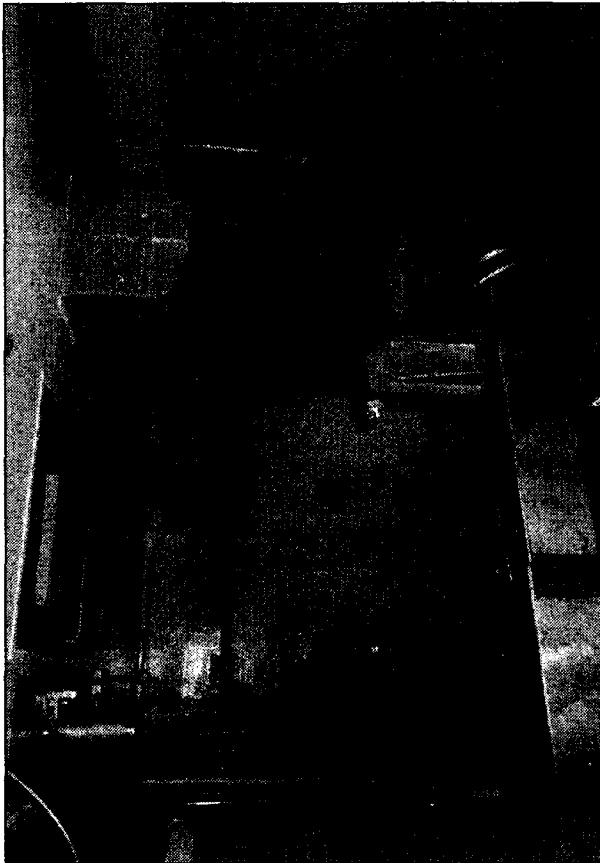
At the reprocessing plant of La Hague, Consulha systems are implemented in order to detect any movements of spent fuel assemblies, when they are unloaded, during their storage in ponds, and before cutting to be reprocessed.

Dry unloading: Consulha is installed in Cell T0 of UP3 plant,

Wet unloading: Consulha is installed in pond NPH2 of UP2-800 plant,

Cutting: One Consulha is installed in Cell T1 of UP3 plant the other one in Cell R1 of UP2-800 plant.

In a period when Agency safeguards face a slow down in budget and an increase in activities in new areas, this unattended monitoring technic plays a very important role. Moreover, the experience gained with the development and the implementation of the Consulha system shows that this technic is not only beneficial for the implementation of Agency safeguards but also for the operator.



*View 2 of the hot cell R1  
where a CONSULHA  
system is implemented*



*View of the CONSULHA  
box implemented in  
NPH2*

### **The video surveillance system EMOSS**

A digital video recording system (EMOSS) with multi-camera video is generally implemented to complement the unattended monitoring system.

This system can monitor at least 4 cameras. These cameras are CCTV and meet the following minimum requirements:

- Minimum sensitivity: 0.2 lux with IR filter or 0.04 lux without IR filter,
- Image sensor: CCD (8.8 mm x 6.6 mm)
- Picture elements: 680 x 490 pixels
- Video output: 1 V, 75 ohms
- Signal/Noise: better than 50 dB
- Operating temperatures: -5°C to + 50°C

Each camera is equipped with a tamper-proof video line indicator. Moreover, generally, the storage medium must be able to at least handle 30,000 images per camera.

Generally, a separate EMOSS is located in the inspectorates office as a review station. This review software can be used friendly and confirms with fuel movements.



## THE MC&A SYSTEM MODEL DEVELOPMENT IN THE RADIOCHEMICAL PLANT OF SIBERIAL CHEMICAL KOMBINE (SChK)

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### Introduction

One of the crucial issues for nuclear states and the world community is to provide safeguards for nuclear materials (NM) nonproliferation and containment. First of all it concerns those materials which can be used for constructing nuclear explosives.

This problem has recently become especially urgent due to a large scale of nuclear disarmament and a large amount of NMs being released, involved into production and transferred to storage facilities.

That is why the state-of-the-art nuclear material physical protection, control and accountancy (MPC&A) is the key issue in solving this problem. The MPC&A requirements have become significantly more stringent. Since October 1994 till October 1996 the (SChK) and a number of other institutions were involved in the project called "Development of NMC&A system model in the SChK Radiochemical Plant".

The main objective of this project was to develop the basis for upgrading the NMC&A system in the SChK Radiochemical Plant.

One of the principal results under the project was the implementation of new criteria and approaches to NMC&A including the step-by-step realization of the principle of measuring all NM flows. It entailed the following:

- the need for application of statistical methods to analyze the results;
- the need for computerization of MC&A system.

It should be stressed that the computerized MC&A system will be developed and implemented in parallel with development and approval of the documents which enforce NM control and accountancy and the procedure of nuclear material physical inventory taking (PIT) for several material balance areas (MBA) for uranium and plutonium. During 1998-1999 new safety criteria will be developed (by the State Scientific Center - Institute of Physics and Power Engineering, Obninsk), different for various MBAs. In order to provide safety (the inventory difference value must not exceed the allowed limit) in MBA-2 (plutonium dioxide production) it will be necessary to perform NM control and accountancy in the real time conditions. It will be impossible to solve these questions without implementation of the computerized MC&A system. The role of this system will grow in time.

The principal objective of the Radiochemical Plant (RCP) is to reprocess nuclear materials in the form of irradiated standard uranium assemblies (ISUA) from commercial reactors to separate plutonium and uranium. Plutonium is extracted in the form of dioxide, and uranium in the form of uranyl nitrate.

Lately, due to the conversion process in the defense industry and expansion of international relations, the RCP has extended the scope of its tasks. They started the work on purifying the reprocessed uranium oxides which are shipped from foreign companies.

The RPC consists of several sections in which NMs are in various states of aggregation and in various concentrations. These sections are connected via the process service lines. Most parts of installations and service lines are assembled below the zero level in canyons which are protected with thick concrete baffles. All the installations are separated with concrete walls.

#### **Description of NMC&A system which exists in the RCP**

The MC&A system in the RCP runs based on the existing legislation of Russian Federation (the law of RF "On the use of nuclear energy", dated 20.10.95, the edict of the president of RF dated 15.04.94 "On the measures of first priority to upgrade nuclear materials accounting system and their containment", "State nuclear material control and accounting system concept", approved of by the Russian Government decree #1205 dated 14.06.96) and requirements of regulatory documents which are in force in the institutions, organizations and plants of MINATOM RF. The particular work on NM control and accountancy in the plant is performed in compliance with internal instructions and documents developed on their basis.

The objectives of the plant MC&A system are:

- to provide constant and systematic control over receiving, use, storage and consumption of NMs at all stages of reprocessing and transferring to other RCP sectors;
- to get the required information about the actual inventory and NM movements at any moment of time;
- the timely detection and prevention of losses or unauthorized uses of NMs and/or attempts of theft.

Currently the MC&A system is not computerized. The information that characterizes the material receipt, its movements in the plant and the final products is registered in the log manually, the recorded data is taken from the remotely controlled instrumentation. The data on NM concentrations is given over the phone from the lab which is located in a separate building or they are registered from the instrumentation displays.

All the calculations in the course of physical inventory taking (PIT) are also performed manually by accounting engineers. They calculate NM balance for the Material Balance Period (MBP) and analyze statistically the value of inventory difference.

All the MC&A documents and reports are worked out manually following a special format approved in the plant.

#### **Nuclear Material Balance Areas (MBA)**

There are two MBAs in the plant today: for uranium and for plutonium. Each of them actually covers the whole plant.

#### **Current MC&A system upgrading**

Under ISTC Project №040-94 all NM transactions within the process limits of the RCP were described in details. The analysis performed resulted in the fact that the plant was divided into four MBAs:

- MBA-01 Plutonium dioxide storage;
- MBA-02 Pu sorption purification, precipitation and calcination;
- MBA-03 Irradiated uranium assemblies dissolution, U and Pu solution reprocessing, liquid radioactive waste included;

- MBA-04 Storage facility and installation for dissolving low-enriched uranium compounds received from foreign companies.

Splitting of a single MBA in the plant into a number of areas was performed with the aim to single out the process sections having similar equipment locations, NM accessibility and attractiveness. For instance, MBA-02 was formed and singled out from the entire process line of the plant because in this area the input material is nitric acid solution with Pu concentration up to 15 g/l, with a relative standard deviation (SRD) for Pu concentration measurement being equal to  $\pm 5\%$ . The output Pu dioxide powder has the SRD for Pu amount measurement equal to  $\pm 0.6\%$ . This fact makes this MBA attractive for potential thieves because there is a high input uncertainty and a much more precise measurement of output Pu amount, so there is a possibility to remove a certain amount of Pu from the process line without being under any suspicion. Taking into account the fact that the installations and process lines are not in the "stone sack" but in the open state with the positive mark relative to the ground level, MBA-02 will be more attractive and vulnerable than MBA-03.

### **NMC&A computerized system**

The MC&A computerized system is designed to automatically account NM reception, movement and storage at the RCP.

The objective of this system development is to provide a constant surveillance over the process material movement, to improve their accountability and administrative work, to upgrade the plant protection against possible NM thefts, stealing and diversion, to rule out any casual errors of operators, to improve the timeliness and significance (reliability) of information about nuclear materials. In terms of its functional goals the MC&A system must meet the following requirements:

### ***Functional requirements to the C&A system under planning***

The NMC&A system at the radiochemical plant should be based on the computerized network. It must keep track of all the material movements in each MBA:

- material receipt from other plant;
- material local movement within the plant;
- material shipment to other plants;
- generation of required documents about NM movements and its accounting.

### **Nuclear material movement**

1. In order to move the material it is needed to have a material identifier which could find the required information in the file.

NM identifiers could be based on:

- the material type;
  - the material location;
  - the number of container or tank.
2. Material transfer from one MBA to another
  3. Material transformation
    - material separation into parts
    - material combination from different parts
  4. Putting material into containers
  5. Material removal from containers

6. Material reception from other plants
7. Material shipment to other plants
8. Possibility to correct errors in the input information

The NMC&A system must provide in each MBA and at the plant as a whole the following:

- recording of all the changes in the key measurement points;
- timely database maintenance covering all the NM movements and transformations in the MBA;
- notification about NMs receipts and shipments in the MBA;
- determination of NM amount in each MBA and at the plant as a whole;
- working out, registration and updating of accounting and reporting data and documents;
- information exchange with the administrative board of the plant;
- control over the correctness of operations with NM movements and changes introduced into the items;
- control over the timeliness of corrections introduced into the NM control and accountancy;
- control and accountancy of personnel access to the automated tools and MC&A logs;
- expeditious search for the location of required products;
- information connection with the physical protection system of the plant;
- required information support to the PIT working groups and inspections.

**Requests on:**

1. Material type
2. Material location
3. Material movement
4. Material movement history
5. Material receipts and shipments
6. Container content

**Physical inventory taking (PIT) procedure:**

1. Data collected from all MBAs
2. Information processing
  - NM balance measured and accounted
  - Calculation of inventory difference and statistical processing of PIT results
3. Generation of the reports

***Administrative board functions in controlling the system***

1. Customers' and administration inquiry service regarding the material management, its availability, location at the plant, etc.;
2. NMC&A and NM database access system control;
3. Information support in the investigation of nonstandard situations related to NM.

**Interfaces**

1. Processing of input information about fissile material availability and movement between MBAs
2. Plant database formation;

3. Output report formation using required format and carriers;
4. Input information compliance control and alarm signals in response to any incompliance detection;
5. Database management, expeditious search and information output;
6. Archive and redundant database copy maintenance.

**Methodological and information support**

***Information support must include the following components:***

- Input information;
- Information about internal movements;
- Output information;
- Regulatory and reference information.

***The principal input information is as follows:***

- The list of MBAs with the information about MC&A procedures and access control systems;
- Accounting documents and reports about each MBA and the plant as a whole covering the period determined in the approved documents;
- Documents with inspection and verification data;
- Special information about NMC&A system operation.

***The following information must be put into the system:***

- Passport data on NMs received by RCP (from the accompanying documents);
- Measurement results;
- Changes in the material characteristics that happened during the technological processing;
- Item movement data;
- Information about people in charge for the materials and any changes in their list;
- Data on changes in the access rules for the MC&A personnel;
- Various reference information;
- Information about directory board instructions for the RCP (about PIT, the next container batch receipt, etc.);
- Results of PITs and PIVs and results of non-standard situation investigation.

***The principal output information is as follows:***

- Data on NM amount, composition, type and location;
- Reports about the actual NM inventory and any changes in it;
- Other types of reports (inspections, special reports if necessary, ect.)

***The system must provide the following:***

- The certain format of accounting documents and reports accepted in separate parts of the plant must have the capability to be transferred into another formats of reports and the data must be selected according to the request in the suitable form.
- Information service must respond to various inquiries of RCP administration, inventory and inspection commissions about the material availability, the state of material processing, statistical reports, detailed or sample data on a certain item.



***The following regulatory documents must be developed for the MC&A computerized system to function correctly:***

- Personnel training programs;
- Computerized system user's manuals;
- Formats of accounting documents and reports.

The system should be use the client-server technology with the use of network amplifiers and long cable paths due to the fact that various process sections are located in various buildings at a certain distance from the shift manager working place.

According to the first approximation the computerized system hardware must include: 25-30 PCs-clients, a server, network components, cable.

The required equipment and structures specifications can be determined more correctly when the following work is complete:

- investigation and description of information transfer routes in the course of accounting data accumulation inside MBAs and in the course of PITs;
- determination of clients' locations in terms of the information to be put into the system, and the way how it can be done (manually, automatically);
- determination of a certain place in the system for the computers used for another tasks: weight, mass, volume measurements;
- determination of the ways and tools with which it will be possible to protect information (at the levels of both hardware and software)
- implementation of the MC&A computerized system.

## **Conclusion**

The requirements mentioned are rather strict. All the system components need to be studied carefully because the insignificant quality of any component of the system impacts the reliability of the whole system.

The information given above cannot be considered as a final one. During the development of NMC&A computerized system various changes and additions can be introduced due to the experience gained at other radio-chemical plants and regulations being developed.

Several issues will be specifically considered during initial design stage of the system.

## **COMPUTERIZATION OF NUCLEAR MATERIAL ACCOUNTING AND CONTROL AT STORAGE FACILITIES OF RT-1 PLANT, PA "MAYAK"**

RU0110725

V. Krakhmalnik, Yu. Menshikov, D. Mozhaev

PA "Mayak", Ozersk, Russia

Computerized system for NM accounting and control at RT-1 plant is being created on the basis of advanced engineering and programming tools, which give a possibility to ensure prompt access to the information required, to unify the accounting and report documentation, make statistical processing of the data, and trace the nuclear material transfers in the chain of its storage at facilities of RT-1 plant.

The system is being developed based on contract No A0251 between PA "Mayak" and Sandia National laboratories. The system is intended for automation of the procedures for NM accounting and control at RT-1 plant.

Up to the present time, the accounting of NM has been carried out without computers. The information characterizing containers with finished products is recorded in operation record book manually, at the department of shipment of finished product; weight of production is written out from remote-controlled measuring devices. The data on NM concentrations in containers and batches are received by telephone from the laboratory, or instrument readings are written out from the remote analytical control instruments.

All documents for accounting and reports are filled in manually. Most documents are made using forms of officially approved patterns.

All procedures for NM accounting and control are regulated by corresponding rules and instructions.

Currently, the accounting is performed in parallel, both by the old methods and with computerized system.

The following functions are performed by the system at the current stage:

- Input of data on the end product's (plutonium dioxide) quantitative and qualitative composition;
- Data input on the location of containers with finished products at storage facilities of the plant and the product's temporary characteristics;
- Selective verification of the data on containers and batches, according to the criteria pre-specified by the user;
- Data protection against unauthorized access;
- Data archiving;
- Report documents formation and providing.

The quantitative composition data are introduced from the technologists-operators' workplaces at the department for finished product shipment by means of dialogue of the operator with the system and automatic recording of the readings from remote-controlled instruments.

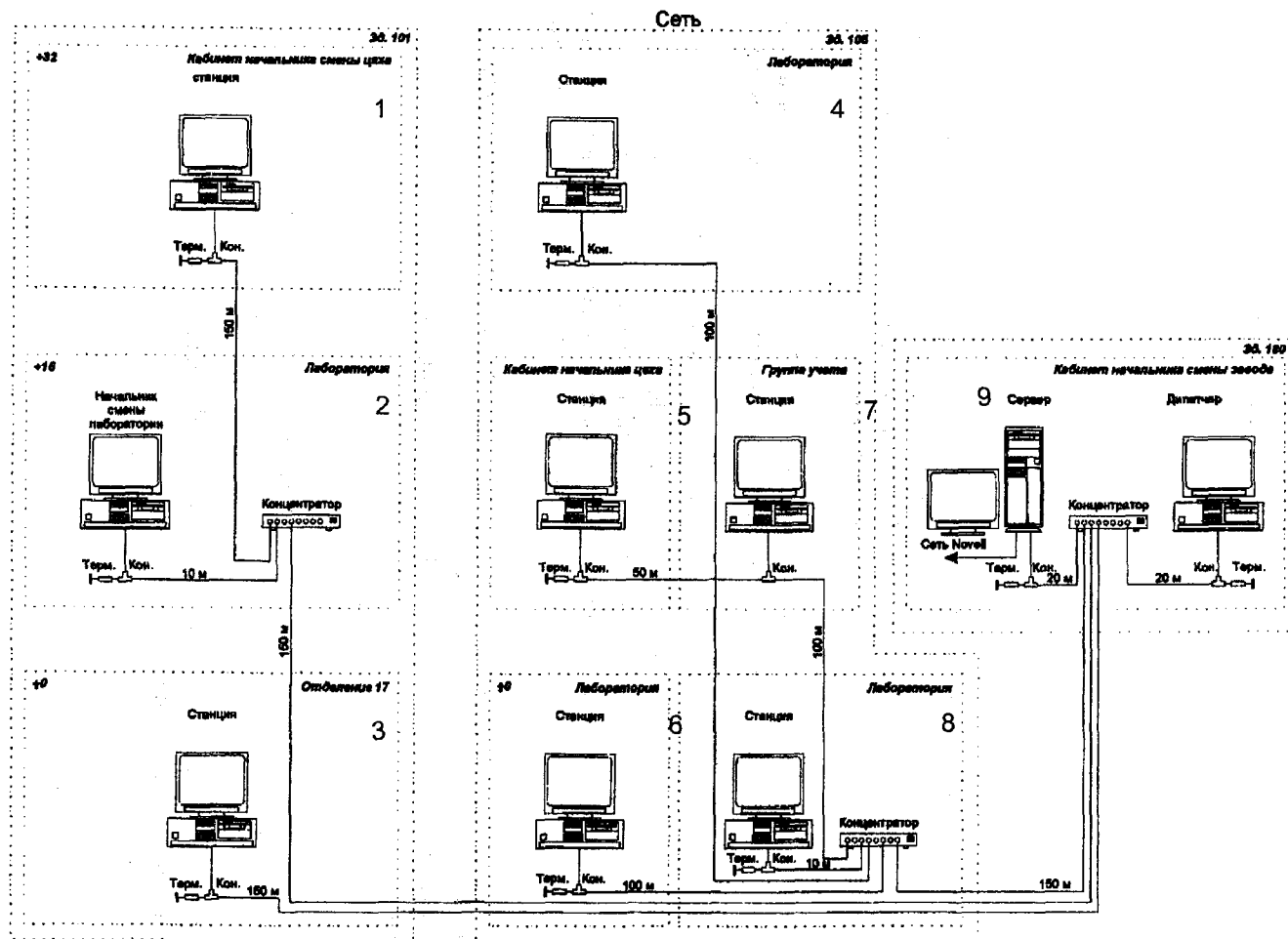


Fig.1. Network

1. Building 101: Office of the chief of department shift. Workstation. Terminal-T-connector 2. Laboratory. Shift manager of the laboratory. Concentrator 3. Department 17. Workstation. 4. Building 105: Laboratory. Workstation 5. Office of department chief. Workstation 6. Laboratory. Workstation 7. Team of accounting. Workstation 8. Laboratory. Workstation. Concentrator 9. Building 180. Office of the shift manager of the plant Server. Dispatcher. Concentrator Novell network

The data on qualitative composition of the end product are introduced from the plant laboratory, points where analyses are performed.

The data on container location at storage facilities and complete sets of the batches are input from workplaces of custodians of the NM special accounting team.

The system's functions include protection against unauthorized access at all levels: both at the level of information input into the system, and data correction and document preparation.

The initial information and output data are stored in databases during an unlimited time.

The computerized system for accounting and control makes it possible to create any kinds of documents for NM accounting and control and any selection of information at the inquiry of users.

The system is functioning all day round in real time.

All information on accounting and control is preserved in case of emergencies (loss of power supply, accidental disconnections, etc.).

The complex of computerized MC&A system engineering support is shown in Fig. 1: the engineering tools are united into a local network Ethernet 10BaseT.

Personal computers of IBM PC type with the configuration shown below are used as workstations:

Pentium 200, 256 Kb cache, 64 Mb RAM, CD-ROM, FDD 3.5",  
HDD 2.5 Gb, VRAM 4 Mb PCI, Ethernet, 32 bit PCI,  
Monitor 17 Glsi, Mouse, Smart-UPS 650 VA.

File-server with the following configuration is used for centralized acquisition and storage of information:

Pentium PRO-200, 2 CPU, 512 Kb cache, 64 Mb RAM,  
CD ROM, FDD 3.5" Ethernet, 32 bit,  
Monitor 15", mouse, Smart-UPS 1000 VA,  
Server certified for operation in WINDOWS NT 4.0.

The system is developed in the architecture client/server, thus preventing the user from direct handling data, decreases a probability of unauthorized access, enhances the system's efficiency as a whole.

Software operates on the basis of Windows NT 4.0 Workstation operation system (OS) (Russian version with documents in Russian) at workstations, and OS Windows NT 4.0 Server installed on the file-server (with license for the required number of users):

Program set for the database development and management:  
Microsoft SQL Server 6.5.

Program set for the application development of database management:  
Microsoft Office for Windows NT 4.0.

Database structure is represented in Supplement 1.

Input data for the system include:

- information on the product quantity and quality in containers and batches;
- information from key measurement points;
- data introduced manually;
- inquiries from personnel;
- reference and regulatory information;
- results of laboratory analyses;
- protection codes.

The data introduced manually and inquiries from personnel are passed to the system as commands from PC, functional keys, commutating equipment.

#### Session 4

Reference and regulatory information (RRI) includes the following data:

- parameter codes;
- product numbers;
- apparatuses' numbers;
- sample numbers;
- regulatory values for parameters;

The following departments of the plant are users of the computerized system for NM accounting:

- team for special accounting;
- bureau for special accounting;
- department (shop) 2;
- laboratory.

Structure of descriptor table for container		
#	Element	Purpose
1	Vessel number	Vessel number (attributed by manufacturer)
2	Container number	The number of container (attributed by manufacturer)
3	Date of container formation	Date of production emplacement into the glass
4	Total weight, g (product + vessel)	Total weight of product and vessel
5	Weight of package (container)	Weight of vessel (indicated by manufacturer)
6	Weight of dioxide	Weight of product
7	Total weight	Weight of container with product (for inventory taking)
8	Sample number	Number of the sample of laboratory analysis for container
9	Extent of moisturizing and undercalcination	Quality characteristics of product in container
10	MED	
11	Container seal of MOL department	Seal markings
12	Container seal by the quality control bureau officer	
13	Location code	Identifier of location at the storage
14	Batch number	Number of the batch the container belongs to
15	Identifier of batch type	Batch type (A,B,C, "N" – standard, "R" – regeneration, "S" – trash collected)

#	Element	Purpose
1.	Number for inventory taking	Inventory number of passport-certificate
2.	Batch number	Product batch number
3.	Sample number	Number of the sample of laboratory analysis for container
4.	Extent of moisturizing and undercalcination	Qualitative characteristics of product in container
5.	MED of 1 kg per batch	
6.	% of main product per batch	% of main product content per batch
7.	% 238	
8.	%239	
9.	%240	
10.	%241	% of isotope content in batch
11.	%242	
12.	Content of impurities (%)	% of impurity content in batch
13.	% Fe	
14.	% Ba	
15.	% Pb	
16.	% Ca	
17.	% Ni	
18.	% B	
19.	% Co	% of impurity content per batch
20.	% Mn	
21.	% Cr	
22.	% K	
23.	% Sm	
24.	% Si	
25.	% Al	



## MC&A PLANNING AND PERFORMANCE TESTING

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### Introduction

Material Control and Accounting (MC&A) regulations have been established as a combination of performance and compliance regulations. Performance based MC&A is based on regulatory standards and implemented in a site-specific approach. In addition to regulatory requirements for performance testing, guidance documents and training programs are available to assist facility personnel developing performance testing programs. A performance testing program is used to assure that the site-specific approach meets the intent of the performance standards. Performance tests may be limited in scope to address a single component of the MC&A system, or they may test the integrated effectiveness of several components or the system as a whole. Statistics of test results are maintained to provide quantitative measures of the MC&A system capabilities. An overview of performance testing as used at U.S. Department of Energy facilities will be provided.

### Regulations

The regulations in the United States specify performance tests be performed by both the sites and regulatory agencies. Sites conduct performance tests as part of the Internal Review and Assessments and also as part of specific Performance Testing programs. These tests serve to two functions: determining performance of the systems and establishing data that can be input into vulnerability analyses. Regulatory Agencies used performance tests to determine regulatory compliance, to assess the effectiveness of the MC&A program, and to assess the effectiveness of policies, procedures, systems, and operations. The elements of the regulatory program are: 1) review site performance testing program, 2) review regulatory oversight program, and 3) conduct tests.

### Key Issues

Performance tests are performed on specific aspects of the regulations or site policy. Some of the key issues in establishing a performance testing program are:

- Identifying what needs to be tested
- Determining how to test
- Establishing criteria to evaluate test results

A performance testing program may be limited in scope to address a single component of the MC&A system, or they may test the integrated effectiveness of several components or the system as a whole. Identifying the need should address regulatory requirements, the system design of the MC&A system, and any vulnerability analyses that have been conducted.

The program elements of a performance testing program consist of:

- Planning
- Coordination
- Conduct
- Evaluation

The planning phase needs to identify the elements to be tested and the type of testing for each of these elements. The planning process should all determine which tests are quantitative and which are qualitative. In addition the tests may be announced or unannounced. Based on the decisions about what is to be tested, schedules and procedures would be developed and submitted for review and approvals.

Before performance tests are conducted, they need to be coordinated to ensure that plant procedures are followed and that the integrity of the MC&A system is maintained. Performance tests should always be coordinated with appropriate personnel at the facility. Some performance tests require that personnel directly tested are unaware that a test is being conducted. Particular care is required to assure that these types of tests are coordinated and safely conducted. Preparatory notifications would be made within the MC&A organization as well as to other site organizations. The notifications need to identify how control will be maintained during tests and that the various organizations can distinguish between simulated situations and actual events. The coordination should establish the protocol for initiation and termination of the tests.

A performance test may be conducted of personnel or equipment. The purposes of equipment performance tests are to: validate that equipment is functional, to validate that the equipment has the proper sensitivity, and to determine whether the equipment will meet its design objectives. The purposes of performance tests of personnel are: to determine whether personnel know and follow procedures, to determine whether procedures are effective, to observe whether plans and procedures accurately describe operational conduct, and to determine whether personnel and equipment interact effectively. Conduct of the performance would include the following elements depending on the complexity of the test:

- Briefings
- Goals
- Methods
- Test Control and Constraints
- Conduct
- Participants
- Controllers and Observers
- Closeout
- Evaluation Criteria

Evaluation criteria need to be established before a performance test is conducted. Criteria may be quantitative or qualitative in nature. Quantitative tests, such as of a measurement system, can use the stated specifications for the system. Written tests or questionnaires can be evaluated



by criteria established for the training program. For qualitative tests, such as observation of activities, evaluation criteria could include such items are

- Procedures were followed
- Correct result achieved
- Appropriate documentation was generated

MC&A exercises are preformed to observe the operation of the system rather than just a component. Exercises are performed to determine that the system performs are described in site plans, and to determine that the system has the capability to meet the goals established by the site. Exercises may be limited in scope because of potential vulnerabilities or safety hazards. Some elements to be tested during an exercise are: command and control, defense-in-depth or redundancy of components, use of information resources, and interface to physical protections program.

### **Functional Areas and Example Performance Tests**

The DOE Orders for MC&A are divided into three functional areas: Program Administration, Material Accounting, and Material Control. Examples Performance Tests may be conducted on Program Administration, Accounting, Measurement and Measurement Control, Inventory, and Containment.

Performance tests in the Program Administration topic address those elements to provide assurance that the MC&A system performs as intended. Tests could include:

- MC&A Training Effectiveness
- Written Tests
- Emergency Response
- Missing Items
- Hoaxes
- Physical Security Alarms

Performance tests of the Accounting topic may be similar to those performed as part of routine audits, or may address detection capability for falsified data. Some types of accounting tests are:

- Receipt Closure
- Data Traceability
- Document Accuracy
- Accounting System Failure
- Material Control Indicators

Measurements performance tests are very similar to the routine activities performed as part of the measurement control program. Example tests could include:

- Scales and Balances
- Nondestructive Assay
- Off-Specification Measurements
- Confirmatory Measurements

Inventory performance tests may be limited to a specific aspect of the inventory program or could be a full scope test. Examples for test of the inventory procedures could address:

- No-Notice Emergency Inventory
- Nuclear Material Locations
- Loss Detection Tests

## Computerization of NMC&A, Material Balance Evaluation, Reports Preparation

- Gross, Partial, and Bias Defects
- Statistical Sampling Plans

Example tests for the containment element could address the following topical areas of material control:

- Material Transfers
- Access Controls
- Portal Monitors
- Tamper-Indicating Devices
- Two-Person Rule
- Daily Administrative Checks

### **Summary**

Performance testing is a valuable tool for determining that the MC&A systems performs as intended. However, when conducting performance tests it is important that care is used to ensure that the integrity of the MC&A system is maintained. The key elements of a performance testing program are establishment of objectives, coordination of each test, and effective evaluation.



## **NUCLEAR MATERIAL DATA MANAGEMENT AND INTEGRATION A SAFEGUARDS PERSPECTIVE**

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### **ABSTRACT**

This paper describes the requirements for a data management system capable of meeting the needs of the various organizations in a complex nuclear facility. Implementation of a material control and accountability (MC&A) systems requires a significant data management capability in order to maintain cognizance of quantities, locations, and activity history of nuclear material (NM) on inventory. For most facilities, this can only be achieved by using a computerized database with auxiliary software capability for performing the data manipulation required for NM safeguards. The primary MC&A activities responsible for data generation are measurements, internal and external transfers, and performance of physical inventories. However, MC&A should not be considered as a stand-alone system with respect to the generation or use of nuclear material data. Data from other activities associated with nuclear facilities (e.g., process operations, environmental protection and safety programs, and physical protection systems) generate data that can be used in performing safeguards activities. Similarly, data generated by the MC&A system may be of value to other organizations. Results from safeguards measurements can provide assurance that processing operations are running as expected, while processing measurements and flow data can provide safeguards assurance. A fully effective data management system must have the capability to ensure that all relevant data is available to all potential customers.

### **INTRODUCTION**

This paper is a discussion of the use of available data in the performance of nuclear material (NM) safeguards. It does not attempt to describe the computer hardware and software used to perform nuclear material accounting. For technical information on computerized accounting software and database applications see references 1-3. Nuclear facilities are required to maintain records to manage and control their NM inventory. The relative difficulty of the task of tracking NM inventories varies with the complexity of the facility's operations involving NM (reference 4). In general, NM processing facilities require more complex accounting systems to meet nuclear material control and accounting (MC&A) requirements than do storage or research facili-

ties. For most facilities, automated, computerized accounting systems are required to meet NM accounting requirements. The primary responsibility for managing data for control of NM rests with the MC&A accounting system; however, there are other sources of data that can contribute significantly. The following discussion considers the various sources of data and the system requirements for collecting and managing that data, and is primarily concerned with domestic safeguards requirements such as those specified by the US Department of Energy (reference 5).

### **PURPOSE OF SAFEGUARDS**

By tradition, NM safeguards is comprised of three systems: NM accounting, NM control, and physical protection. Two of these systems, NM accounting and NM control, are the components of the MC&A system. The purpose of NM safeguards is three fold:

- prevention, deterrence, and/or detection of theft of special nuclear material (SNM);
- identifying anomalies involving SNM and flagging them for investigation; and
- resolving questions of potential unauthorized activities involving SNM

The safeguards system is most effective in meeting its goals when the three components are integrated allowing sharing of data to maximize the capability to detect unauthorized activities that might indicate an attempt to steal SNM. Additional effectiveness may be achieved by including data from non-safeguards sources.

### **SAFEGUARDS ROLE OF MC&A**

The primary role of MC&A in meeting safeguards goals is to detect and assess anomalies involving nuclear materials. Anomalies are defined as any irregularity or inconsistency in the data. Indications of anomalies include:

- items not in their recorded locations;
- items with missing or wrong tamper-indicating devices;
- inconsistencies in measurement data;
- unusual quantities reported as processing losses, inventory differences, or shipper/receiver differences; and
- data on NM transfers and other activities not accompanied by appropriate authorizing paper work.

Anomalies may result from inadvertent errors that occur in any routine operations, or from deliberate performance of unauthorized actions. A system of consistency checks and reviews of the MC&A data are required to detect anomalies. All anomalies must be assessed to determine whether they may have resulted from attempt to steal SNM. Important MC&A functions for detecting anomalies include daily administrative checks and physical inventories.

### **DATA MANAGEMENT CONCEPTS**

The basic approaches to using data to meet safeguards requirements and goals can be summarized as follows.

- The traditional and most common approach is to focus on the NM accounting data. This data is most readily available, and is normally maintained in a computerized form.
- An expanded approach would be to add other safeguards data. Data from the NM control and physical protection systems provide information on accessing NM and can increase the ability to detect and assess anomalies.
- In a fully integrated approach, the safeguards data is supplemented by information from non-safeguards sources within the facility. Many functions involving nuclear materials are

not recorded in the safeguards systems records, but could provide valuable information on the status of NM as well as cross checks for accuracy of the safeguards data.

## **SOURCES OF DATA**

As noted above, the data that can be utilized to meet safeguards goals can be categorized into three types: NM accounting data, other safeguards data, and data from non-safeguards sources.

### ***NM Accounting Data***

The primary sources of NM accounting data result from the following activities:

- external receipts and shipments;
- internal transfers;
- periodic and special physical inventories;
- measurement of item and bulk material quantities;
- physical and chemical changes during processing; and
- reported inventory adjustments (operating losses, shipper/receiver differences, and inventory differences).

This data is normally provided directly to or generated by the NM accounting system.

### ***Other Safeguards Data***

The other two parts of the safeguards system, NM control and physical protection, have data that is of value in detecting anomalies.

NM control data sources include:

- NM transfer controls;
- NM and data access controls;
- tamper-indicating devices (seals); and
- bar coding.

Some NM control data may be included on the NM accounting system computer, maintained on a separate computer, or available only in paper records.

Physical protection data sources include:

- NM surveillance devices;
- alarms; and
- written records (operational logs).

Physical protection data may be maintained on a separate computer or in paper records; however, it is normally not included on the accountability system computer.

### ***Non-Safeguards Data***

Many activities outside of the safeguards function can provide data for consistency checks of safeguards data and/or new data to assist in anomaly detection. Three such functions are NM processing, health and safety, and NM management. Processing data of use to safeguards include processing measurements of SNM quantities, tank volumes and bulk material weights, in-process transfer activities, and physical measurements (temperature, pressure, density). Useful data from Health and Safety includes nuclear criticality control records, radiation levels, and records of safety alarms (e.g., air monitor alarms. The nuclear material management records include data on NM quantities, form, etc., and information control of the use of the NM for the intended purpose. There may be other examples of non-safeguards data that can be used to increase the effectiveness of the safeguards system.

## **DATA MANAGEMENT SYSTEM REQUIREMENTS**

In order to allow the use of all available data, the safeguards data management system must provide for exchange of information. Communications must be established among the three elements of the safeguards system, and between safeguards and non-safeguards organizations. The nature of the communications established between the various sources of data are likely to differ depending upon how the data is maintained. The NM accounting system is required to be computerized for most nuclear facilities. It normally includes on-line input of some accounting data and direct access to other accounting data. Access to other sources of data may be direct or indirect. Data from NM control sources are often included in the computerized accounting system (e.g., TID records and bar code data). When possible, the MC&A system should also be able to access other NM control data by direct connection. The data from the physical protection system is not normally included in the computerized accounting system, but may be accessed directly provided the data is computerized. If direct communication is not possible, indirect transfers (disk or electronic files) should be available. The data available from non-safeguards activities is not always computerized, and may be available only by transfers of paper work. More direct access is desirable but may not be feasible.

The concept of a single computer system to collect and manage data from all safeguards and non-safeguards sources is attractive from the point of view of efficiency. However, this may not be a desirable situation due to considerations of separation of duties, "need to know" requirements for sensitive information, and the fact that supervision of the various sources rests with different organizations. The preferred approach for an integrated system is separate computerized systems with direct inter-system communications. Where direct communication is not feasible, care should be taken to implement an effective indirect communication system. Regardless of the level of communication between the various data management systems, it is essential that a system of consistency checks be implemented to assure the reliability of the data used for safeguards purposes.

## **CONCLUSION**

The preferred configuration for integrated data management does not necessarily require a single computer system; however, separate computerized systems with direct inter-system connections is preferred. Use of all relevant data (NM accounting, NM control, physical protection, and non-safeguards) is necessary to assure the most effective protection for the NM inventories. Where direct exchange of data is not possible, a systematic program to implement indirect exchange is essential.

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## METHODS FOR NUCLEAR MATERIAL CONTROL USED IN THE BASIC PRODUCTION OF A TYPICAL RADIOCHEMICAL PLANT

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Techniques for destructive and non-destructive assay of the component and isotopic composition of nuclear materials are described, namely, gravimetric, titrimetric, coulometric, mass-spectrometry, as well as those based on the registration of neutron and  $\gamma$ -radiations. Their metrologic characteristics are described. The techniques described are suggested to be used for MC&A purposes at the model radiochemical plant for processing irradiated fuel subassemblies from power reactors. The measurement control program is also described. This program is intended for the measurement quality assurance in the framework of MC&A system.

### INTRODUCTION

This paper is mainly based on the materials of work performed Contract # B802819 between SSC-VNIINM and Brookhaven National Laboratory. The work completed in 1997, was dedicated to the development of requirements to measurements of bulk fissile materials (FM) for MC&A as applied to the model radiochemical plant.

The basic measurement techniques, designed in the Analytical Division of SSC RF - VNIINM and introduced into practice after the commissioning of the first domestic radiochemical plant RT-1, were considered and described in the course of developing the requirements to the methods of FM measurements. The FM measurement techniques are enumerated and briefly described. In the paper Consideration is given not only to their physical and chemical principles, but also to the sequence of analytical operations implemented in the technique.

The techniques are described in relation to the particular materials analyzed in the corresponding material balance areas with the indication of the accuracy achieved, along with its comparison with the recommended international target values and with the conclusion about the level of consistency between the achieved values of accuracy and international criteria.

In those cases where the analysis techniques do not agree with the recommended international target values in terms of the accuracy criteria, the reasons for these conditions are indicated and recommendations on their improvement are given.

### 1. DESTRUCTIVE ASSAY TECHNIQUES

#### *1.1 Isotopic composition analysis by a mass-spectrometric technique*

The mass-spectrometric technique is used in order:

- to determine the content of uranium, plutonium and neptunium in the starting nitrate solutions;

- to determine uranium and plutonium isotopic composition in the end products (uranium and plutonium dioxide fusion cake).

The isotopic composition of uranium and plutonium (both metal compounds) is analyzed by means of a mass-spectrometric technique with the use of thermoionization mass-spectrometers MI-1201 commercially available (resolution is not less than 500) and MI-3306 (resolution is equal to 800). The resolution is determined as a ratio  $M/\Delta M$ , measured at the level of 10% of the peak height.

The metal Pu samples to be analyzed are dissolved in nitric acid; the plutonium dioxide samples are dissolved in the mixture of concentrated nitric acid and hydrofluoric acid or hydrochloric and iodhydric acids. After that the sample solutions are transformed into the nitrate form by means of evaporation with concentrated nitric acid. The plutonium solution in 0,1 M  $\text{HNO}_3$  is applied onto the ion source. 1-3 mg uranium or plutonium are applied onto the ion source evaporator.

The U,  $\text{UO}_2$ ,  $\text{U}_3\text{O}_8$  samples are dissolved in nitric acid, evaporated up to wet salts and dissolved in 0,1  $\text{HNO}_3$ .

For the isotopic analysis the plutonium samples are first of all subject to purification from americium by means of a chromatographic technique. The presence of other actinides does not impact the results of a mass-spectrometric isotopic analysis of standard plutonium.

The isotopic sensitivity of the analysis is equal to  $5 \times 10^{-4}$  and  $2 \times 10^{-7}$  at.% for MI-1201 and MI-3306, respectively (detection limits). The result error depends on the type of the instrument and on the isotopic content and covers the range of 0.008-0.02% (MI-1201) and 0.005-0,01% (MI-3306).

The metrological support for this technique is based on the system of state and sector reference samples of uranium isotopic composition which cover the entire range of isotopic content required.

The capability of mass-spectrometric methods for isotopic analyses is also used in the isotopic dilution technique in order to determine the uranium and plutonium content in various materials and solutions. In case of isotopic dilution a certain amount of the element under determination is introduced into the sample. The isotopic composition of this element differs from the composition in the sample ("tag"). the variation in the isotopic composition is measured by mass-spectrometer. After that the content of the element is calculated. The following isotopes are used as "tracers": U-233, U-235, U-238, Pu-242 (in case of high enrichment in these isotopes).

A relative error of the results obtained by means of isotopic dilution ranges from 1% to 1.5%.

## 1.2 Potentiometric titration technique

The technique is used in order to determine uranium:

- in the starting nitrate solutions in the products of the first and second cycles of extraction;
- in the end product in the form of uranium fusion cake.

### 1.2.1 Uranium potentiometric titration: the Davies-Grey method

Uranium (VI) is reduced to uranium (IV) with iron (II). The excess of iron (II) is oxidized with nitric acid in the presence of Mo (VI) as a catalizer. Uranium (IV) is titrated with potassium bichromate ( $\text{K}_2\text{Cr}_2\text{O}_7$ ) in the presence of vanadyl sulfate as an indicator. For this purpose a Russia lab titrator T-108 with ionometer EV-74, is used. A platinum electrode EPV-1 (TU 25-05.2143, Russia) is used as an indicator and a mercury/mercurous sulfate electrode ( $\text{Hg}/\text{H}_2\text{SO}_4$ ) or a silver-chloride (EVL-1MI or EVL-1M3, Russia) are used as a reference electrode. One these reference electrodes used in the redox titration method have the advantage over calomel electrodes in a more distinct fixation of the titration end point.



5 ml 1,5 M/L sulfaminic acid solution and 40 ml 85% phosphoric acid solution are added to the sample solution which has about 300 mg uranium. The solution is stirred after the addition of each reagent. Then 5 ml 1M/L iron (II) sulfate solution is added and the sample is being held up for 0.5 - 1 min. The vessel walls are washed with 10 ml oxidizing mixture (the mixture consists of 8 M/L nitric acid, 0.15 M/L sulfaminic acid and 0.4% ammonium molybdate) and the solution is stirred vigorously avoiding any splashes. After the dark-brown colour is vanished (20-40 s) the solution is being held up for 3 min and then 100 ml water and 110-130 mg vanadyl sulfate are added and right after that titration starts. For this purpose electrodes are installed into the sample vessel and the stirrer and ionometer are switched on. The ionometer covers the range of "-1+4" in case of the mercury-mercurous sulfate electrode and the range of "4+9" in case of the silver-chloride electrode. Then at a high speed the dispensing burette doses about 90% equivalent volume of titrant. After that the burette is switched over to a low speed and external control. The "START" key is pressed and the process of titration starts. It will last till the preset potential is achieved (70-160 mV with regard to the  $\text{Hg}/\text{H}_2\text{SO}_4$  electrode and 550-600 mV with regard to the silver-chloride electrode). As soon as the "end" lamp flashes the readings of equivalent volume in the burette display are registered in the log. The time span between the moment when the sample is ready and the moment when the titration is over should not exceed 7 min. During titration it is necessary to register the titrant solution temperature, which is needed for the calculation of uranium mass fraction in the sample being analyzed. A relative error of this method is equal to 0.3%. Plutonium does not affect the measurement.

#### *1.2.2 Plutonium potentiometric titration: by the McDonald-Savage method*

The method can be used in order to determine Pu:

- in nitrate feed solutions,
- in products of the first and second extraction cycles.

Plutonium is oxidized to the VI-valent state with cerium nitrate. The excess of cerium nitrate is reduced with sodium arsenite in the presence of osmium tetroxide as a catalizer. The excess of sodium arsenite is decomposed with potassium permanganate. The excess of potassium permanganate is reduced with oxalic acid. The final stages of Pu determination consist in Pu (VI) reduction to Pu (IV) with a standard solution of Fe (II), a small excess of which is determined by the back titration with a standard solution of potassium bichromate. The titration end point is determined potentiometrically.

In order to determine plutonium we use a redox titration by the Davies-Grey method. However we think that in the future it will be possible to use the titration by the McDonald and Savage method which is described here based on the information from the USA. No data on the relative error of this measurement are available.

#### *1.3 Coulometry in determination of uranium and plutonium*

This method is used in order to determine U and Pu:

- in nitrate feed solutions
- in products of the first and second extraction cycles
- in the end product in the form of plutonium dioxide and uranium fusion cake.

The controlled - potential coulometry (CPC) makes it possible to make precision analyses without any preextraction of the components and can achieve a high selectivity. The titration end point (TEP) is determined directly from the current value without any special indication system.

One of the methods within the framework of CPC technique is a potential - scanning coulometry (PSC). In this case the valency transfer potential is gradually varied starting with the

initial valency potential. By means of that it is possible to significantly reduce background currents and thus to increase sensitivity and accuracy of measurements.

The analytical control is carried out step-by-step in the following way:

- The amount of sample measured very accurately is introduced into the electrolytic cell and diluted with a background electrolyte. The preliminary reduction of uranium and plutonium is carried out with a gradual decrease in the potential from +0.5V to -0.17V. This value is maintained to the residual current which does not exceed 1 mA. Then uranium and plutonium are jointly oxidized with a gradual increase in the potential up to +0.90V. This value is maintained till the residual current is equal to 10 mA.
- Then plutonium is selectively reduced with a gradual decrease in the potential to +0.5V, with the residual current of 10 mA.
- Plutonium is selectively oxidized with a gradual increase in the potential up to +0.90V, the residual current being equal to 10 mA.

The duration of each step and current integrals which passed through the cell during this stages are measured. The data obtained are used with the aim to calculate the analysis results with the help of certain constants, parameters and results of out-of-process measurements. A relative error of this method is 0.3%.

The specific feature of this method is actually absolute automation of electrochemical analysis and mathematical processing of the obtained values, their filing and possible application in the computer nuclear material control and accounting system.

#### **1.4 Direct U and Pu measurements by injection-spectrophotometry**

This method is used in order to measure U and Pu

- in the products of the first and second extraction cycles.

The method consists in injecting the sample solution into the carrier flow (nitric acid, 3M) and measuring the U and Pu inherent light absorption at various wave lengths, in particular at 416 nm and 563 nm, respectively.

The device used for this purpose consists of separate components of a liquid chromatograph "TSVET-306" made in Russia:

- vessels for a carrier solution,
- a pump to supply the carrier solution with the required speed into the analytical line (BPZh-49 type),
- a sample injector which consists in a 6-way gage tap with a dose metering loop (30 ml) and two positions: "sampling" and "analysis",
- a spectrophotometer SF-00 (Russia) which contains a monochromator with a spherical mirror ( $F=160$  mm) and a diffractive lattice (1200 pieces/mm) with a flow-through vial of 15 mm in length and 25 ml in volume. It serves in order to get the electrical signal proportional to the value of luminous radiation transmission,
- a logarithmic amplifier (UL-02 type) to transform signals from the spectrophotometer in order to get the output signals proportional to the optical density of the solutions being analyzed or to the concentration of metals under measurement within the interval of 0.01-1.28 units.

The device provides the level of zero signal fluctuation noise not higher than 0,005 mV in the dry cell and the detection limit of benzene of  $5 \cdot 10^{-7}$  g/ml.

Analysis. The carrier (nitric acid 3M solution) goes through the device with the volumetric velocity of 5 ml/min and with the "sampling"cock position of the tap. Through the metering loop the carrier is supplied to the analytical line and the loop is washed and filled with the solution

being analyzed. Then the tap is switched over to the "analysis" position and the sample is forced out from the loop to the flow-through vial of the spectrophotometer by the carrier flow. The solution optical density is registered first at the wave length of 416 nm and then, at the second sample injection, of 563 nm. The time required for changing the wave length and setting the base line does not exceed 5 min.

The uranium and plutonium concentrations in the solutions being analyzed are determined by means of the graduation curve and calibration factor obtained on the basis of standard solutions with the definite content of these metals in them. The relative error of this method does not exceed 1%.

The specific features of this method consist in its very rapid determination, no need in sample preparation small volume of samples (several tens of microliters), easy automation,.

### **1.5 Extraction chromatography/spectrophotometry techniques**

The method is used to measure uranium and plutonium in raffinates and liquid waste.

The aqueous and organic solutions of uranium, neptunium and plutonium have a specific colour, thus making it possible to determine these elements and to identify their various valency forms.

Spectrophotometric methods of direct determination of uranium, neptunium and plutonium are mainly used in order to solve particular analytical tasks due to low sensitivity and selectivity of these methods.

Spectrophotometric methods of U, Np and Pu determination based on the formation of intensively stained complex of these elements with organic reagents have assumed a significant importance for analytical purposes. Arsenazo III and xylenol orange are used most frequently as these reagents. As the arsenazo group agents form the stained compounds with many elements in the degrees of oxidation +4 and +6, it is necessary to preliminary pre-extract the element being analyzed from the solution in order to purify it from different hindering components and impurities. In order to selectively extract and purify U, Np and Pu from the solutions and materials being analyzed the chromatographic methods are often used, in particular, the method of extraction chromatography with "solid-extractants". Solid extractants are in the form of granulated porous polymers impregnated with the extrantant. The extrantant mass content in the solid extractant can achieve high values, exceeding 50%.

The spectrophotometric determination of uranium (III) with arsenazo (III) is usually carried out at pH = 5.5. For this purpose the solution aliquot (about 5 ml) which contains 5-50 mg of uranium is placed into a measuring flask (25 ml). Then 15 ml of acetate buffer with pH 5.5 and with sodium ethylenediaminetetraacetate (trilone B) are added. After that 1 ml saturated sulfanilic acid solution and 2 ml 0.1% arsenazo (III) aqueous solution are added. The tag is reached by means of certain addition of distilled water. Then the solution is stirred and left for 15-20 min. After that the optical density is measured with the spectrophotometer at the wave length of 650 nm in the vials with the length of 50 mm based on the comparison with the arsenazo (III) solution. The uranium content is determined with a graduation curve.

The method sensitivity is equal to 1 µg. The relative error of this method is 10%.

Uranium in the organic solutions is reextracted with of acetate buffer solution which contains arsenazo (III). Then the optical density of the product is measured of two wave lengths: 750 nm and 650 nm, based on the comparison with the reextrantant solution. The relative error is equal to 5%.

### 1.6 Gravimetry for uranium measurement

This method is used in order to determine uranium in uranyl fusion cake.

This method is based on the following: the sample is dissolved in nitric acid, uranium is precipitated at  $\text{pH} = 2.25\text{--}2.50$  in the form of uranyl ethylenediaminetetraacetate which in 10-15 min after addition of hydrogen peroxide is transformed into uranium peroxide. The following introduction of formate buffer solution provides the optimum pH and a quantitative precipitation of uranium within the next 15-20 min. Uranium determination is performed by the calcination of the precipitate and its weighing in the form of  $\text{U}_3\text{O}_8$ .

A certain amount of fusion cake (uranyl nitrite hexahydrate) which contains about 0.5 g uranium is weighted with the error not more than 0.0005 g, put into the can, then 10-20 ml 1 M nitric acid and 20-40 ml distilled water are added and the solution is heated up to boiling. The solution is diluted with distilled water up to 100 ml and then 5 ml solution of diammonium salt of ethylenediaminetetraacetic acid, 0.5 ml thymol blue solution and ammonia solution (1:6) are added till the time when the colour changes from pink to orange. In this case uranyl ethylenediaminetetraacetate of a light yellow colour is precipitated. Then 15-20 ml of hydrogen peroxide are added. After that the solution above the precipitate should become pink. In 10-15 min 15 ml of buffer solution are added and in another 15 min the solution is filtered through the two-layer paper filter "white strip". The precipitate is washed by means of decantation. The precipitate is transferred onto the filter and washed with 50-100 ml washing solution. The washed precipitate together with the filter dried out. The dried filter placed in the preliminary weighed crucible is installed into the muffle furnace with the temperature not higher than  $300^\circ\text{C}$ . Then the temperature increases up to  $900\text{--}1000^\circ\text{C}$  and is maintained at this level for an hour. Then  $\text{U}_3\text{O}_8$  formed in the result of all these procedures is cooled in the exsiccator and weighed. In parallel with this analysis a control test is performed for each stage of the analysis. The relative error of the method is 0.3%.

### 1.7 Measurement of $^{237}\text{Np}$ concentration in solutions with the methods of extraction chromatography, isotopic dilution and gamma-spectrometry

The method is used with the aim to determine neptunium concentration in the solutions of Pu product.

It is rather difficult to determine Np concentration in solutions which have a complex salt composition and contain uranium, plutonium and fission products, without its pre-extraction and purification from impeding impurities. The analysis which involves chemical procedures usually assumes either actually complete extraction of the nuclide to be analyzed (and that is a rather labour-consuming and complicated task), or controlling its chemical yield by the method of isotopic dilution. The isotopic dilution technique permits exclude the errors in measurement results which occur due to the analyzed nuclide losses in the purification process. Below is the description of the technique for measuring  $^{237}\text{Np}$  concentration in solutions which is based on the combination of methods of extraction chromatography, isotopic dilution and  $^{237}\text{Np}$  and  $^{239}\text{Np}$  - gamma-radiation spectrometry by means of a semiconductor gamma-spectrometer. Trioctylamine is used as a extrantant. This extrantant allows to achieve a high level of purification of Np extracted from the solution from U, Pu and gamm-emitting fission products.  $^{239}\text{N}$  in its radioactive equilibrium with the mother  $^{243}\text{Am}$  is used as a tracer.

The analysis is performed in the following way. The aliquot with the volume of 1-2 ml is taken from the solution being analyzed. Then 2 ml  $^{243}\text{Am}$  solution is added to this aliquot and carefully stirred. Next comes the addition of 0.1 ml Fe (II) sulfamate solution. The final solu-

tion is left for 2-3 min and then transferred into the chromatographic column and filtered with the speed of 1 ml/min. When this procedure is over the column is washed with three portions of 3 ml 1.5 M/L nitric acid each. Neptunium extracted by trioctylamine is eluted with 2 ml 0.3 M/L  $H_2C_2O_4$  solution in a special vial made of organic glass.

The  $^{237}Np$  concentration in the eluate is determined by gamma-spectrometry. By means of the gamma-spectrometer of high resolution  $^{237}Np$  gamma-radiation with the energy of 86.5 keV and  $^{239}Np$  gamma-and/or X-radiation with the energy of 228.2 or 277.6 keV or (106.1 + 103.7) keV are registered. maximum  $^{239}Np$  yield is gamma-radiation with the energy of (106.1 + 103.7) keV. But it is necessary to take into account the contribution of  $^{237}Np$  gamma-radiation with the energy of 106,1 keV.

The  $^{237}N$  content in the solution being analyzed is calculated by means of the following formula:

$$m_{237} = \frac{G_{237}}{G_{239}} \cdot \frac{N_{237}}{N_{239} \cdot e^{\frac{\ln 2}{2,347} t}} \cdot m_{243},$$

where  $G_{237}$ ,  $G_{239}$  are graduation factors which are determined during graduation based on the results of multiple measurements;

$N_{237}$  - a number of pulses in the  $^{237}N$  peak at the energy of 86,5 keV during the time of measurement;

$N_{239}$  - a number of pulses in the  $^{239}Np$  peak during the time of measurement;

2,347 is a  $^{239}Np$  half-life, days;

t - the time period since the beginning of extraction till the beginning of measurement, days

$m_{243}$  is the  $^{243}Am$  mass added as a tracer into the solution being analyzed, mg.

Graduation is carried out by means of reference solutions with the known concentration of  $^{237}Np$  being in the equilibrium with  $^{233}Pa$  and reference solutions of  $^{243}Am$  in the equilibrium with  $^{239}Np$ . The solutions are placed into the vials identical to those which are used for the analysis and the measurements are performed in the same geometry. The graduation factors are calculated by means of the formulae:

$$G_{237} = \frac{m_{237}^k}{N_{237}^k \cdot 0,87} ; \quad G_{239} = \frac{m_{243}^k}{N_{239}^k},$$

where  $m_{237}^k$ ,  $m_{243}^k$  are  $^{237}Np$  and  $^{243}Am$  masses, respectively, in the reference solutions, mg;

$N_{237}^k$ ,  $N_{239}^k$  are numbers of pulses in  $^{237}N$  and  $^{239}Np$  peaks, respectively, during the time of measurement;

0,87 is a coefficient which takes into account the contribution of  $^{233}Pa$  gamma-radiation into the  $^{237}Np$  peak.

The relative error of this technique within the range of  $^{237}Np$  content in the sample being equal to 1 - 200 mg is from 30% to 6% at the confidence probability of 0,95.

There is another available technique similar to the one described above with the only difference which consists in the fact that  $^{237}Np$  is used as a tracer.

## 2. NDA METHODS

### 2.1 X-ray fluorescence technique

This method is used in order:

- to measure U, Pu, Np concentrations in the feed solution;
- to measure U, Pu, Np concentrations in the extrantant products.

The X-ray fluorescence method can be applied both for the feed analysis and the analysis of U, Pu and Np products.

The method consists in excitation of heavy element atoms by X-ray tube irradiation or with an isotopic source and in registration of characteristic radiation of K, L or M series.

The most commonly accepted excitation source is a X-ray radiation generator through isotopic sources are currently used with increasing frequency. The detection systems available are both with the wave length dispersion and with energy dispersion. Detection is mainly carried out in the L-area.

In case of the wave length dispersion a high resolution is provided, but at the same time it is accompanied with a low detection efficiency. And vice versa, the energy dispersion option provides a better efficiency but worse resolution. The spectrometer with the wave length dispersion within the range of U/Pu ratio from 50 to 300 provides the relative standard deviation of about 0,5% in the result of measurement results.

The spectrometer with the wave length dispersion and with X-ray tube excitation was designed and used in operation for a certain period of time at the reprocessing plant.

The X-ray fluorescent method can be recommended to be used for the analysis of both feed solutions and the product of the first extraction cycle and U, Pu and Np products.

### 2.2 Gamma absorption method for controlling U and Pu solution concentrations

This method is used in order:

- to control U and Pu concentration in the process solutions.

In order to nondestructively control U and Pu concentration in process solutions of radiochemical fuel-element fabrication plants the method of gamma absorptiometry is widely used. It is based on measuring the of external source radiation attenuation by a solution layer depending on the uranium concentration in it. The advantage of this method consists in the fact that it is not sensitive to the isotopic composition of the material being measured. There is no serial production of gamma absorptiometers as devices for assaying U and Pu concentrations. They are usually designed as applied to the requirements of each specific task.

$^{241}\text{Am}$  is used as a gamma-radiation source in the absorptiometers applied for the process control. Scintillation counters with a thin scintillator are usually used as detectors. The solution layer thickness between the source and detector is chosen depending on the range of uranium concentrations measured in the test point and is a fixed value in the gamma absorptiometer design.

For the conventional one-beam absorptiometer the dependence of U or Pu concentration ( $C_{NM}$ ) in the solution on the number of registered pulses is described by the formula:

$$C_{NM} = \frac{\ln(N_0 / N)}{\rho x (\mu_{NM} - \mu_s)} - \frac{\mu_s}{\mu_{NM} - \mu_s},$$

where  $N_0$  is the number of registered pulses without any source;  
 $N$  is the number of registered pulses with the source available;  
 $r$  is the solution density,  $\text{g}\cdot\text{cm}^{-3}$ ;

$x$  is the solution layer thickness, cm;

$m_{NM}$  is the mass factor of the source gamma-radiation absorption by U or Pu,  $\text{cm}^2 \cdot \text{g}^{-1}$ ;

$m_s$  is the mass factor of the source gamma-radiation absorption by the carrier,  $\text{cm}^2 \cdot \text{g}^{-1}$ .

The dependence  $C_{NM} = f(N)$  is well-defined if  $r$  and  $m_s$  are constant. In practice it may not be so and thus it requires a thorough analysis of corresponding errors.

### **2.3 Neutron methods of control**

The techniques based on detection of inherent neutron radiation of nuclear materials makes it possible to measure process product characteristics directly related to the fissile material mass. These characteristics are as follows: mass, concentration or distribution of fissile material in the particular equipment. Some of these techniques can be used with the aim to measure technological process characteristics related to the fissile material amount in the equipment. As compared to other NDA methods these techniques are distinct in the relative simplicity of methodological solutions and equipment being used, thus allowing a high reliability of their running to be achieved. The feasibility to implement this method under the conditions of high radiation fields directly in the process lines makes this method be the most important and widely used tool for remote control of technological process at radiochemical and chemical-metallurgical plants. The physical basis of this method consists in measuring neutron radiation of process products. Under the particular geometrical conditions and with the reproducible nuclide composition this radiation value can be related to the NM mass or concentration. Transuranium nuclides are the principal source of neutron radiation for the process products at radiochemical plants. In the feed solution of VVER spent fuel or spent fuel subassemblies this is curium, and in case of low burn-up these are plutonium isotopes. After solution purification from fission products and transplutonium nuclides the principal neutron source is plutonium. In the course of uranium affinage the uranium neutron radiation can be used for the purpose of control (due to  $^{238}\text{U}$  spontaneous fissioning). Neutron counters and fission chambers are used as neutron detectors.

The following techniques are used or can be used for purposed of control and accountancy of bulk nuclear materials:

- measurement of NM mass (concentration) in tanks;
- measurement of the amount of unsolved fuel on cladding;
- measurement of NM distribution in the extraction equipment;
- measurement of NM mass in the sorption equipment and monitoring of process dynamics;
- monitoring of spent fuel dissolution dynamics.

Besides, the neutron radiometric methods are used to monitor Pu content in solid radioactive waste; though for this purpose evidently it will be more reasonable to use methods based on neutron coincidence or active methods with the application of a neutron generator or isotopic source or their combination.

In some cases neutron radiation measurements are accompanied with the measurement of gamma-radiation. For instance, the amount of unsolved fuel on cladding can be accounted based on neutron and gamma-radiation. Gamma radiation can be also converted into neutron radiation by means of a beryllium radiator.

### **2.4 Measurement of Pu content with gamma-spectrometry and neutron coincidence technique**

The method is used in order:

- to analyze the end product in the form of plutonium dioxide,
- to determine Pu concentration in waste.

The principal NDA methods currently in use for measuring Pu mass in various Pu bearing materials are neutron coincidence counting and calorimetry; their accuracy is quite competitive with destructive methods. However these methods require certain data on isotopic composition of Pu being analyzed. These data can be obtained by non-destructive gamma-spectrometric methods with the use of semi-conductor gamma-spectrometers of high resolution with codes such as MGA, PC/FRAM, etc. for gamma-spectra processing.

The passive neutron coincidence method is based on the selective registration of neutrons generated in one event of spontaneous fission of Pu even isotopes nuclei ( $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{242}\text{Pu}$ ). These prompt neutrons are correlated in time, whereas ( $\bar{\alpha}$ , n)-reaction neutrons form the background randomly distributed in time. The required selectivity is achieved by means of registration of neutron pulse coincidences within the time interval which depends on the physical properties of the detection system.

Methods and tools for NM assay based on double neutron coincidences have been widely used, since Development and application in the Los Alamos National Laboratory of shift register package and the mechanism of semi-empirical correction of neutron multiplication in samples measured. Neutron coincidence counters based on the shift register have been demonstrating a very successful operation for the MC&A purposes for about 20 years.

The principal contribution into the neutron spontaneous fission counting comes from  $^{240}\text{Pu}$  both in case of Pu from the fuel with a low burn-up value, where the content of this isotope covers about 6%, and in case of Pu from the fuel with a high burn-up value, when its content runs to ~ 15% -25%. For the sake of convenience the value called  $^{240}\text{Pu}$  effective mass ( $^{240}\text{Pu}_{\text{eff}}$ ) is used.  $^{240}\text{Pu}_{\text{eff}}$  mass is the mass of  $^{240}\text{Pu}$  which gives the same coincidence count as a real sample with the above-mentioned even isotopes. If the isotopic composition of Pu in the sample to be analyzed is known or measured than one can calculate the  $^{240}\text{Pu}_{\text{eff}}$  value as a fraction of the total Pu mass using the formula:

$$f(^{240}\text{Pu}_{\text{eff}}) = f(^{240}\text{Pu}) + 2.52 f(^{238}\text{Pu}) + 1.68 f(^{242}\text{Pu}),$$

where  $f(^i\text{Pu})$  is mass (or atomic) fraction of Pu even isotopes which can be obtained in the result of gamma-spectrometry measurements. The coefficients 2.52 and 1.68 take into account the difference in the numbers of specific neutron production in spontaneous fission neutrons per a unit of mass of  $^{238}\text{Pu}$  and  $^{242}\text{Pu}$  relative to  $^{240}\text{Pu}$ . The value  $f(^{240}\text{Pu}_{\text{eff}})$  is given by the codes MGA and PC/FRAM as a result of measurements.

Having measured the spontaneous fission neutron count rate in the sample being analyzed ( $n_{sf}$ ) we can get the value of  $^{240}\text{Pu}_{\text{eff}}$  mass in grams via the formula:

$$m_{\text{Pu}} = m_{^{240}\text{Pu}_{\text{eff}}} = K \cdot n_{sf},$$

where K is a graduation factor in g·s, which is determined during of neutron coincidence counter calibration by measuring a count rate of spontaneous fission neutrons from Pu standard sources with the known isotopic composition (the known values of  $m_{^{240}\text{Pu}_{\text{eff}}}$  in grams).

The total Pu mass in the sample being analyzed in grams is determined with data on neutron coincidence counts and gamma-spectrometry by means of the formula:

$$m_{\text{Pu}} = m_{^{240}\text{Pu}_{\text{eff}}} / f(^{240}\text{Pu}_{\text{eff}}).$$

However, a significant drawback of such a combined method is that the gamma-spectrometry technique is capable to determine the relative content of all the Pu isotopes with the exception of  $^{242}\text{Pu}$  which cannot be distinguished in the spectrum due to a very low intensity of its gamma-radiation. Because of that the content of this isotope is calculated based on the correlation with



other isotopes. A lot of different investigations resulted in a great number of algorithms for determining  $^{242}\text{Pu}$  content. However each of them taken separately is applicable for a rather limited group of Pu samples depending on the reactor type, burn-up value, initial enrichment in terms of  $^{235}\text{U}$  and other factors. In the MGA code the correlation proposed by R. Gunnink and applied in many codes is used in the following form:

$$^{242}\text{Pu} = K (^{240}\text{Pu}/^{239}\text{Pu}) (^{241}\text{Pu}/^{239}\text{Pu}).$$

The disadvantage of this correlation is its dependence on the relatively short-lived  $^{241}\text{Pu}$  whose content reduces with the rate of about 5% per year. So in the MGA code another polynomial correlation formula is used, less sensitive to  $^{241}\text{Pu}$ :

$$[242] = a + b[239] + c[240] + d[241] + e[239]^2 + f[240]^2 + g[241]^2,$$

where the numbers in brackets mean the measured values of isotopic contents and the coefficients from a to g for four ranges of  $^{239}\text{Pu}$  contents are experimentally determined and tabulated.

The error in  $^{242}\text{Pu}$  determination by means of correlations can be significant (more than 10%, they are indicated in the MGA print-out. This fact impacts the results for other isotopes (1%) and total Pu mass (up to 2%).

Double coincidence counters measure only first two moments of counting whereas there are at least three unknown parameters:  $^{240}\text{Pu}$  effective mass, multiplication factor and the ratio of ( $\alpha$ , n) reaction neutrons to spontaneous fission acts. The principal solution of this problem consists in measuring the moments of the highest order (at least, the third one). A significant progress has been achieved both in developing theoretical models and in designing multiplicity counters.

In order to analyze plutonium dioxide in the end product the combined method can be used. It is based on the use of gamma-spectrometry to analyze the isotopic composition of Pu and a multiplicity counter designed in the Los-Alamos National Lab with the aim to measure the mass. It will make it possible to decrease the effect of humidity and impurities on the results of measurements.

The well diameter and depth in the counter developed in the LANL for the scrap analysis is 20 cm and 41 cm, respectively. The multiplicity counter consists of a detector unit with a high voltage supply and preamplifiers located in its upper part, an electronic package and a computer. The counter well with two graphite end plugs is surrounded with 80-130  $^3\text{He}$  - tubes installed into a polyethylene moderator. The neutron registration efficiency is equal to 40-55%. For the plutonium dioxide sample with the mass of 1 kg and with insignificant amount of impurities the relative error of Pu mass measurement will be 0.3 - 2% with the confidence probability of 0,68 and with the time of measurement being equal to 1000 seconds.

The Pu content in waste is assumed to be determined by the combined method based on the use of gamma-spectrometry in order to analyze the Pu isotopic composition and the use of passive neutron coincidence counter in order to determine Pu mass. The modern commercial passive neutron coincidence counters allow to measure from 0.3 g up to several kilograms of Pu both in large and small containers. The relative error of measurements is 1%-5%.

### 3. MEASUREMENT CONTROL PROGRAM

The objective of the measurement control program consists in assessment of the efficiency of the measurement system being used and planned to be used, in quality assurance of measured values being used for the material accountancy and in getting the values of reproducibility and accuracy to be used in evaluation of inventory difference limits as well as error limits for evaluation of shipper-receiver data.

**The measurement control program** envisages the following parts:

**Program of electronic scales and balances.** All types of scales and balances being used for accountancy must be maintained in a good condition, be calibrated according to the time fixed and their accuracy, and scale linearity must be checked every day.

**Analytical quality assurance.** The routine measurement data must be statistically analyzed in order to provide accuracy and reproducibility of measurements.

**Sampling conditions.** The error related to the sampling uncertainties must be determined and set at the regular basis.

**Physical measurement control.** Reproducibility and accuracy in measuring volume, temperature, pressure, density, etc. must be definitely set and guaranteed.

**Instrument calibration.** All the instruments must be calibrated with the use of corresponding standards or at least the measurement values must be compared with more accurate results of measurement.

**Standard samples.** All calibration and working standards used in the measurement control program must meet be traceable with the national measurement system. In this case the standard sample errors must be lower than the errors of analytical methods in which they are used. Working standards must be representative with respect to the type and composition of the material under measurement when the NM matrix impacts measurement results.

**Sample exchange program.** A radiochemical plant likewise any other plant must participate in the programs of sample exchange with other plants or labs in order to provide independent verification of the internal quality assurance of analytical measurements.

**Statistical control.** For each measurement technique meant for the NM accountancy certain procedures for correcting measurement conditions must be determined and documented for the cases when the preset error limits are exceeded. These limits should be set at the level of two standard deviations (a warning signal) and three standard deviations (an alarm signal).

In the first case when the warning limit is exceeded the method must be corrected and the reasons for that must be revealed. In the second case when the alarm limit is exceeded the measurement method must be thoroughly analyzed, the reasons must be revealed and eliminated. After that the method should be demonstrated as workable within the limits of preset statistical control.

**Measurement technique verification.** The program of measurement technique verification must be available at any plant. The objective is to demonstrate applicability of these techniques before taking measurements for the purpose of accountancy. Both for DA and NDA methods this demonstration should be performed at least once a day before starting real measurements. When it is impossible or unreasonable, the check measurement is carried out after each five measurements.

**Check measurement procedures.** These procedures must be stated in the written form. The correct implementation of check measurement procedures is provided with a corresponding program.

Currently the measurement verification program is being realized at the radiochemical plant according to the sector regulatory documents (OST95.10430-90, RD95.10396-89, RD95.10398-89 and OST 95.10289-87). On the whole these documents correspond to the **Measurement Verification Program** with the exception of the list of required standard samples.

Quantitative and Qualitative Measurements in NMC&A at RCP

**TABLE 1. Summary data on MC&A methods at reprocessing plant**

Material being analyzed	Method	$\Delta^*,\%$	Target values, %	
			Random	Systematic
Fuel Assemblies	Gamma spectrometry and neutron count	5.0	—	—
Feed solution of uranium, plutonium, neptunium	Coulometry	0.3	—	—
	Potentiometric titration	0.3	0.15	0.15
	Mass-spectrometry with isotopic dilution	0.5-0.7	0.2	0.2
	X-ray fluorescence	0.7	—	—
Uranium and plutonium waste	Coulometry	0.3	—	—
	Neutron count	3-5	—	—
Uranium and plutonium solutions	Coulometry	0.3	0.15	0.15
	Redox titration	0.3	0.15	0.15
	Mass-spectrometry with isotopic dilution	0.5-0.7	0.2	0.2
First cycle products: uranium, plutonium, neptunium	Coulometry	0.3	0.15	0.15
	Injection Spectrophotometry			
	Gamma-Absorptiometry	1.0	—	—
	X-ray fluorescence	1.0	—	—
		0.5	0.5	0.5
Liquid waste	Potentiostatic Voltammetry	0.5	—	—
	Extraction Chromatography/spectrophotometry	0.5-1.0	—	—
	Mass-spectrometry with isotopic dilution			
		0.5-1.0	0.2	0.2
Plutonium and Neptunium solutions	Coulometry	0.3	0.15	0.15
	Injection spectro photometry	0.5	—	—
	Gamma absorptiometry			
		1.0	—	—
Plutonium dioxide and neptunium powders	Coulometry	0.3	0.15	0.15
	Gamma spectrometry with neutron coincidence counting	1.0	—	—
Liquid waste	Potentiostatic voltammetry	0.5	—	—
	Extraction chromatography which ends in spectrophotometry	0.5-1.0	—	—
	Gamma spectrometry with isotopic dilution			
		1.0	—	—
Solid waste	Neutron counting	5	—	—
Uranyl Nitrite fusion cake	Weighinig	0.3	0.05	0.05
	Coulometry	0.3	0.15	0.15

$\Delta^*$  - Relative error at the probability of 0.68.



## METROLOGY AND EXTERNAL QUALITY CONTROL IN REPROCESSING PLANTS

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### Introduction

The large amounts of nuclear material processed in modern reprocessing plants has the consequence that measurements of fissile material for fissile material control need to be carried out with the lowest possible uncertainties. At the input or dissolver stage, the concentrations and isotopic abundances of uranium and plutonium have to be measured, and these amounts are then compared with the purified uranium and plutonium, including amounts in waste streams. Metrology has an important role to play in ensuring the reliability of the analyses and therefore reducing the uncertainty on the fissile material balance.

### Type of analyses employed in Safeguards

Today, the majority of measurements made for fissile material control can be classed as non-destructive. Techniques employed include gamma-spectrometry, neutron counting and K-edge absorptiometry. These techniques have a number of practical advantages: they can be relatively rapidly carried out, therefore producing results in a short time; they are capable of producing results with very good precisions; and they produce waste of a type which can be recycled back to the plant.

Destructive analysis techniques, properly carried out, are capable of producing the highest quality results (i.e. those with the smallest uncertainty on the result,  $u_c$ ). Indeed, the techniques commonly applied in this field, gravimetry, titration, coulometry, isotope-dilution mass-spectrometry, are all classed as "primary methods of analysis", which signifies that they meet the CCQM criteria:<sup>1</sup>

*"A method whose operation can be completely described and understood, for which an uncertainty statement can be written down in terms of SI units, and whose results are therefore, accepted without reference to a standard of the quantity being measured".*

The disadvantages of destructive analysis methods, lie in the relatively long time needed for analysis, the need for highly skilled analysts and in the waste produced during the chemical manipulations.

### Safeguards measurements and Use of Reference Materials

Destructive analysis methods are considered the basis of fission material control measurement techniques, for the reason that they can be made traceable.

Safeguards fission material control is based on the principle of the comparison of independent measurements. That is, the operator declarations are checked by samples being independently measured by a safeguards laboratory. The laboratory is put under pressure to produce good results for comparison within a short time, which is the reason non-destructive analysis

methods are used for the most part. However, comparability of measurement results depends on each measurement being traceable to a common base. This can only be done by means of Reference Materials: there is no such thing as a completely unbiased measurement!

Reference materials are of 2 types: Isotope Reference Materials and Amount Reference Materials.

One form of an Amount Reference Material: an element assay RM is used to quantify small biases, or systematic deviations arising for example in titration or coulometric titration, where the extent of completion of the determining reaction will typically be slightly less than 100%. This small factor is determined by measuring an element assay Reference Material under the same conditions. Thus the daily and local variations or biases in the measurement results can be corrected for.

However, where the isotope abundances are required which is the case for uranium and plutonium mass-spectrometric techniques have to be used. A mass-spectrometric measurement suffers also from biases, of which the most important is mass-fractionation. This bias arises from mass-specific processes during the measurement: evaporation in the source being the largest effect. This mass-fractionation has to be measured for the measurement conditions employed and this is done by measuring an Isotopic Reference Material under the same conditions that were used in measuring the samples. A well set-up mass-spectrometer should have the same mass-fractionation factor under given conditions, irrespective of the magnitude of the isotopic ratios measured. An isotopic reference material, as a first and very good approximation, does not therefore need to match the isotopic abundances of the sample exactly: if this is deemed necessary, it is a sign that the measurement is not under control.

When isotope amounts are to be determined, a spike an Isotopic Amount Reference Material, as well as an isotopic reference material. The isotope dilution equation is in principle simple and most importantly allows the uncertainties involved in each term to be written down and combined to yield the uncertainty of the isotope amount measured. Clearly, for this measurement process, there are uncertainty contributions from the fractionation factor and from the uncertainty of the spike, as well as uncertainties from the measurement itself. Nevertheless, all these uncertainties can be quantified and their values held low.

As a consequence: the measurements will be in the last effect, limited by the uncertainties on the isotope reference materials and spikes. Such recognition is pushing production of Reference Materials to even better defined materials.

The role of a metrology laboratory in this process is to provide Reference Materials which meet the requirements of the measuring laboratories. In particular, the Reference Material must be certified with a total combined uncertainty ( $u_c$ ) and this uncertainty should be considerably less than that of other measurements (when properly budgeted)<sup>2</sup>. This must be complete, since the measurement laboratory will use the certified value and its uncertainty to calculate the value of the isotope abundance being measured, together with the uncertainty on this value. The measurement results are then Traceable to the SI.

### **SI-Traceability in Safeguards**

In the competition between operator laboratory and safeguards laboratory to produce the best results, the advantage of having SI-traceable values is considerable. Bluntly expressed, the results from a laboratory which can prove its measurements to be SI traceable, cannot be contested, except for mundane causes such as, for example, sample confusion.

In a local situation, traceability can be, and often is, local. That is, if both measuring laboratories base their results on the same local reference material, (not SI-traceable) the results will be comparable, even if this RM itself has a considerable bias. If the measurements are used only in this local situation, there will be no consequences, but the modern world is becoming

smaller, and measurements made, especially in the nuclear field, are frequently compared to results from other parts of the world. The need to have Reference Materials traceable to basic physical constants is thus clear. Such Reference materials are called Primary Reference Materials (PRM) and are defined by the CCQM:<sup>2</sup>

*"A Primary Reference Material is an RM established and certified by means of a primary method of measurement".*

### Quality Control in Safeguards

Having defined how laboratories measure, and how they should use RMs, what type of control is possible for the users of the data to ensure that the laboratory reaches the levels of results required?

All laboratories have internal quality control, to ensure the continuity (reproducibility) of measurements from one analysis to the next. Large establishments and national organisations will also organise QC campaigns to ensure

1. the required uncertainty level of measurement results is maintained,
2. to compare internally the performance of laboratories,
3. to bring out biases, in the form of systematic differences between the laboratories.

Using mean values in around robin process only allows conclusions to be made which apply internally between the laboratories taking part. However, to provide an absolute basis for QC comparison campaigns, the use of certified test samples (CTS) with SI-traceable values has many advantages:

- the reference value is absolute and therefore unbiased,
- the total uncertainty is given for the CTS. This makes comparison of measured results with the certified value more straightforward.
- Allows world-wide comparison of measurement results

The materials are called Certified Test Samples to keep clear the difference with regard to RMs: a CTS is a RM whose values are undisclosed, except when the measurements have been carried out and submitted by the analytical laboratory.

The disadvantages of using a RM for QC lie in the difficulties of producing and certifying the materials and guaranteeing longevity and homogeneity. This applies in particular to the nuclear fuel cycle, where solutions (for instance dissolved fuel, or high concentrations of Pu in solution) can often be unstable with time.

### Conclusions

Measurements on fissile material carried out in reprocessing plants, for material control or safeguards purposes have high requirements of accuracy because of the high degree of control needed for the material flow. To these requirements, we can add "traceability", as without this comparisons of measurements results cannot be made.

The role of reference materials is to provide the traceability needed for comparison of the measurement results and therefore they - and the metrology needed to produce and certify them - are essential, not only on reprocessing plants, but also in the entire nuclear fuel industry.

Measurement results need also to be assured, and this is done by means of external QC campaigns using certified test samples

### References

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<sup>2</sup> Guide to the Expression of Uncertainties in Measurements: ISO, IEC, OIML, BIPM, ed by ISO, 1992.



## MIXING AND SAMPLING TESTS FOR RADIOCHEMICAL PLANT

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### Introduction

Among the more difficult sources of measurement uncertainty to assess are mixing and sampling. Because there are no standards, sampling studies and/or tests are required to assess sampling systems statistical contributions to overall measurement uncertainty. Over the years in the U.S., there have been a number of approaches to such tests. This paper describes results and test procedures used to evaluate uncertainty and bias effects introduced by the sampler systems of a radiochemical plant, and similar parameters associated with mixing. In addition, these tests are also the basis to establish operational parameters to assure minimum effects of sampling and mixing on overall measurement uncertainties.

Design of a radiochemical plant and the processing equipment is always a compromise between competing requirements and disciplines. Safeguards must compete with operational and criticality concerns. Safeguards must compromise in these arenas yet achieve acceptable measurement statistics in light of other concerns. The tests and evaluations described can guarantee acceptable measurement performance within constraints of other concerns.

This report will concentrate on experiences at the Barnwell Nuclear Fuels Plant (BNFP). In the history of reprocessing in the U.S., initial experiences were in the weapons plants at Hanford and Savannah River built in the 40's and 50's. The first commercial reprocessing plant built in the U.S., which deviated from the canyon concepts at Hanford and Savannah River, was at West Valley. This plant operated from 1966-72 and set the groundwork for advanced reprocessing concepts in the U.S. Some experiences were gained at the Idaho Chemical Processing Plant (ICPP) as they planned for upgrades through the 1970's. However, the BNFP plant incorporated the latest technologies and experiences from the U.S. reprocessing histories. It was on a par with the THORP plant currently operating in the U.K. with respect to equipment design and the efforts to qualify measurement equipment and technologies. This paper will concentrate on experiences at the BNFP and discuss related experiences in the earlier generation plants as well.

### Tank/Equipment Design

As mentioned, a number of concerns must be mediated in the final design of plant equipment, and thus measurement systems. When it comes to the overall vessel configuration, several variations are available to the designers. The simplest is the simple cylindrical tank, oriented with the straight-walled sections horizontal or vertical. The issue here is criticality. Concentra-

tions of fissile material must be limited in these tanks or soluble poisons must be added. Typically the tanks are used in the head end area of a light water reactor processing facility, in chemical or waste handling areas, or in uranium finishing areas where the isotopic and overall elemental concentrations are limited and controlled. Internal structures for heating and cooling as well as mixing and sampling can complicate issues related to measuring, but the sampling and mixing issues are peculiar to the cylindrical design.

Sometimes vessel configuration becomes more complex such as in the case of evaporators and concentrators. These are special cases for the sampling and mixing questions. Generally, they pose the concerns when addressing the requirements during inventory taking to close material balances during conventional shutdown inventories or on-line inventories to support near-real-time accounting or other timely loss detection techniques.

In areas of a plant where high fissile material concentrations are expected, such as plutonium purification and conversion or highly enriched uranium processing, other tank designs are required for criticality control. Annular or slab tanks are typically used in these applications. These designs pose a separate set of questions related to sampling and mixing. Another option in tank design under criticality constraints is a tall, small diameter tank, often referred to as "pencil tank". It again presents a unique challenge to operations related to mixing and sampling. Designers have resorted to other configurations for criticality control. One design uses runs of critically safe pipe slightly off the horizontal and connected at the ends by vertical runs of critically safe pipe. These look like a harp and are often referred to as "harp tanks". In a few cases, critically safe geometry pipe has been connected in a "D" shape to control criticality and facilitate mixing/re-circulation. All of these options present challenges to the safeguard measurement technologist.

The question of capacity also relates to design and considerations for mixing and sampling. Throughput or temporary storage requirements dictate capacity. Physical space requirements also influence design. In specific areas of processing plants. As examples, slab tanks or pencil tanks may be interconnected to form a tank system. The safeguard measurement specialist must consider these options.

Most of this discussion related to liquid processing considerations. An entirely new dimension is added in sections of processing plants where solids or slurries are handled. This paper will concentrate on liquid processing measurement problems.

In all designs, the issue is to guarantee a homogeneous solution in the process vessel to allow a representative sample to be drawn. This requires a method for mixing/agitation. In some applications mechanical means may be applied such as motor driven paddles. Various forms of pumps can be used to re-circulate solutions. Both of these methods are limited to applications where maintenance on the pumps or motors can be carried out. There are some air drive pump techniques that can be used for highly radioactive solution measurement points, but remote/no-maintenance methods are usually required. These are usually air-driven techniques to stimulate agitation/mixing, such as internal air-lift mixers, or air spargers. Spargers may take the form of submerged rings with a series of air holes drilled where the air bubbles exiting under pressure create turbulence to simple tubes where the bubbles create turbulence and stimulate re-circulation by convection.

Sampling is another matter. Where radiation exposure and contamination concerns can be addressed by gloveboxes or ventilated hoods, samples may be drawn from simple gravity fed devices such as valves/spigots. There are sampling accuracy concerns here, but these are rather simply addressed by pulling enough samples to "flush" the valve. In radio-chemical plants the majority of samples must be remotely drawn. In the U.S., the most widely used sampling tech-



nology is the vacuum assisted airlift, re-circulating sampler. This is a rather elegant device that uses an airlift in a sampling location to deliver solutions part way to a sampling location, usually in a hot cell remote from the processing operations. The actual sampling station consists of a sampling block with two needles. The airlift pipe to the tank/solution terminates at one needle. The other needle connects to a vacuum supply and drains back to the tank/process. The airlift design is not sufficient to deliver solution all the way to the needle. A sample bottle with a rubber diaphragm cap is pushed onto the two needles. Installation of the bottle closes the system between the airlift and the vacuum and the vacuum assist pulls the solution the rest of the way up the supply line to create circulation of the solution through the bottle back to the tank/process. The safeguards questions here relate to the minimum re-circulation times to guarantee a "representative sample from the solution, and to quantify potential concentration effects from the vacuum/airlift.

In the current safeguards regime, all system must be qualified to establish statistical parameters associated with the contribution to measurement uncertainty and establish operational parameters guarantee performance and to minimize the statistical effects. The test method to establish these parameters is the subject of the remainder of this paper.

### **The Role of Density Measurements in Tests and Operations**

If there is one message to be conveyed in this paper it is the importance of process solution density measurements. It can be argued that this is the single most important analytical measurement that can be made in radio-chemical plants. The state-of-the-art technique for solution density measurement is the vibrating tube method. This method relates the harmonics of a vibrating glass capillary tube with process solution to the tube with air and de-mineralized water. Such equipment is manufactured and distributed by Paar-Mettler as one source. The Paar-Mettler equipment is capable of density measurement precision in the fourth or fifth decimal place, depending on the model. Accuracy is dependent on calibration and accuracy related to process measurement must consider temperature effects.

The implication of temperature effects is that a totally homogeneous solution will change in density with temperature, but according to predictable relationships. The accuracy of commercially available equipment can be guaranteed by control of temperature. A controlled water bath for samples, coupled to modern vibrating tube density measurement equipment can ensure accuracy to the fourth or fifth decimal place. The implication here is that solutions measured in the controlled temperature measurement application can be compared at the fourth or fifth decimal place. Again, results at the fourth or fifth decimal place level depend on the model equipment purchased, but decisions on comparisons of solution can be made at the hundredth of a percent or better.

### **Mixing Tests**

The objective of mixing tests to establish the effectiveness of the installed mixing systems and the operating parameters to guarantee performance of the systems during routine operations. As an example, a particular process tank is equipped with a sparge ring mixing system. How long must that mixer be operated to guarantee a "homogeneous" solution and to what degree is that solution "homogeneous".

Some qualitative tests were run over the years at several test facilities. For example, the Idaho plant built a plexi-glass slab tank to study mixing. They added acids and acid sensitive dyes to the tank. They initiated mixing and looked for color based indications of homogeneity and local-

ized dead spots for mixing. These were generally tests with slab tanks and sparge tubes and they were able to optimize placement of the tubes for mixing and elimination of dead spots. However the dye methods did not necessarily quantify mixing parameters.

The questions were answered at the Barnwell Nuclear Fuels plant using a mixing test involving density measurements. The tank in question was partially filled with an acid of one concentration. Acid of a different concentration, and consequently a significantly different density was carefully added to create a "stratified" condition in the vessel. At this point the mixing system was started and a sample drawn immediately. Subsequent samples were drawn every five minutes. All samples were analyzed for density using the vibrating tube density measurement equipment with the constant temperature water bath. Sequential density results were considered. When sequential samples agreed within the Precision/accuracy of the density measurement, it was concluded homogeneity was attained. When subsequent samples (two or three) agreed within the statistical parameters, it was assumed the homogeneity was within those limits of the density method. For the Barnwell tests, agreement to within 0.0008 was the criteria with solutions of density in the neighborhood of 1.2-1.3mg/ml. This corresponds to an "accuracy" in mixing on the order of 0.07%. This was with density measurement equipment capable of accuracy in the fourth decimal place. Modern equipment is capable of measurements to the fifth decimal place, with corresponding improvements in assessment of mixing capabilities.

These tests were run on a number of process vessels at Barnwell. Mixing time to achieve accuracy of less than 0.1% were in the neighborhood of 15-30 minutes for most single tank systems. A number of tests were run on systems consisting of several connected slab storage tanks. These tanks are mixed by re-circulating pump systems. In this test, a single tank was filled with acid of considerably different density from the others. Mixing was initiated and sampling commenced in the same way as for single tanks. Initial tests showed minimum mixing times of several hours to achieve homogeneity to the accuracy of the density method. Re-circulation pump capacities were increased and re-tests were conducted. Satisfactory mixing (within the accuracy of the density method) could then be achieved within 30-60 minutes. But the testing method using density as the parameter allowed for this judgement and corrective action.

### Sampling Tests

Sampling parameters for simple sampling systems can be established based on calculations, once the homogeneity of the solution in the vessel is established. For instance, a spigot sampler draws liquid from a tank. Mixing will not generally affect the solution trapped in the spigot itself, but will guarantee the homogeneity in the vessel. The sampling procedure must simply guarantee a flush of the full capacity of the spigot to guarantee the "homogeneous" solution from the tank is delivered.

The problem becomes more complex with more complex samplers. For example, consider the re-circulating airlift sampler system. In this case the solution must enter the sampler system, flush the lines, flush the sample bottle and consider any systematic effects such as concentration of the sample by the flow of air involved.

Again, going back to the Barnwell experience in the U.S., a test was developed to quantify sampler system effects. Test equipment was fabricated to back flush the samplers on the air-lift side with water. At the conclusion of the mixing experiments, when the density of the homogeneous acid solutions in the tanks had been established, the sampler systems were back-flushed with water. Re-circulation in the sampler was by installing a bottle on the needle block. Sample bottles were exchanged on the block every 3-5 minutes. This provided a "new" sample of the

delivered solution with each bottle. All bottles were analyzed for density. Again, when subsequent bottles agreed within the accuracy of the density method, it was concluded that the sampler was delivering the homogeneous solution from the tank.

The minimum re-circulation times were established by the test. Based on the test results, it was concluded that with minimum re-circulation times, the sampler system could deliver samples within a "precision" equal to the "accuracy" of the density measurement. The question of "systematic" uncertainties remained. To answer this question, during the pre-operational tests at Barnwell, grab samples from the tanks were taken and analyzed for density at the conclusion of the sample tests. Comparison of the density of the sampler delivered samples to the grab samples was made. It was concluded in these tests that sampler system systematic errors were less than the precision of the density method (i.e., less than 0.07% with the equipment used in those tests). Tests for systematic effects in operating facilities where "grab" samples are not attainable would be problematic and rely on in-tank density measurement comparisons.

### **Conclusions**

Mixing and sampling tests can be conducted to establish the statistical parameters for those activities related to overall measurement uncertainties. Density measurements by state-of-the-art, commercially available equipment is the key to conducting those tests. Experience in the U.S. suggests the statistical contribution of mixing and sampling can be controlled to less than 0.01% and with new equipment and new tests in operating facilities might be controllable to better accuracy.



## **NONDESTRUCTIVE ASSAY MEASUREMENTS APPLIED TO REPROCESSING PLANTS**

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### **Abstract**

Nuclear materials accounting and verification in reprocessing plants is essential, because it is the first time in the nuclear fuel cycle that plutonium can be measured. With the main characteristics of reprocessing plants being large flows and holdup of nuclear materials, inaccessibility of the material in a major part of the plant, complex and automated processes, materials accounting and verification are difficult challenges in these plants. Effective nuclear materials accounting systems and international safeguards inspections in reprocessing plants rely heavily upon nondestructive assay measurements of the plant's nuclear materials in both item and bulk form. Radiation-based nondestructive assay (NDA) techniques play an important role in providing timely accounting data on the full range of process materials and rapid verification of previous measurements. NDA for reprocessing plants relies on passive gamma-ray spectrometry for plutonium isotopics and plutonium mass values of medium-to-low-density samples and holdup deposits; on active x-ray fluorescence and densitometry techniques for uranium and plutonium concentrations in solutions; on calorimetry for plutonium mass in product; and passive neutron techniques for plutonium mass in spent fuel, product, and waste. This paper will describe the radiation-based nondestructive assay techniques used to perform materials accounting measurements. The paper will also discuss NDA measurements used in inspections of reprocessing plants.

### **I. INTRODUCTION**

Reprocessing plants are very significant from a safeguards point of view, because they separate fissile materials, which could be used in a short period of time for nuclear weapons with a minimum of additional work. Even small plants are capable of producing many kilograms of plutonium per year. Thus, nuclear materials accounting and control (MC&A) in reprocessing plants is very important, but reprocessing plants also represent the largest and most complex challenge to safeguards.

Today, a commercial reprocessing plant can process up to 1000t of fuel per year, and produce nominally 7t of plutonium. Therefore, it is very important that nuclear materials control and accounting in a reprocessing plant provide accurate assessments of the facility's inventory of nuclear materials, in all forms and locations within the plant. Nondestructive techniques are an integral part of these assessments and are used frequently by safeguards inspectors and facility operators.

Spent fuel assemblies arriving at the plant are identified and stored temporarily under water in a storage pond. The safeguards at the storage pond are no different than those at the storage pond of a reactor. The nuclear material is in identifiable items, but quantities are known only approximately by reactor calculations that can only be verified by indirect measurements.

The next step is to dismantle the assemblies and chop the fuel rods at the head end of the reprocessing plant. At this point, the item integrity of the fissile material is lost. The chopped material is dissolved into a liquid phase for separation of the fissile materials. This is the only opportunity for determining the amount of plutonium input to the separation process by analysis of the dissolved fuel in the accountability tank.

A major difficulty arises from the fact that in the separation or chemical processing area of reprocessing plant, the nuclear material is inaccessible. Because the plant is working with highly irradiated material fuel, the early separation stages must be carried out behind shielding, which is normally thick, concrete walls. Thus, another important measurement is at the output accountability tank. To establish a material balance for a separation campaign, representative samples are taken of input and output, and all streams leaving a plant. This requires that many different measurement techniques be available to provide complete materials accounting data.

The output measurements for the process area are at the same time the input measurements to the storage area. The measurement and loading of oxide powders at the packaging station serve to define the identity of the cans and thus establish the basis for item accountancy in the product storage area. The loading of the oxides into the cans therefore constitute a transition from bulk accountancy to item accountancy. All transfers of product canisters into and out of the storage area are subject to item counting, item identification and, as appropriate, NDA testing.

## **II. NONDESTRUCTIVE ASSAY (NDA) TECHNIQUES**

Nondestructive measurements are usually separated into two types, qualitative or attribute measurements, which indicate that the irradiated fuel contains the correct radioactive signatures; and quantitative measurements, which relate to the specific characteristics of the irradiated material, burnup or fissile content, and separated materials, quantity and isotopic abundances.

While weighing is by far the most precise nondestructive measurement of nuclear materials, it requires knowledge of its chemical and physical composition, which is usually determined by destructive analysis on samples of the material. This becomes difficult, however, when the matrix materials, chemical composition, or the impurities vary in the materials and cannot be determined accurately by destructive analytical measurements. Radiation-based NDA measurements is then the preferred approach and has its greatest utility when the purity and chemical makeup of the material are not well known. The following sections provide descriptions of various NDA measurements used in the common material-balance areas (MBA) of a reprocessing plant.

## A. NDA measurement at input and output of the processing area

### Input/Output Solution Analysis

Active non-destructive assay techniques such as energy-dispersive absorption edge and X-ray fluorescence spectrometry are successfully applied for quantitative measurements on the input solutions right after the dissolution of the spent fuel elements. The respective measurements involve the determination of the uranium and plutonium element concentrations on samples withdrawn from the input accountability tank. The information thus obtained may be used either for verification of the input accountability in Safeguards, or for plant control purposes.

For practical reasons the underlying measurement techniques are utilized in a variant allowing the application and exploitation of the highest possible radiation energies, viz. K-edge densitometry (KEDG) for the absorption edge measurements, and analysis of fluoresced K X-rays for the X-ray fluorescence (XRF) measurements. For both methods this means the spectrometry of radiations with energies around 100 keV. The advantages offered by this kind of radiation are:

- ability to penetrate safe sample containment (typically stainless steel);
- reduced sensitivity to matrix effects;
- sufficient element discrimination power for simultaneous uranium and plutonium assay.

The direct analysis of input solutions is accomplished with a unique NDA instrument known as 'Hybrid K-edge Densitometer' (HKED) as shown in Figure 1. The HKED combines the above mentioned techniques of KEDG and XRF to achieve a simultaneous determination of the uranium and plutonium concentration in the input solutions [1]. The instrument is normally attached to a hot cell facility as shown in the Figure to receive from there the highly radioactive input samples for analysis. Alternative installations, where the HKED is directly interfaced to pneumatic sample transfer lines, have been also realized for applications in process control.

Since undiluted input solutions typically have a total  $\beta, \gamma$  activity level of about  $10^9$  Bq per ml, a very intense external photon source is required to make KEDG and XRF measurements feasible for such type of samples. The HKED employs X-rays from an industrial X-ray generator with a

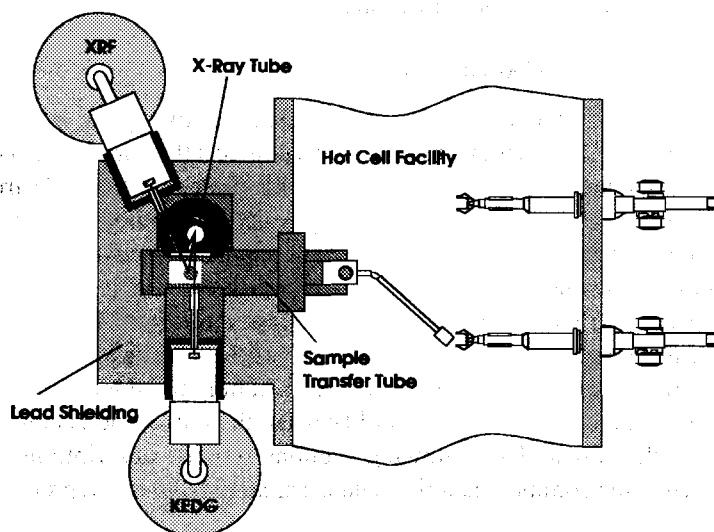


Figure 1. Layout of a typical HKED installation.

The basic components include an X-ray generator, two high-resolution germanium detectors for KEDG and XRF, and a sample transfer tube as an interface to a hot cell facility

maximum voltage rating of 160 kV for the interrogation of the samples. For optimized measurement conditions the X-ray tube should emit an X-ray continuum up to 150 keV, and it should be located at the shortest possible distance from the sample in order to ensure at the sample position a total flux of about  $10^{11}$  photons/cm<sup>2</sup>/s<sup>-1</sup> with energies above the K-absorption edges of uranium (115.6 keV) and plutonium (121.8 keV).

The technique of K-edge densitometry, which from its principle is preferably used at higher element concentration levels, has proven as a precise and extremely reliable analysis technique in its range of applicability. For precise KEDG measurements the element of interest ideally should present an areal density of at least 100-200 mg/cm<sup>2</sup> to the crossing photon beam. For reprocessing input solutions this condition is easily met by the main constituent uranium when the assayed solution thickness is chosen to about 2 cm. The resulting discontinuity at the K-absorption edge of uranium as shown in the bottom curve in Figure 2 then allows a fairly precise (about 0.2 % relative) determination of the uranium concentration at relatively short measurement times (about 1000 s). The K-edge measurements on input solutions remain practically unaffected by the presence of fission products, which on the average just lead to a predictable systematic reduction of about 0.1-0.2 % at maximum of the measured uranium concentration compared to a calibration performed with synthetic solutions free from fission products.

For concentration measurements by K-edge densitometry using disposable liquid samples one must accurately know the path length of the transmitted photon beam across the assayed sample because the directly measurable quantity, like for any type of absorptiometry measurement, is proportional to the product of concentration times solution thickness. In most of the existing HKED applications for Safeguards accountancy verification measurements the input solutions are transferred within a hot cell facility into special sample containers of well known dimensions before being transferred into the HKED for analysis. Alternatively, if the sample containers used within the plant's sampling system assure well-specified tight dimensional tolerances, it is also

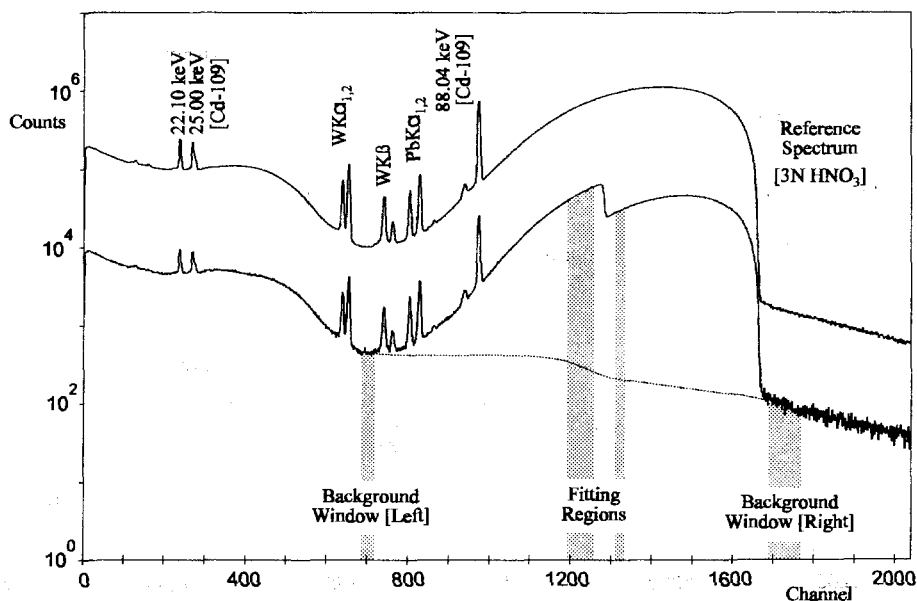


Figure 2. K-edge spectrum from an input solution showing the K-absorption edge discontinuity of uranium (bottom curve), and a reference spectrum from a blank nitric solution (top curve). The dashed spectral regions are used for the evaluation of the K-edge jump.

possible to perform truly non-destructive measurements on the original samples without the necessity of intermediate sample handling. This approach is practiced in one of the European reprocessing plants.

For the more important plutonium assay in input solutions one has to make use of the XRF measurement capability of the HKED, which offers the necessary detection sensitivity and dynamic range for simultaneous element analysis at element ratios of 100 or higher. Figure 3 displays the relevant portion of an XRF spectrum taken with the HKED from a typical input solution sample, showing the fluoresced K-X rays from uranium and plutonium. The indicated dashed spectral regions are utilized for the correct evaluation of the net peak counts in the  $K\alpha_1$  X-ray lines from uranium and plutonium. The XRF spectrum also reveals the presence of a few fission product gamma rays, which point to the fact that the XRF analysis of input solution samples may be slightly affected by the self-radiation of the samples if not properly corrected for.

From the XRF measurement the plutonium concentration in input solutions is usually determined with a precision between about 0.5-1% within a measurement time of 1000 s. The actual plutonium concentration in input solutions is normally derived from the combination of a U/Pu-ratio result from XRF with the uranium concentration result from KEDG. This measurement strategy in the HKED, which completely bases on ratio measurements alone, has consistently proved to provide very accurate and reliable assay results for uranium and plutonium in reprocessing input solutions.

In some installations the standard HKED set-up as shown in Figure 1 has been augmented by an additional gamma detector for measuring sample self radiations. Applications of the additional gamma detector include burnup verification measurements from input solutions based on the spectrometry of specific gamma rays from long-lived fission products, and plutonium isotopic composition measurements on plutonium product solutions using the analysis code MGA as described below.

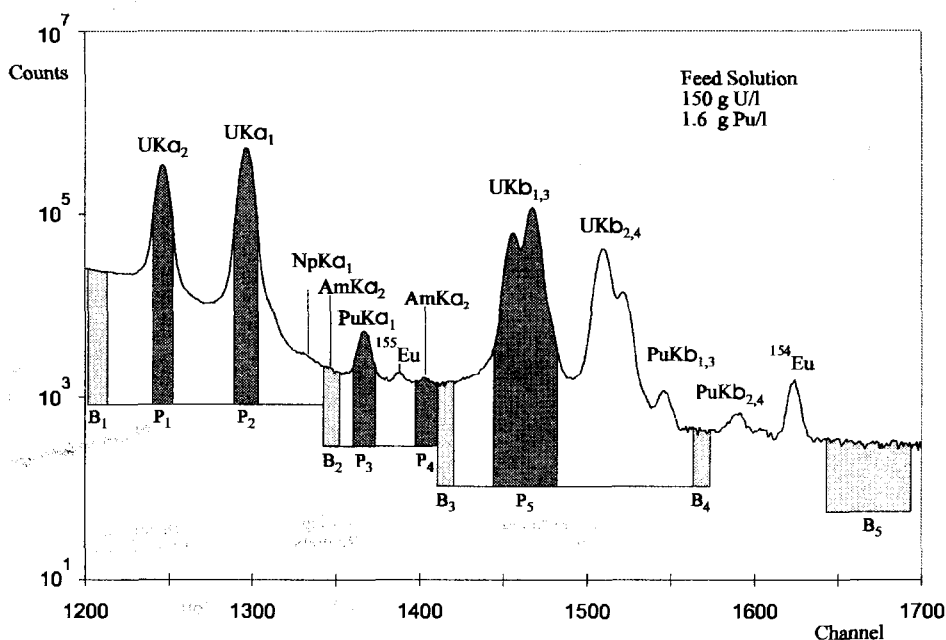


Figure 3: Sectional display of a typical XRF spectrum from an input solution sample showing the K-series X rays from uranium and plutonium. Some gamma rays from fission products are also visible. Dashed spectral regions are utilized for peak area evaluation.



The KEDG and XRF techniques incorporated in the HKED instrument offer, of course, also measurement capabilities for quantitative uranium and plutonium assay in other types of process solutions from reprocessing. In fact, any concentration level of uranium and plutonium above about 0.1 g/l in any type of process solution, from the highly radioactive input solutions down to the purified concentrated product solutions, can be quantitatively determined with the one or other technique. Dedicated KEDG/XRF instrumentation will therefore play an important role in forthcoming Safeguards on-site laboratories in reprocessing plants /2/.

HKED instruments are currently installed and used in all European reprocessing plants to perform the key Safeguards verification measurements on input solutions /3/. The most important features of this key NDA application are:

- Operational simplicity: Direct analysis of the original input samples without any prior sample treatment. Convenient instrument user interface for operation by inspectors.
- Timeliness: Measurements performed on site, providing the analysis results within 1-2 h upon sample availability.
- Independence: Application of a completely independent method of analysis versus the standard operator method (IDMS) for accountancy measurements.
- Performance: Random measurement uncertainties (0.2 % for U and 0.5 % for Pu) and systematic uncertainties (<0.2 for U and Pu) conforming to established International Target Values for measurement performance.
- Reliability: Demonstrated long-term measurement stability and reliability as a result of the adopted scheme of robust ratio measurements.
- Calibration: Use of less expensive reference solutions certified for the uranium and plutonium element concentration.
- Ease of implementation: The HKED is easily interfaced to existing plant installations for the handling of highly radioactive samples.
- Safety: Nondestructive analysis through safe sample containment as required for the highly radioactive samples.

#### **A. NDA measurements in the Storage Areas**

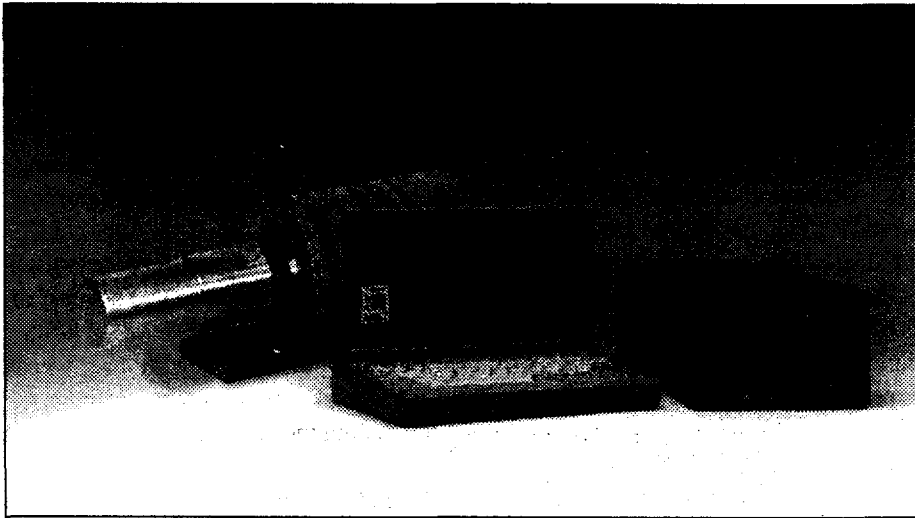
##### **1. Gamma-Ray Isotopic Measurements**

The isotopic composition of a plutonium sample is used to calculate  $^{240}\text{Pu}_{\text{eff}}$  or specific power (watts/gram), that are used to interpret passive neutron coincidence or calorimetry measurements for absolute plutonium mass. MGA can determine plutonium isotopic abundances with accuracies better than 1% using a high-resolution, low-energy, planar germanium detector and measurement times of less than 10 minutes /3,4/. Gamma-ray spectrometry systems like the one shown in Figure 4 allow inspectors to acquire spectra and analyze results in the field.

MGA can include analysis of a second spectrum of the high-energy (300-600 keV) plutonium gamma rays that significantly improves the determination of  $^{240}\text{Pu}_{\text{eff}}$  in high-burnup plutonium. MGA can also provide isotopic information from the high-energy regions only, when the low-energy gamma-rays cannot be observed due to absorption by thick sample-container walls or sample shielding.

The MGA code has the following unique analysis characteristics:

- It uses the most intense gamma-ray peaks in a plutonium spectrum; the 94- to 104-keV region and the 129- and 148-keV peaks.
- All data are represented by a single set of internally consistent and appropriately weighted equations.



*Figure 4. A portable gamma-ray spectrometry system.*

*The components include a high-resolution germanium detector (left), portable computer for data analysis (center), and data-acquisition electronics (right).*

- The relative detection efficiency for a measurement is determined from the data. The functional form used approximates the following three basic physical processes involved in the detection process:
  - the gamma-ray interactions with the detector,
  - attenuations due to absorbers (cadmium is usually used),
  - sample self-attenuation.

The advantage of basing the functional form on known processes is that its shape is determined not only by the data but also by the known physical-interaction processes.

Although MGA can determine the abundances of  $^{235}\text{U}$  and  $^{238}\text{U}$  in plutonium samples, the MGAU code performs isotopic analyses of samples containing only uranium. Like MGA, it requires no calibration, can determine  $^{235}\text{U}$  enrichments with accuracies within 1 to 2%, and can be used over the entire enrichment range from depleted to highly enriched. Also like MGA, it uses the 89-100 keV region of a spectrum in its normal operational mode. However, later versions of MGAU now also include the 'enrichment meter' approach which makes use of the 186 keV gamma ray.

MGA uses the 94- to 104-keV region of the low-energy spectrum. Although the peak structure of this region is very complicated, this region is very useful because it contains the most intense peaks of all of the plutonium isotopes and  $^{241}\text{Am}$  (except for the region below 59 keV which is visible only when analyzing freshly processed materials). This feature is particularly important when considering  $^{238}\text{Pu}$  and  $^{240}\text{Pu}$  which each emit only three low-energy gamma rays, when they decay. The emission probability of the three gamma rays diminishes greatly with increasing energy so that the 160-keV peak of  $^{240}\text{Pu}$  becomes very difficult to measure. This difficulty becomes acute when measurements are made of high burnup plutonium with high concentrations of  $^{241}\text{Pu}$ , as shown in Fig. 5. Thus the  $^{240}\text{Pu}$  peak at 104 keV provides significantly better information despite the complexity of this peak region.

The 94- to 104-keV energy region contains more than 16 peaks due to x and gamma rays emitted by the isotopes in the sample. It is impossible to determine precisely the area of each of

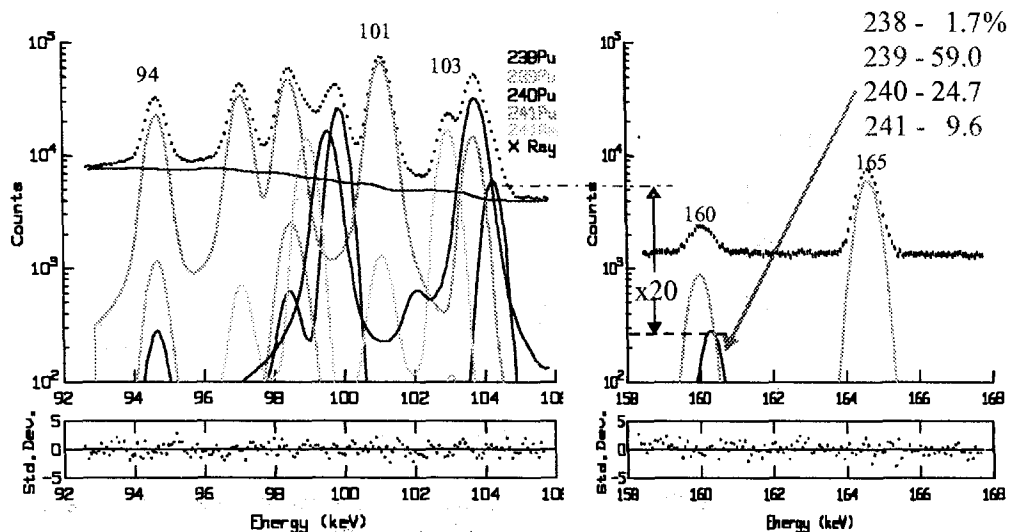


Figure 5. The right-hand figure illustrates the computed isotopic response functions used to analyze the 94-104 keV region. The intensity difference of the 104- and 160-keV peaks of  $^{240}\text{Pu}$  in high burnup Pu are also shown.

these peaks individually. However, three important characteristics of the peaks are known. First, we have developed an algorithm that accurately calculates peak shapes as a function of energy. It is important to note that the peak shape of the x rays is quite different from gamma rays, due to the Lorentzian line shape of the x rays. Second, the exact relative energies of the peaks are known, so that their relative positions can be accurately determined. Finally, the relative intensities of peaks belonging to each isotope can be easily calculated from the known emission probabilities and the relative efficiency curve.

Using these three characteristics, a 'response spectrum' can be calculated for each of the isotopic components of the 94- to 104-keV region. By doing this, not only is the number of variables reduced from over 30 down to 6 (in principle), but the remaining variables appear as linear terms in the equations, thereby allowing us to solve 170-180 equations fitting the data by a simple least-squares method. Figure 5 illustrates the characteristics of the responses to the 94- to 104-keV region.

The relative detection efficiencies for the gamma- and x-ray peak intensities are needed in the isotopic analysis. This relative efficiency curve, also called the 'intrinsic' efficiency curve, must be determined from the spectral data. Most gamma-ray analysis codes use a functional form for the efficiency curve that does not necessarily reflect the physical processes involved in detecting the gamma- and x-ray emissions. However, in MGA we use a functional form that describes the three principal interactions involved in the detection process. These are 1) the detector efficiency, 2) absorption by cadmium (or other) filters, and 3) self-attenuation by plutonium in the sample. The following equation is used to describe these processes:

$$A_j = \sum_{k=1}^{k=3} (p_{j,k} \cdot X_k) \cdot \exp(-\mu_j \cdot \text{CD}) \cdot ((1 - \exp(-\mu_j \cdot \text{PU}) / \mu_j \cdot \text{PU}) \cdot \epsilon^0 (1 - bE - cE^2)) \quad \text{Eq. 1}$$

where  $A_j$  are the areas of ten peaks in the low-energy region of each spectrum, due to  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{237}\text{U}$ , and  $^{241}\text{Am}$  and  $X_k$  are the unknown amounts of these isotopes. The absorption coefficients,

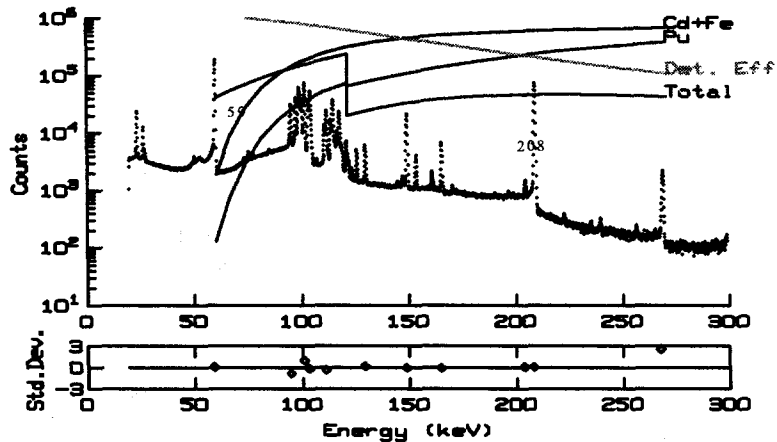


Figure 6. Plot of the three principal processes that characterize the low-energy 'intrinsic efficiency' curve.

$m_i$ , at each peak energy are known for CD and PU, but the thickness of the Cd filter and Pu sample thickness are usually treated as unknown variables. The final term in Eq. 1 describes the detector efficiency as a function of the energy,  $E$ , where the slope,  $b$ , and the curvature,  $c$ , are also usually treated as unknown variables. Equation 1 is very nonlinear in form and therefore the variables, shown in bold, must be solved by an iterative least-squares method. The characteristics of Eq. 1 are shown in Fig. 6. The resulting curve for the total efficiency is used to determine the relative efficiency values at the peaks of interest.

MGA obtains its analysis information from the low-energy regions, when they can be measured. However, improved results and additional isotopic information can be obtained from higher-energy regions (above 300 keV). Because the gamma rays in this region are considerably less intense than those at lower energy, a large coax-type germanium detector should be used. By combining the data obtained from the second higher-energy region with data from the low-energy spectrum, not only are the results improved but additional information is obtained. For example, the  $^{238}\text{U}$  abundance can be obtained from the 1001 keV peak, a more accurate analysis for  $^{237}\text{Np}$  is made, certain fission products can be detected and analyzed, and isotopic inhomogeneities in the sample can be detected. A separate intrinsic efficiency curve must be determined for the higher energy spectrum, as shown in Fig. 7. Like the low-energy curve in Fig. 6, the components of the efficiency are based on the physical processes involved in attenuating and detecting the gamma rays.

Recently a need has been identified for performing plutonium isotopic analyses of samples stored in lead-lined containers. The low-energy gamma rays are completely absorbed by the lead, rendering the low-energy spectrum useless. However, since MGA was already capable of analyzing high-energy spectra, a new mode of analysis could be implemented with only a little additional work. Therefore, latest version of MGA can now determine the abundance of plutonium isotopes of plutonium samples stored in lead-line containers by analyzing only the high-energy spectrum.

There is also current interest in using neutron coincidence methods to measure low levels of plutonium found in waste containers. However, isotopic information is needed to interpret the neutron coincidence measurements. Since high accuracy measurements are not required, the isotopic results need not be as good as for accountability measurements. However, significant

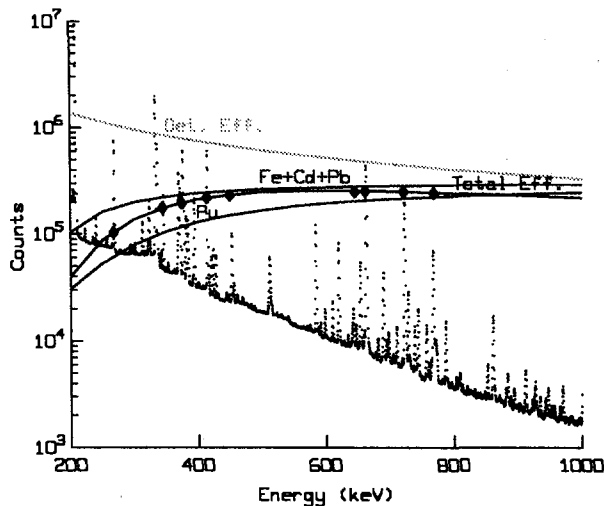


Figure 7. High energy region showing components affecting the efficiency curve.

efforts were devoted to 'harden' the code to survive statistically poor data, or exit properly so that unattended measurements may continue. Our experience indicates that plutonium abundances of about 10 mg can be measured in counting times of 10-30 minutes.

In summary, MGA is a code that analyzes plutonium gamma-ray spectra to determine relative isotopic abundances. Its most important features are:

- Requires no calibration
- Measures almost any size and type of plutonium
- Uses the most intense regions of the spectrum
- Requires only a few minutes of measurement time
- Can attain accuracies of better than 1%
- Determines the amounts, relative to Pu, of several other actinides, e.g.  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{237}\text{Np}$ , and  $^{241}\text{Am}$
- Can measure the U/Pu ratio in mixed-oxide (MOX) samples
- Can be used to measure the Pu concentrations in solutions. Only a single source is required to perform a one-time calibration
- Runs on PC computers with short data-processing times
- Requires little or no user interaction. Several convenient user interfaces have been written to control analysis parameters and select spectra for analysis. Some versions are available commercially.

The method for determining the  $^{235}\text{U}$  enrichment of a uranium sample based on gamma spectroscopy was called the 'enrichment meter' method /6/. It was based on measuring the intensity of the 186 keV peak and required a calibration, 'thick' samples, and reproducible measurement conditions. A new method was developed, and incorporated into a code called MGAU, which uses the data in the 89-120 keV region of a high-resolution germanium detector spectrum /4/. It requires no calibration, can be used on any size sample, and is capable of 1 to 2% accuracies in a few minutes counting time.

The daughter products of  $^{238}\text{U}$  emit two gamma rays in 89-100 keV region, one at 92.367 and the other at 92.792 keV. There are also Th and Pa x rays in this region due to the decay of  $^{235}\text{U}$  and its Pa daughter activity. However, since several of these peaks are very close in energy, a 'response function' approach, similar to that used in the MGA, is used to unfold the overlapping peaks in this region. When this approach is used, the isotopic response curves are easily fit by

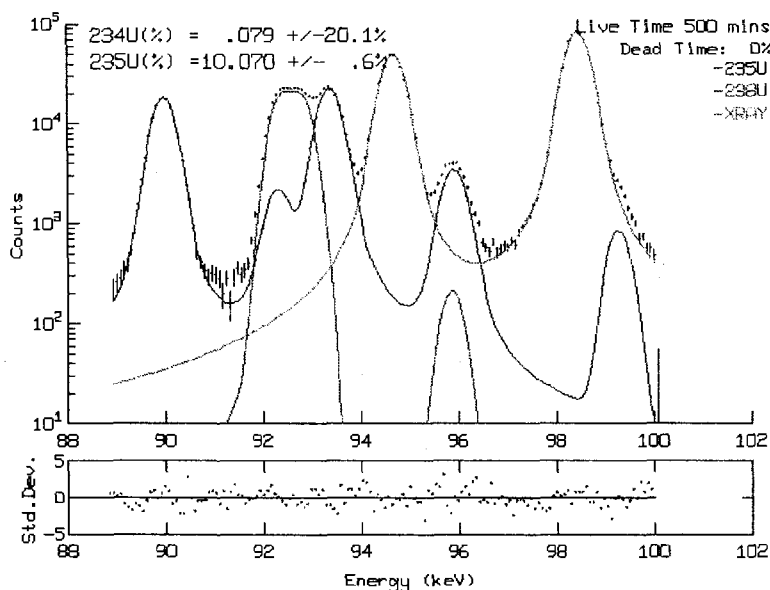


Figure 8. Isotopic response fit to the 89-100 keV region of a uranium spectrum.

the method of least-squares to determine the relative abundances of  $^{235}\text{U}$  and  $^{238}\text{U}$ . Response spectra for the 89-100 keV region of a typical spectrum is shown in Fig. 8.

Although the peaks are close in energy, an efficiency curve must be calculated for this region so that the relative intensities of the peaks in the various 'response spectra' can be determined.

Recent versions of MGAU are also capable of determining the  $^{235}\text{U}$  enrichment of a sample by the traditional 'enrichment meter' method of analysis. This method was implemented because it continues to be the only practical, nondestructive method for analyzing uranium samples that are **not** in equilibrium with their daughter products, or that are contained in thick, (highly absorbing) steel containers. However, the 'enrichment method' **does require** a detector-system calibration before analyses can be performed on unknown samples. Furthermore, the method usually can be used only when measuring samples with several mean paths of sample thickness at the 186 keV gamma ray energy.

The latest versions are also capable of analyzing samples containing thorium. Such samples produce several additional peaks, including thorium x rays that directly interfere with the peaks that are analyzed in the 89-100 keV region. These versions also account for the daughter products interferences of  $^{234}\text{U}$  when analyzing samples containing U ore.

Results we have obtained from many users and studies show that

- The method does **not** require a calibration and can be used on samples of any size.
- $^{235}\text{U}$  enrichments can be determined with accuracies of 1-2% or better.
- The entire range from depleted to 93% enrichment can be measured. However, the measurement uncertainties increase at the extremes of this range.
- The optimum accuracy is in the 3-20% range of enrichments.
- Many measurements can be made in only a few minutes of counting time.
- The method **requires**:
  - Parent-daughter activity equilibrium
  - A container wall thickness of less than about 10 mm steel
  - A high resolution Ge gamma ray spectrometer
  - That samples not contain Pu or significant amounts of other activities

MGA has had a long development history and has been subjected to many tests and used at many institutions including the IAEA and Euratom /8/. MGAU is more recent, but has also been evaluated by many groups and organizations. Nonetheless, both of these codes continue to evolve as new applications and measurement conditions arise. Fortunately, more powerful computers are available to address our demands for larger codes and for faster calculations. Special efforts are continually made to minimize the need for human interaction by incorporating decision logic into the codes.

## *2. Calorimetry Instruments*

Calorimetry involves the measurement of heat. In the case of samples typical of reprocessing plants, the heat measured is the result of the radioactive decay of the sample. By far the most common radiometric calorimeter in use today is the isothermal heat flow type. A sample chamber contains the heat producing material and is surrounded with a thermal resistance. A constant temperature heat sink is provided which may be the air surrounding the calorimeter, a temperature controlled water bath or any other sink held at a constant temperature. Heat flows from the source to the sink and is allowed to continue until a steady state is achieved. At this point, termed equilibrium, some parameter is measured that is proportional to the heat produced by the sample. The actual parameter measured is specific to the exact type of calorimeter used but is in general an electrical current, a voltage or both. Regardless of the parameter measured the final result is the same for all -- the heat generated by the sample. A detailed discussion of the various types of calorimeters can be found in reference 9.

Some of the advantages of calorimetric assay are listed below (Ref. 10 and 11):

1. The entire sample can be measured.
2. The assay is independent of sample geometry (only equilibrium time is affected).
3. The assay is independent of matrix material composition and distribution, including nominal moisture concentrations.
4. The assay is independent of nuclear material distribution within the sample, including the effects of sample self-attenuation.
5. Electric current and potential measurements are directly traceable to National Bureau of Standards (NBS) reference materials.
6. Calorimetric assay discriminates between uranium and plutonium isotopes in most cases (only plutonium is assayed).
7. Calorimetric assay is applicable to a wide range of material forms (including metals, alloys, oxides, fluorides, mixed oxides, waste, and scrap). Representative plutonium standards are not needed.
8. Calorimetric assay is comparable to chemical assay in precision and accuracy if the isotopic composition is well characterized.
9. Calorimetric assay is a completely nondestructive assay procedure when coupled with high-resolution gamma-ray spectroscopy isotopic analysis.

The last two points refer to the fact that like neutron coincidence counting, calorimetry requires an additional piece of information to calculate the plutonium mass from the measured heat output. As with neutron counting, isotopic information about the plutonium in the sample is required.

**The total mass of plutonium is calculated using the equation**

$$M = W/P_{\text{eff}} \quad \text{Eq. 2}$$

where M is the plutonium mass in grams, W is the measured heat output in watts (or milliwatts) and  $P_{\text{eff}}$  is the specific power in watts (or milliwatts) per gram of plutonium. The total specific power,  $P_{\text{eff}}$  can be calculated from

$$P_{\text{eff}} = \sum w_i P_i \quad \text{Eq. 3}$$

where  $w$  is the isotopic weight fraction,  $P$  is the specific power and  $i$  refers to each plutonium isotope and  $^{241}\text{Am}$ .  $P$  is calculated for each isotope based on the rate of decay of the isotope and the decay energy associated with the decay. Nominal values for  $P$  for each plutonium isotope and for  $^{241}\text{Am}$  are:

Isotope	Specific Power (mW/g)
$^{238}\text{Pu}$	567.57
$^{239}\text{Pu}$	1.9288
$^{240}\text{Pu}$	7.0824
$^{241}\text{Pu}$	3.412
$^{242}\text{Pu}$	0.1159
$^{241}\text{Am}$	114.2

It should be pointed out that the presence of other actinides in the reprocessed materials must be evaluated to determine the contribution to the total heat output. Uranium does not contribute significantly but isotopes of americium other than 241 may be present as well as various curium isotopes, which may cause biases in the measurement if not taken into consideration.

The list of advantages above suggests that calorimetry can be used for a variety of different material forms including waste. However in practice the primary material forms assayed are relatively pure metals and oxides. Even for these material forms, the time required for a calorimetric assay - four to twelve hours - recommends the selection of another method for routine use if greater uncertainty in the measurement can be tolerated.

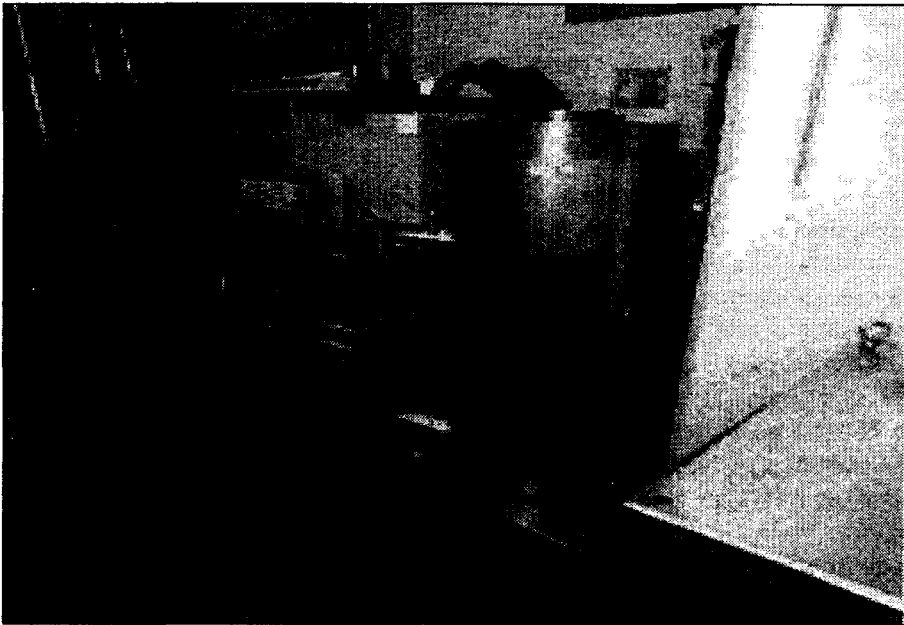
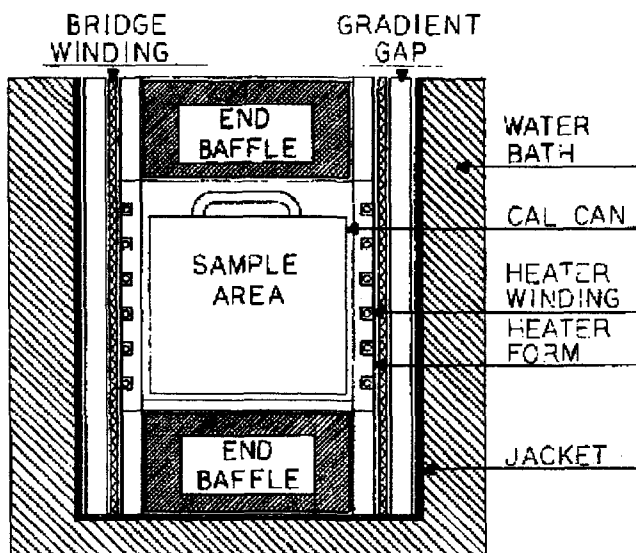


Figure 9. A calorimeter in use at the Department of Energy's Savannah River Site.



Figure 9 shows a version of the Mound transportable calorimeter system as permanently installed at the U.S. Department of Energy's Savannah River Site (SRS). This calorimeter is termed a servo controlled gradient bridge calorimeter. It is routinely used to assay plutonium ingots produced in the F Canyon/ F B-Line reprocessing facility. This facility's original mission was to produce plutonium for the U. S. weapons program but is now being used to reprocess and stabilize legacy materials ranging from irradiated reactor fuel remaining after the shut down of the SRS reactors to a variety of plutonium bearing scrap materials.

Figure 10 (Ref 13) shows the general structure of this type of calorimeter. The sample chamber is held at a constant temperature a combination of the output from the heater and the heat from the sample. The heat supplied from the heater is reduced from its initial set point by the amount of heat generated by the sample. The difference between the supplied heater power and the set point power is therefore the heat output of the sample. For this type of calorimeter typical performance is given in the table below /13,14/.



*Figure 10. Schematic of a gradient bridge calorimeter.*

**Performance Specifications for Mound Gradient Bridge Calorimeters**

Accuracy	0.5% at 2 Watts
Precision	0.1 %
Design Range	500 g to 5 kg (2 to 12 Watts)
Assay Time	4 hours for product 8 to 16 hours for scrap

### 3. Passive Neutron Assay Technique

Passive neutron assay is used to determine the spontaneous fission rate  $F_s$  of plutonium isotopes with even mass numbers. This value, together with the isotopic composition, is used to estimate the total mass of plutonium. A passive neutron assay instrument consists of three principal parts; the neutron detection head, the electronic chains, and the neutron pulse analyser.

A passive neutron detection head ideally has a  $4\pi$  geometry and consists of a polythene moderator surrounding the item to be measured. The moderator has thermal neutron detectors incorporated and is clad with cadmium on all free surfaces. Fast neutrons generated inside a sample may be partially slowed down and absorbed in the sample. The fraction of fast and epithermal neutrons leaking from the sample into the thermal neutron detector head is slowed down in the moderator, diffuses as thermal neutrons, and is partially absorbed by  $^3\text{He}(n,p)$  neutron detectors. Many different passive neutron detection heads, based on the above principles, have been designed over the past two decades.

The electronics chain transmits the electronic pulse train generated by detected neutrons to the analyser. Most commonly the AMPTEK chip /15/ in conjunction with a mixer based on OR gates with one gate for each signal line is used. In case of high count rates a special mixer as described in ref./16/ is recommended.

The signal pulse train analyser can be a variable dead time counter, an autocorrelator, also known as the shift register, or a time correlation analyser (or multiplicity counter). The former one has been proposed by Jacquesson /17/ and first used by Bondar and Birkhoff /18,19/. Although the theory of this counter has been considerably improved /20,21/, the use of the latter ones is recommended.

Bondar and Birkhoff /22/ were the first to apply the autocorrelator concept to mass assay of Pu samples for safeguards purposes. Thereafter Boehnel designed a dedicated hardware instrument and specified a basic interpretation model /23/. The shift register theory was further developed at Los Alamos /24/, KfK Karlsruhe /25/ and JRC Ispra /26/. The autocorrelator opens for each neutron signal followed by a pre-delay, an observation interval of a fixed time. It records, beside the total number of signals  $T$ , for each signal the number of counts in the observation interval. This number is accumulated in the 'R+A' register. The same is done in the 'A' register, which is delayed by the time  $T_d$  after the triggering interval (where  $T_d$  is much larger than the decay time of the detector head). At the end of the measurement the 'A' register contains the accidental counts,  $A$ , and the 'R+A' register contains in addition the correlated pair counts,  $R$ . These correlated pair counts, also called 'Reals', are generated by signals in the observation interval resulting from the same spontaneous fission process as the neutron triggering the observation interval. The accidentals are generated from signals in the observation interval originating either from a different spontaneous fission or from  $(\alpha, n)$  reactions.

Neutron coincidence counting or neutron pair correlation using the shift register is used extensively both for measurements of plutonium in bulk material and in waste. As a relative technique it works well for measuring samples which are characterised with known isotopic composition and multiplication close to one. The technique often fails, however, if samples are not well characterised or when the calibration is inappropriate.

Neutron correlation analysis, or neutron multiplicity counting, is an extension of neutron coincidence counting and considers single neutrons, correlated neutron doublets (pair correlation) and in addition correlated neutron triplets. In essence the frequency analyser (or TCA) determines the frequency of events where groups of a given number of neutrons are detected in observation intervals of fixed time. From the factorial moments of such frequency distributions the so called singlets  $R_1$ , correlated doublets  $R_2$  and correlated triplets  $R_3$  can be derived. Both pair and triple neutron correlation analysis were developed mainly at the Los Alamos National

Laboratory by Krick, Ensslin and Stewart/27,28/ and at the Joint Research Centre in Ispra /6,29-32/. An advantage of deriving the singlets, correlated doublets, and correlated triplets is that these quantities can be expressed in three equations, linear in the spontaneous fission rate (proportional to the Pu mass), which are functions of instrumental and sample parameters. The three equations are shown below.

$$R_1 = \varepsilon F_s M v_{s(1)} (1 + \alpha) \quad \text{Eq. 4}$$

$$R_2 = \varepsilon^2 F_s M^2 v_{s(2)} \left[ 1 + (M - 1)(1 + \alpha) \frac{v_{s(1)} v_{I(2)}}{v_{s(2)} (v_{I(1)} - 1)} \right] \quad \text{Eq. 5}$$

$$R_3 = \varepsilon^3 F_s M^3 v_{s(3)} \left[ 1 + 2(M - 1) \frac{v_{s(2)} v_{I(2)}}{v_{s(3)} (v_{I(1)} - 1)} + (M - 1)(1 + \alpha) \times \frac{v_{s(1)} v_{I(3)}}{v_{s(3)} (v_{I(1)} - 1)} \left( 1 + 2(M - 1) \frac{v_{I(2)}^2}{v_{I(3)} (v_{I(1)} - 1)} \right) \right] \quad \text{Eq. 6}$$

where

$$M \cong \frac{1 - p}{1 - p v_{I(1)}} \quad \text{Eq. 7}$$

$$\alpha = \frac{S_\alpha}{v_{s(1)} F_s} \quad \text{Eq. 8}$$

$$v_{j(\mu)} = \sum_{\mu=v} \binom{v}{\mu} P_{jv} \quad \text{Eq. 9}$$

and the symbols are:

$S_\alpha$  = ( $\alpha, n$ ) neutron emission rate of the sample

$F_s$  = spontaneous fission rate of the sample

$M$  = multiplication factor

$\mu$  can be 1, 2 or 3 to evaluate  $v_s$  or  $v_I$

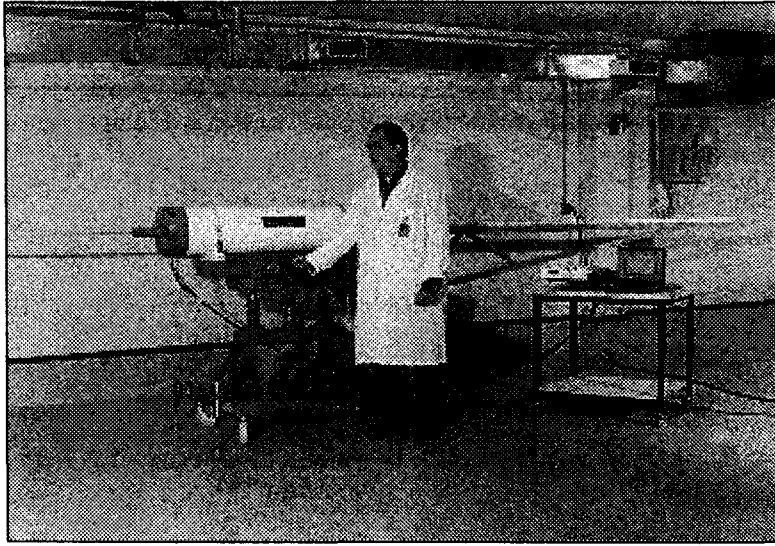
$p$  = probability that a fast neutron generates an induced fission event

$\varepsilon$  = probability for the detection of a neutron

$P_{jv}$  = probability of the emission of fast neutrons per prompt fission caused by a primary neutron generated by reaction  $j$  ( $j = 1$  for induced fission,  $j = s$  for spontaneous fission).

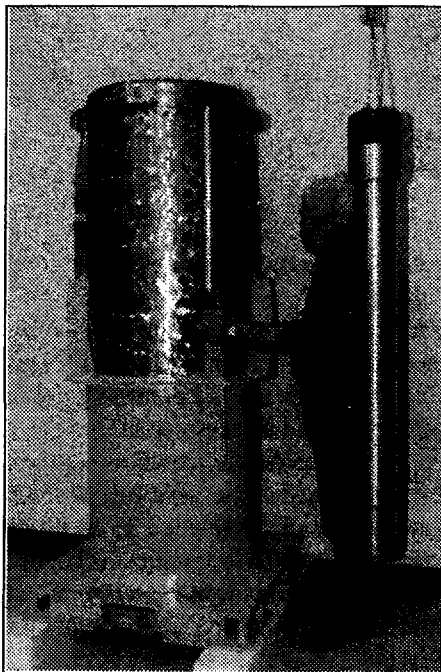
When applying the equations 4 to 6, three unknowns may be determined. In addition to  $F_s$ , it is possible to solve for two unknowns selected from the three parameters  $S_\alpha$ ,  $\varepsilon$  and  $M$ . In many situations at least one of these parameters may be considered known or negligible.

Over the last decade numerous tests were performed at the Joint Research Centre in order to validate the model as expressed in the equations 4 to 6. Reference 33 and 34 show results on plutonium containing waste drums of different sizes and various matrix compositions. For waste drums of large volume and variable matrix materials, the neutron detection probability cannot be considered known. In such applications the triple correlation analysis has shown particular advantages compared to calibration methods.



*Figure 11. MOX fuel pin counter for NDA of Pu.*

Passive neutron assay systems can also be applied directly to the process line in reprocessing plants. In many circumstances however, it is not possible to apply an absolute assay as suggested by equations 4 to 6. The reasons are different for the various process steps. In some areas limited space or access to the nuclear material prescribes compromises in the design of the detector head to the extent that the approximated  $4\pi$  geometry is not maintained and the point model of equations 4 to 6 is not applicable. At other stages the chemical solution of plutonium contains such a high level of gamma and  $(\alpha, n)$  neutron emission that the signal from correlated neutron bursts disappear in an uncorrelated background. In these cases calibrations or count rate verifications remain the



*Figure 12. Scrap counter for large MOX samples.*

only applications of passive neutron instrumentation. Calibration methods are often used together with sampling of liquid solutions at various process steps. The Reals obtained from a passive neutron measurement in a purpose built sample counter can be used to derive the fissile content of the sample if the instrument is carefully calibrated with known samples of the same chemical composition. Primitive passive neutron counters, which measure the total neutron count rate or the Reals count rate, are sometimes used to monitor holdup in pipework and tanks.

At the final stage of the reprocessing plant however, the neutron triple correlation can be applied to the mass assay of the separated products. At the Joint Research Centre several instruments have been built for this purpose. The figures 11 and 12 show two examples of instruments from the Joint Research Centre currently assaying Pu products from two European reprocessing plants.

The MOX fuel pin counter of figure 11 is used in conjunction with a frequency analyser to assay the Pu mass in fresh MOX fuel pins. This configuration is not problematic in terms of signal count rates as moderate rates are observed. However, counting times of only a few minutes are necessary to produce an absolute determination of the mass due to the high detection efficiency of the instrument. The only complication as compared to an ideal configuration is the fact that the fuel pins are longer than the sample cavity and that some considerations are necessary to handle the effect of the fuel outside the cavity.

The scrap counter of figure 12 is also used together with a frequency analyser to provide an absolute mass determination (no calibrations) of plutonium in MOX. The instrument is designed to assay containers of the dimensions shown in figure 12. The total MOX content of each container is about 30 kg. This large mass results in count rates as high as 350,000 to 400,000 counts per second. In order to calculate the Pu mass the measured quantities, the singlets, correlated doublets, and correlated triplets, must be corrected for dead time effects. At the Joint Research Centre the new analytical dead time correction /32/ has been implemented in the analysis software. The results from the first measurement campaign were very good. The deviation of the assayed to the declared Pu mass was typically 1 to 2 % for individually samples. At the end of the campaign the total assayed mass of Pu agreed with the declared mass with a negligible deviation.

### **III. CONCLUSIONS**

Accurate and timely accountability and verification measurements in reprocessing plants are essential, because this is the first time in the nuclear fuel cycle that plutonium can be measured. Radiation-based nondestructive assay measurements provide a rapid and accurate determination of SNM content at the input and the output of a reprocessing plant. When accompanied by destructive analytical measurements, they result in timely and accurate MC&A data for better nuclear material accountancy. These NDA measurements are employed by both plant operators for both safeguards and process control, and for verification by plant, national, and international inspectors.

A summary of the measurement techniques discussed and their average performance values is given in Table 1 /36/.

NDA Method	Sample type	Random Error (%)	Systematic Error (%)
K-Edge	Input, U, 1000 sec measurement time	0.25	0.20
K-Edge + XRF	Input, Pu, 1000 sec measurement time	1.0	0.30
K-Edge	Output, U, 1000 sec Measurement time	0.20	0.15
K-Edge	Output, Pu, 1000 sec Measurement time	0.20	0.15
High Resolution Gamma-Ray Spectrometry	Pu oxide, MGA code Pu-238	1.1	0.4
	Pu-239	0.25	0.3
	Pu-240	0.6	0.2 - 0.9
	Pu-241	0.5	0.2 - 0.5
	Am-241	1.0	1.0
Water Calorimetry	Low burn-up	0.5	0.2 - 0.5
	High burn-up	0.2	0.5 - 1.0
Air Calorimetry	Low burn-up	0.8	0.2 - 0.5
	High burn-up	0.3	0.5 - 1.0
Passive neutron	Pu metal	< 1	1 - 2
	PuO <sub>2</sub> Powder	< 1	2
	MOX Powder	< 1	3 - 5

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## DA AND NDA MEASUREMENT OF NUCLEAR MATERIALS "MAYAK" REPROCESSING PLANT

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### 1. Introduction

Methods for nuclear material (NM) analysis at RT-1 plant of PA "Mayak" are reviewed. Taking into account the wide range of fuel elements reprocessed, the paper considers the methods used at the main technological facility of the plant dealing with reprocessing of NPP fuel elements. In addition to principal characteristics of the methods involved, their applicability for quantitative accounting of NM is evaluated.

In order to better represent the material, the review is commenced with a brief characteristics of the NM accounting and control system at the facility.

### 2. System for NM accounting at RT-1 plant

#### 2.1. Technological flowchart

Major steps of reprocessing of the irradiated fuel of VVER include:

- cutting and dissolving the fuel in nitric acid;
- two-stage extraction purification of U, Pu, and Np with U and Pu separated during the first cycle;
- evaporating uranyl nitrate to the state of fusion cake;
- precipitation of plutonium oxalate followed by its calcination to dioxide;

Figure 1 shows major flows, where the nuclear materials are accounted at the facility.

Major characteristics of the NM flows are shown in Table 1.

As it follows from the Table above, RT-1 plant laboratory performs measurements of NM in a very wide range of concentrations - from tens weight per cent to million fractions. Some characteristics of the analytical methods used for nuclear material accounting and control (MC&A) are described below.

TABLE 1. Characteristics of major flows of NM at RT-1 plant

Flow No	Flow	NM content per flow		
		U	Pu	U-235
1	Irradiated fuel assemblies	up to 96%	up to 1%	1 – 1.5%
2	Solution of irradiated fuel	300 g/l	3 g/l	1 – 1.5 %
3.	Solution of enriched uranium	30g/l	-	16 – 90%
3'.	U oxide (yellow cake)	> 85%	< 30 ppb	16-90%
4.	Claddings after dissolution	< 0.5%	< 0.5%	1 – 1.5%
5.	Liquid wastes	< 10 ppm	< 1 ppm	1 – 1.5%
6.	Pu nitrate solution	< 50 ppm	up to 30 g/l	-
7.	U nitrate solution	up to 100g/l	< 5 ppb	2 – 2.5%
8.	Np nitrate solution	< 10 ppm	< 10 ppm	-
9.	Plutonium dioxide	< 30 ppm	up to 86%	2 – 2.5%
10.	Fusion cake of uranyl nitrate	up to 50%	< 5 ppb	2 – 2.5%
11.	Np nitrate concentrate	< 10 ppm	< 10 ppm	-

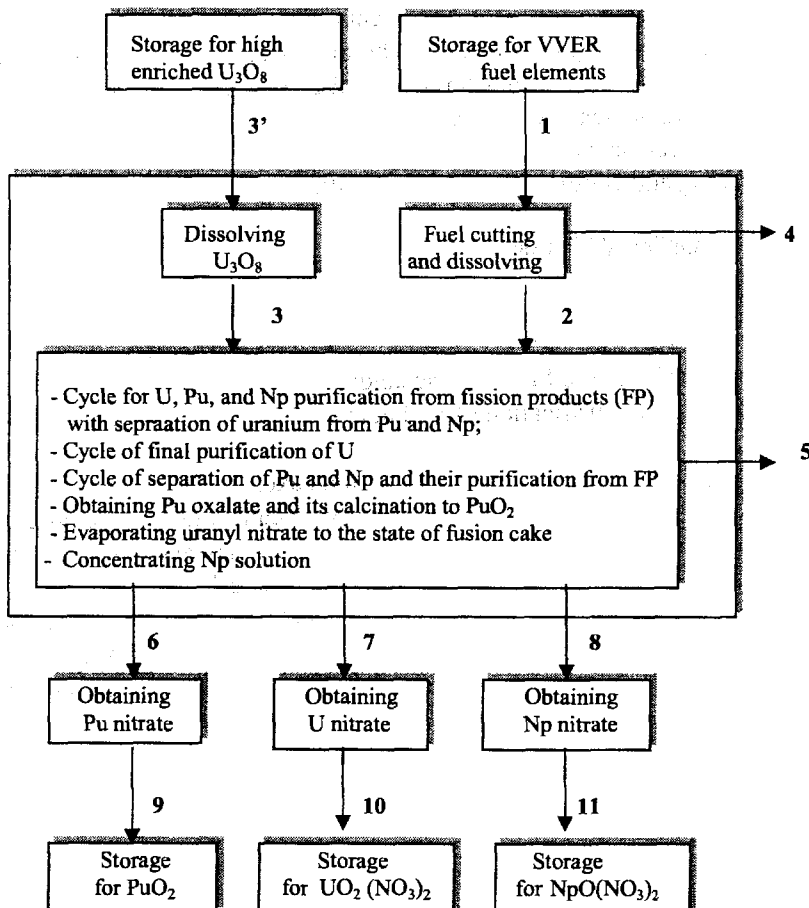


Fig.1. Major flows of nuclear materials at RT-1 plant facility for VVER fuel element reprocessing (figures at the arrows - numbers of flows containing NM)

### **3. Review of methods used for NM measurements at RT-1 plant**

Actually, at all key measurement points (KMP) the quantity of NM is defined as NM concentration as flow rate, volume, or mass (weight). Thus, NM concentration is a key accounting parameter. Brief description and quantitative characteristics of the methods for NM concentration measurement at the plant laboratory is given below (random error values are given at confidence level of 95%).

#### **3.1. Uranium concentration measurement**

##### *3.1.1. Irradiated fuel solution*

Method of  $\gamma$  absorptiometry is used for the measurement of uranium in this solution, based on the measurement of  $\gamma$ -ray absorption by the sample (matter). Radiation source is americium isotope; the thickness of the sample absorbing layer is 5mm, detector - scintillator crystal with photomultiplier. The method's error is up to 1-2%. It is used under hot cell conditions.

##### *3.1.2. High- and low level liquid wastes*

Method of spectrophotometry of dyed complexes of uranium with Arsenazo-3. Preparation of the sample is carried out when necessary, with the purification from interfering impurities in solid ion exchangers. The method error is 15-30%. The method is used in protection boxes.

##### *3.1.3. Product within the uranium line (Fusion cake of uranyl nitrate)*

Weighing method is used. Uranium in the weight form  $U_3O_8$  is obtained by calcinating the precipitate of uranium peroxide obtained by precipitation from formate buffer solution. The method error is 0.15%.

##### *3.1.4. Uranyl nitrate solutions*

As dependent on the presence of impurities and their types, uranium is measured either by the volume method - Davice-Grey titration with visual fixation of the equivalent point, or by  $\gamma$  absorptiometry (see 3.1.1.).

The titration error is 3%, that of  $\gamma$  absorptiometry - 1-2%.

#### **3.2. Plutonium concentration measurements**

##### *3.2.1. Solution of irradiated fuel*

Spectrophotometry method is used - measurement of light absorption by  $Pu(3+)$  ion stabilized with  $Fe(2+)$ . The method's accuracy for hot cell conditions is not very high, the error being 5-7%.

##### *3.2.2. High- and low level wastes*

The method is used identical to that described above for uranium spectrophotometry of dyed complex of plutonium with Arsenazo 3. The method error is 15-30%.

##### *3.2.3. Plutonium dioxide*

The controlled-potential coulometry method is used. The sample is preliminarily dissolved in nitric acid with fluoride ion added. The method error amounts to 0.3-0.4%.

##### *3.2.4. Plutonium nitrate solutions*

Method of  $\gamma$  absorptiometry is principally used, similar to that described in p. 3.1.1. for uranium. The error reaches 4-5%.

### 3.3. Measurement of uranium-235

#### 3.3.1. Solution of irradiated fuel

Method of mass spectrometry with thermal ionization of the sample material is used. The sample preparation includes chromatographic purification of uranium on solid sorbents. The method error for spent fuel of VVER reactors is of 4-5%.

#### 3.3.2. Enriched uranium oxide (yellow cake)

Method of mass spectrometry is used with thermal ionization of sample. With isotope content variation within the range of 1 - 99%, relative random error ranges from 4 to 0.2%.

#### 3.3.3. Solution after dissolving oxide (p.3.3.1)

Method of  $\gamma$ -spectrometry is used with measurement of 185.7 keV peak intensity. Ge detector is used for these measurements, the error being 5%.

### 3.4. Nuclear material measurement in residues after fuel dissolution

NM measurement in claddings after fuel dissolution (in flow 4) is carried out by neutron nondestructive assay (NDA) with error of 60-100%. This is the only NDA technique used at RT-1 plant for MC&A purposes.

These are principal methods for uranium and plutonium concentration measurements in various materials used for the MC&A system at the plant. Quantitative characteristics of the methods described above are summarized in Table 2.

As it can be seen from the data above, there are considerable errors in measurements of plutonium in solution of irradiated fuel, of uranium and plutonium in liquid and solid wastes, and U, Pu in intermediate flows. However, in order to determine the significance of these errors for MC&A, the analysis of accounting procedure is necessary for the facility.

**TABLE 2. Relative error levels of NM analysis methods used at RT-1 plant (confidence level of 95%)**

KMP	Component measured	Measurement method	Error, %
2	Uranium	$\gamma$ absorptiometry	0.9
2	Plutonium	Plutonium (III) spectrophotometry	5
3	Uranium	Devise-Grey titration	3
3	Uranium-235	$\gamma$ -spectrometry	2.6
3'	Uranium	weighing	0.3
3'	Uranium-235	mass-spectrometry	0.1
4	U, Pu	neutron NDA	50-100
5	Uranium	Spectrophotometry of U complex with Arsenazo III	15-30
5	Plutonium	Spectrophotometry of Pu complex with Arsenazo III	15-30
6	Plutonium	$\gamma$ -absorptiometry	5
7	Uranium	$\gamma$ -absorptiometry	0.9
8	Neptunium	Controlled-potential coulometry	0.5
9	Plutonium	Controlled-potential coulometry	0.4
10	Uranium	Weighing method	0.15
10	Uranium-235	Mass-spectrometry	0.2
11	Neptunium	Controlled-potential coulometry	0.5

#### **4. Measurement error effect on the quality of NM accounting at RT-1 plant**

##### **4.1. Effect of plutonium measurement error on the quality of NM accounting at RT-1 plant**

In order to assess this effect, the analysis of mathematical model of error of plutonium balance was carried out with the consideration of the flowcharts shown in Fig.1. Some final results of the analysis will be presented below, with details omitted.

1. The calculation of inventory difference uncertainty limiting the value of the inventory difference depends on the production output of the facility, frequency of inventory taking, sizes of NM batches, error of measurements such as batch volume and size, and NM concentrations.
2. The current levels of measurement error measurement currently existent provide the quality of plutonium accounting only for the facility production output below a certain level (400 t/year) and with the number of inventory taking procedures exceeding a certain value (more than once every 12 months). As the production output increases, with a decrease of inventory taking procedures, it is necessary to upgrade the measurement quality in order to obtain good results of NMC&A.
3. In case of an optimum production output and periodicity of inventory taking, the dispersion of inventory difference has the following structure:
  - 94% - dispersion of Pu detection in solution of irradiated fuel;
  - 5.8% - dispersion of detection in insoluble claddings of fuel;
  - 0.19% - dispersion of detection in plutonium dioxide;
  - 0.002% - in liquid wastes, high- and low level ones.

Thus, major components of the uncertainty include the error of analysis in the input solution and in the claddings after fuel dissolution. The rest components do not render a considerable effect on total uncertainty, and the quality of plutonium measurement at these KMPs can be considered satisfactory.

4. The analysis shows that the most effective way might be a decrease of measurement error for plutonium in irradiated fuel solution is to 1-1.5%. A further decrease proves to contribute to the upgrading of accounting quality very little, because in this case other components of total error for the balance become more significant.

##### **4.3. On the error of uranium measurement**

Since maximum admissible inventory difference for uranium of VVER fuel elements is considerable (about 3000 kg), it can be stated a priori that techniques for uranium analysis now in force are acceptable from the standpoint of their accuracy. Calculations show that the current level of errors for uranium analysis allows to have the error for inventory difference, that is one order of magnitude lower vs. maximum admissible value.

#### **5. Measurement quality control at RT-1 plant laboratory**

The measurement quality control at the laboratory is regulated by the laboratory order prepared in accordance with the industry standard OST 95 10889-87 now in force. Essentially, this order implies daily supervision of quality of the operators' work and equipment performance and periodic verification measurements of various types of reference materials. RMs of various levels are used for this purpose: national (federal), industry sector, enterprise RM, and certified mixtures. State and sector (departmental) RM are developed and manufactured mostly by institutes

subordinate to ministries and specialized organizations; RM for internal use at facilities and certified mixtures are manufactured at facilities.

Functions of an independent supervisor of the plant laboratory are implemented by the central laboratory and the service of chief metrologist of the enterprise, which perform annual commission supervision of the laboratory work quality. Besides, central laboratory carries out periodic quality examination using "unknown" samples.

### **Conclusions**

Methods of NM analysis and their quality assurance at the laboratory of RT-1 plant of PA "Mayak", are reviewed as applicable to the NMC&A system. The impact of analysis quality on the NMC&A system quality is analyzed. It has been shown that major component of the inventory difference for Pu is caused by the error of its measurement in the initial solution. The expedience of decreasing this error from 5% to 1-1.5% has been substantiated.

Errors of plutonium detection in the other flows of NM at the facility prove to be acceptable for the NMC&A system.

Accuracy of uranium concentration measurements proved to be satisfactory for all NM flows.



## THE CHOICE OF NUCLEAR MATERIAL MEASUREMENT STRATEGY IN "BULK-FORM" IN MATERIAL BALANCE AREA

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### INTRODUCTION

During the life cycle (from mining to final disposal), all nuclear materials (NM) appear in the "bulk-form" [1] at certain stages of the technological process, and sometimes, of the usage. Besides, the amount and content of the NM as "items" [1] are determined objectively by the measurement of the material quantities and isotopic composition of the bulk-form at technological stages preceding the appearance of items themselves. These data are registered in the forms and records (Technical specifications [TU], passport, special form, etc.). After that, these data, with results of indirect measurements, verification, or radiation passportization, appear to be the most reliable and accurate during the whole period of the use of this "item".

Therefore, one of major conditions determining the accuracy and reliability of nuclear material balance at federal level, as well as the guarantee of their non-proliferation (non-diversion) is a substantiated choice of strategy for NM measurement in "bulk-form" available and shipped-received by the nuclear material balance area (MBA) during material balance periods.

### BATCH OF NUCLEAR MATERIAL

In practical production and use of NM, the term "batch" is used in the following variants of meaning:

- Dispatching (shipment) batch of NM;
- Technological batch of NM;
- Accounting batch (portion) of NM [1,2].

The choice of quantitative characteristics of batch in each case is determined by various considerations. Let us consider the concepts and assumptions used as a basis for quantitative estimations.

#### *"Dispatching (shipment)" batch of NM*

It follows from the caption that the quantity of material in the batch is determined by economic justification for the transportation of such batch, or other ones (e.g., radiation-related). The quantity of material in such batch is indicated in technical requirements for the transportation referred to in the specifications, passport, form, etc, as well as in the shipment list. Obviously, the quantity of material in a shipment batch is not directly related to the choice of strategy.

***"Technological batch" of NM***

When designing technological processes of production or use of NM, as dependent on the capacity of technological equipment available and modes of its operation, NM is subdivided into parts – technological batches – according to quantitative requirements. This part of the flow is technological batch of NM. Thus, for example, a technological batch for NPP reactor is the number (mass) of fuel subassemblies (SA) loaded into reactor core for operation cycle. For the section of pellet sintering it implies the number (mass) of pellets per one loading into electrical furnace. Obviously, the choice of quantity in technological batch is not directly related to the choice of measurement strategy of NM in MBA.

***"Accounting batch", or simply "batch" of NM***

According to [2], batch is "...a portion of nuclear material treated as a unit of measurement (weighing) for the accounting purposes at a key measurement point (KMP), its composition and quantity being determined by a unified complex of NM specifications or measurements. Nuclear material in the batch can be in "bulk-form" or "items".

As it is obvious from this definition, this batch must be in a certain way associated with the NM measurement strategy in the MBA. However, the publication referred to does not give any idea, how this association is realized, and which is the effect dividing the total amount of NM in MBA into batches in the measurements.

Let us attempt to show with statistical approach to the mathematical processing of results that the latter variant makes it possible to considerably decrease the error in estimates of measured amounts of nuclear materials in MBA with a pre-specified confidence level vs. the former two variants.

**MEASUREMENT STRATEGY CHOICE**

Let us consider the following case of the realization of technological process for NM production within a certain MBA, for which primary information [3] and the value of MBA transfer function [4].

Structurally, this MBA includes storage for raw materials, technological sector for NM processing, storage for finished products, and that for "scrap" [1] formed as a result of the raw material processing as wastes and rejects. For a simplification, let the transfer function of technological process be assumed as:

$$Q_k = KQ_k = 0.9 \cdot Q_H, \quad (1)$$

where:  $Q_k$  - quantity of ready products ready for further use (kg);

$Q_H$  - quantity of source materials (kg);

$K$  - data transfer coefficient equal to  $K=1-K_n$ ,

$K_n$  - percentage of losses in the processing of raw materials (here assumed as  $K=1-K_n$ ).

Let the NM production capacity for the given MBA during the material balance period be  $Q=3000$  kg. For the rythmical operation of this MBA shipping the source material, reliable source material stock of 50% of production capacity is stipulated for the inter-balance period:  $Q_1=1500$  kg. Raw materials are shipped by batches distributed as approximately equal by time three times during the inter-balance period. Batch sizes are:  $Q_2=1000$  kg;  $Q_2=1300$  kg;  $Q_3=700$  kg. The material balance period -  $\tau=2$  months. The personnel of the section works in one shift, 6 h a day, 5-day working week. There are 40 shifts during the material balance period (3 shifts for the production section outage to "clean" the equipment and physical inventory taking and closing



the balance). Technological cycle -  $\tau_c = 6$  hours (one shift). The size of technological batch  $Q_T = 75$  kg. Three key measurement points (KMP) have been established for this material balance area (MBA):

- KMP 1 verification of source material quantity (input);
- KMP 2 verification of finished product quantity (output 1);
- KMP3 verification of the amount of "scrap" (output 2).

It is assumed here for a simplification of further considerations, that isotopics of NM in this MBA is not verified; the isotope composition is ensured by previous measurements, as well as by the use of tools and devices for preservation and surveillance, access control tools, and physical protection).

The instruments for NM quality assurance installed at each KMP are calibrated.

The system component of the instrument error is excluded by the use of corresponding standards.

The random component of the error -  $\Delta_{\text{random}} = 1\%$  of the value measured, the confidence limit being  $\gamma = 0.95$ .

And this value of error is ensured to be preserved for the whole range of measurements carried out at KMP1, KMP2, and KMP3.

The following strategies for NM quantity at MBA are possible, to be considered below.

#### **STRATEGY 1. Single measurement of dispatching, technological, and shipment batches**

Measurement of NM received by/shipped from MBA and those present in the MBA during the physical inventory taking is carried out by total quantity, i.e.:

1. The quantities of nuclear materials in the MBA are measured at the moment of reception, as one weight

a.  $Q_2' = 1000$  kg;

b.  $Q_2'' = 1300$  kg;

c.  $Q_2''' = 700$  kg.

2. The quantity of NM present at the MBA at the beginning of material balance period is measured as one weight.

$Q_1 = 1500$  kg.

3. Each NM shipment is measured as one weight

a. Ten shipments  $Q_4 = 270$  kg each (finished product);

b. One shipment  $Q_5 = 300$  kg ("scrap").

If it is assumed that no thefts occurred during the material balance period in this MBA, then the balance equation for the data of previous inventory taking, as well as for shipment and receiving data must give a result as:

$$Q_1 + (Q_2' + Q_2'' + Q_2''') - 10Q_4 - Q_5 - Q_3 = ID, \quad (2)$$

where  $Q_3$  - result of physical inventory taking (NM quantity actually available by the end of the material balance period);

ID - inventory difference (MUF, see [1]).

When all these values are measured ( $Q_i$ ) with the error at  $\gamma = 0.95$  taken into account, we have:

$Q_1 = Q_3^k = 1500 \text{ kg} \pm 15 \text{ kg}$  (first and last measurements), or  $1485 \text{ kg} \leq Q_{1,3} \leq 1515 \text{ kg}$ .

$Q_2' = 1000 \text{ kg} \pm 10 \text{ kg}$  or  $990 \text{ kg} \leq Q_2' \leq 1010 \text{ kg}$ ;

$Q_2'' = 1300 \text{ kg} \pm 13 \text{ kg}$  or  $1287 \text{ kg} \leq Q_2'' \leq 1313 \text{ kg}$ ;

$Q_2''' = 700 \text{ kg} \pm 7 \text{ kg}$  or  $693 \text{ kg} \leq Q_2''' \leq 707 \text{ kg}$ ;

$Q_4 = 270 \text{ kg} \pm 2.7 \text{ kg}$  or  $267.3 \text{ kg} \leq Q_4 \leq 292.7 \text{ kg}$ ;

$Q_5 = 300 \text{ kg} \pm 3.0 \text{ kg}$  or  $297 \text{ kg} \leq Q_5 \leq 303 \text{ kg}$ .

Since we measure  $Q_4$  10 times (10 shipments 270 kg each), then total error for  $10Q_4$  will amount to:  $\Delta_4 = 2.9 : \sqrt{10} = 0.92 \text{ kg}$ .

Using the methods of mathematical statistics for total error of several independent measurements (which do not belong to one general aggregate), in this case, according to [5], we have:

$$\Delta_{\gamma=0.95} = \sqrt{(\Delta_1^2 + (\Delta_2')^2 + (\Delta_2'')^2 + (\Delta_2''')^2 + \Delta_4^2 + \Delta_5^2 + \Delta_3^2)} = |ID|, \quad (3)$$

where,  $\Delta_1 \dots \Delta_5$  - extreme values of error measurement for  $Q_1 \dots Q_5$ ;

$|ID|$  - maximum admissible (critical) value of ID.

Substituting the error values, we have:

$$|ID| = \sqrt{(15^2 + 10^2 + 13^2 + 7^2 + 0.92^2 + 3^2 + 15^2)} = 27.9 \text{ kg} \cong 28 \text{ kg}.$$

Thus, if physical inventory taking carried out by this strategy renders the value of NM physically available at this MBA ( $Q_3$ ) in the range of  $\{1472-1528 \text{ kg}\}$  this will mean that no theft of nuclear material is recorded.

As we can see, the range of uncertainty in this strategy amounts to 56 kg, with confidence range of  $\gamma=0.95$ . This may be not acceptable, because significant amount of  $U^{233}$ , e.g., is equal to 8 kg, whereas  $U^{235}$  - 25 kg.

### STRATEGY 2. Measurement of NM by technological batches.

As it has been stated before, the size of technological batch for this MBA amounts to  $Q_T=75 \text{ kg}$ . The nuclear material from the raw material storage is given to the custodian of the shift in the amount needed to ensure the work of the shift, and at the end of the working day (shift), as technological cycle is completed, the same custodian returns this material to the storage for finished product and storage for scrap, it might be convenient and expedient to subdivide all the material into technological batches and measure it by these batches, even at the input and output sections of this MBA.

The measurement strategy, in this case, includes the following:

1. The quantity of NM received at the storage for source materials of this MBA is measured by technological batches ( $Q_T=75 \text{ kg}$ ).

a.  $Q_2' = 1000 \text{ kg}$  ( $13 \frac{1}{3}$  batch 75 kg each);

b.  $Q_2'' = 1300 \text{ kg}$  ( $17 \frac{1}{3}$  batch 75 kg each);

c.  $Q_2'' = 700 \text{ kg}$  ( $9 \frac{1}{3}$  batch 75 kg each);

Total: 40 batches 75 kg each.

2. The quantity of NM available at the storage facility of this MBA by the beginning of material balance period is measured by technological batches.

$Q_1 = 1500 \text{ kg}$  (20 batches 75 kg each)

3. Each shipment of NM is measured by batches.

$Q_4 = 270 \text{ kg}$  (3.6 batches 75 kg each)

Note: It might be justified to increase the size of shipment batch to 300 kg (9 shipments 300 kg each).

a. One shipment of scrap  $Q_5 = 300 \text{ kg}$ . (4 batch 75 kg each).

Balance equation (2) in this case can be written as:

$$20Q_T + 13 \frac{1}{3} Q_T + 17 \frac{1}{3} Q_T + 9 \frac{1}{3} Q_T + 36Q_T + 4Q_T + 20Q_T = ID, \quad (4)$$

where  $Q_T = 75 \text{ kg}$  – the size of technological batch.

Random component of measurement error in this case amounts to  $\Delta_{\text{random}} = 1\% \times Q_T = 0.01 \times 75 = 0.75 \text{ kg}$ , and total error of all measurements can be calculated according to [6].

$$\Delta_{\Sigma_{\text{random}}} = |ID| = \pm t_{\gamma, k} \Delta_{\text{random}} / \sqrt{n}, \quad (5)$$

where  $t_{\gamma, k}$  – Student's coefficient for the pre-set confidence level  $\gamma = 0.95$  and the number of degrees of freedom  $k = n - 1$ ;

$n$  – the number of measurements (in our case = 120);

$\Delta_{\text{random}}$  – random component of error of single measurement ( $\Delta_{\text{random}} = 0.75 \text{ kg}$ ).

Substituting, we have:  $|ID| = \pm 1.98 \frac{0.75}{\sqrt{120}} = \pm 0.135 \text{ kg}.$

Thus, with physical inventory taking by this strategy, the quantity of NM practically available at this MBA –  $Q_3$  will be within the range  $Q_3 I \{1499.865 \text{ kg} - 500.135 \text{ kg}\}$  - this will mean that no theft took place.

As we can see, with this strategy of measurement, the range of uncertainty for the estimation of NM practically available is  $\{0.27\} = |ID|$ , which is significantly less, than for the first strategy case. (Equivalent error of the metering instrument, which would allow to measure 1500 kg with this accuracy level:  $\Delta_{\text{eq}} = 0.007\%$  is unlikely to be achieved).

The result cited  $Q_3 = 1500 \pm 0.135 \text{ kg}$  seems to be giving hopes from the standpoint of safeguards for non-diversion of nuclear materials, because it ensures safeguards related to critical values for NM. I.e., for example, the amount of theft is 8 kg (Pu or U <sup>233</sup>), will be recorded beyond a doubt in both single measurement, and in inventory taking, because  $|ID| = \pm 0.135 \text{ kg} \ll 8 \text{ kg}$ . However, as we agreed for this example, the period of physical inventory taking is 2 months, but the theft (e.g., 0.5 kg per shift) can take place at the beginning of the material balance period, and to detect this theft, we shall have to wait till the beginning of inventory taking procedure.

- Note: The quantity of theft is 0.5 kg/cm in weighing of 75 kg of NM falls within the range of uncertainty of a single measurement  $\Delta_i = 1.5 \text{ kg}$ . I.e., in intended inaccurate weighing

and formation of the technological batch by the lower tolerance limit for a batch ( $Q_T \geq 74.25$  kg) 16 shifts would be enough to withdraw 8 kg of NM from the production line. Here, the gamesmanship is observed: results of measurements are within the tolerance limits for measurement error.

Therefore, the strategy of measurement (weighing) NM by “technological batches” could prove insufficient for the safeguards objectives, because the time of evaluation of the material conversion into ready components from metallic uranium or plutonium (according to [1]) is 7-10 days.

### **STRATEGY 3. Measurement of NM by “accounting batches”**

Now let us consider the notes necessary for the definition of numerical value of the “accounting batch”.

Significant quantity – approximate amount of nuclear material, for which the possibility of creating a nuclear explosive cannot be excluded, taking into account any conversion processes used [1,6].

For the same sources numerical values of significant quantities of NM are established as well. For the nuclear material, which can be directly used for fabricating nuclear explosives:

Plutonium (containing less than 80%  $Pu_{238}$ ) - 8 kg (for all isotopes);

Uranium-235 – 8 kg (for the isotope only) ;

Uranium (containing 20% or more  $U^{235}$ ) – 25 kg ( $U^{235}$ ).

For nuclear materials of indirect use (material, which can be used for the fabrication of nuclear explosive only after a corresponding conversion processing):

Uranium (containing less than 20% of  $U^{235}$  – 75 kg ( $U^{235}$ );

Thorium – 200 kg (for all isotopes of the element).

As we have seen already, for the case of withdrawal of a significant amount of NM from technological cycle in fractions smaller than the value of error for a single measurement, there is a possibility during a certain time period to withdraw this quantity, without detecting this fact.

Consequently, either more strict requirements should be addressed to single measurements' accuracy (which is not always feasible), or the quantity of NM measured (size of “accounting batch”) should be diminished so that the error of single measurement made it possible to accumulate the amount of NM necessary only during the whole material balance period, when theft of this amount of NM will be registered beyond a doubt when physical inventory taking will be performed. In this case, the value of “accounting batch” can prove small enough, and labor input for measurements – considerable enough, which can prove economically non-efficient, especially in case of high production output of technological section, in a low accuracy of measuring tools available (e.g.,  $\Delta_{\text{random}} \geq 5\%$ ). Therefore, it is expedient, simultaneously with the optimization of the “accounting batch” size, to use statistical approaches to the estimation of possible thefts during a time shorter, than the material balance period (1,2,3... n shifts).

- Note: It should be noted that the size of “accounting batch” thus defined (further to be termed as “critical value of accounting batch size”) will depend on the accuracy of measurements and the quantity of NM, which had passed the MBA and remained therein for the period between two inventory taking procedures consecutive in time.

Since there are four unknowns in this task (the “critical value of accounting batch size” , value of random component of error for its measurement, the amount of NM passed through the MBA and remaining therein by the moment of physical inventory taking, as well as the material balance period parameter), then this task should be solved by the method of consecutive

approximations, decreasing the “critical value of accounting batch size” stepwise. The solution of the optimization task with four unknowns in a general presentation would not be expedient from the engineering standpoint because of large labor input necessary and complicated calculations).

*The first step of iteration (approximation)*

The size of “accounting batch” ( $Q_y$ ) is equal to the size of “technological batch” ( $Q_y \setminus Q_t = 75$  kg). Measurement error (random component)  $\Delta_{\text{random}} = 75$  kg.

The size of thefts anticipated ( $X$ ) is assumed to be for variants as follows:

Variant 1 –  $X_1 = 0.5$  kg/sh.;

Variant 2 –  $X_2 = 0.2$  kg/sh.;

Variant 3 –  $X_3 = 0.1$  kg/sh.

Assuming the material under study to be  $\text{Pu}^{239}$ , we take the significant quantity of NM to be equal to  $|Q_{sq}| = 8$  kg.

Thus, in order to “accumulate” this quantity, the intruder has to carry out thefts during period ( $T_i$ ).

$$\text{Variant 1: } - T_1 = \frac{|Q_{sq}|}{X_1} = \frac{8}{0.5} = 16 \text{ shifts (16 batches 75 kg each);}$$

$$\text{Variant 2 - } T_2 = Q_y = 40 \text{ shifts (equal to material balance period);}$$

$$\text{Variant 3 - } T_2 = \frac{|Q_{sq}|}{X_3} = \frac{8}{0.1} = 80 \text{ shifts (exceeding the material balance period).}$$

It is clear that with error of  $\Delta_{\text{random}} = \pm 0.75$  kg, a single measurement would not give reliable data to state that theft had taken place, if in a repeated measurement (at the end of the shift) the quantity of NM became 74.5 kg. (74.8; 74.9) instead of 75 kg, measured at the beginning of the shift. Using expression (5), we find the time, during which the theft can be registered for variants 1, 2, 3.

$$1.1. \text{ 3 shifts } \Sigma X_1 = 1.5 \text{ kg } (\Sigma X_2 = 0.6 \text{ kg, } \Sigma X_3 = 0.3 \text{ kg}).$$

$$|ID| = \pm t_{\lambda, k}^* \frac{\Delta_{\text{random}}}{\sqrt{n}} = 4.3 \frac{0.75}{\sqrt{3}} = \pm 1.86 \text{ kg} > 1.5 \text{ kg} > (0.6 \text{ kg}) > (0.3 \text{ kg}).$$

\* - value  $t_{\lambda, k}$  is tabulated in [7].

Thus, for the three shifts one cannot state with certitude that no thefts ( $X_1$ ;  $X_2$  or  $X_3$ ) had occurred.

$$1.2. \text{ 4 shifts } \Sigma X_1 = 2 \text{ kg } (\Sigma X_2 = 0.8 \text{ kg, } \Sigma X_3 = 0.4 \text{ kg}).$$

$$|ID| = \pm t_{\lambda, k} \frac{\Delta_{\text{random}}}{\sqrt{n}} = 3.182 \frac{0.75}{4} = \pm 1.19 \text{ kg} < 2 \text{ kg } (> 0.8 \text{ kg; } > 0.4 \text{ kg}).$$

i.e., it can be stated with certitude that theft took place for the 4 shifts case, if the withdrawal per each shift had exceeded  $X_1 > 0.5$  kg (in the extreme case, registered amount is  $X_1 > 0.3$  kg/sh);

$$1.3. \text{ 5 shifts } \Sigma X_2 = 1 \text{ kg } (\Sigma X_3 = 0.5 \text{ kg}).$$

$$|ID| = \pm t_{\lambda, k} \frac{\Delta_{\text{random}}}{\sqrt{n}} = 2.776 \frac{0.75}{\sqrt{5}} = \pm 0.93 \text{ kg} < 1 \text{ kg } (< 0.5 \text{ kg}).$$

i.e., it can be stated with certitude that theft took place for the 5 shifts case, if each withdrawal had exceeded  $X_2 > 0.2$  kg/shift (instead of 40 shifts !)

1.4. 6 shifts  $\Sigma X_3 = 0.6$  kg.

$$|ID| = \pm t_{\lambda,k} \frac{\Delta_{random}}{\sqrt{n}} = 2.571 \frac{0.75}{\sqrt{6}} = \pm 0.78 > 0.6$$

i.e., total theft amounting to 6 kg (0.1 x 6) is not registered.

1.5. 7 shifts  $\Sigma X_3 = 0.7$  kg.

$$|ID| = \pm t_{\lambda,k} \frac{\Delta_{random}}{\sqrt{n}} = 2.365 \frac{0.75}{\sqrt{7}} = \pm 0.67 < 0.7 \text{ kg}$$

i.e., total theft of  $\Sigma X_3 = 0.7$  kg in this case is reliably registered for 7 shifts ( $\ll 40$  shifts).

*The second step of iteration (approximation)*

2. Let us decrease the size of "accounting batch"  $Q_y$  to 25 kg (i.e., "technological batch" ( $3Q_{sq} = 3Q_y$ )).

In this case,  $\Delta_{random} = \pm 0.25 \text{ kg} = 0.01 Q_y$ .

Let us assume the amount of anticipated thefts:

Variant 1 -  $X_1 = 0.2$  kg (of one "accounting batch");

Variant 2 -  $X_2 = 0.1$  kg

In order to accumulate a "significant amount" of NM  $|Q| = 8$  kg, it is necessary:

Variant 1 -  $T_1 = \frac{|Q|}{X_1 \cdot m} = \frac{8}{0.2 \cdot 3} = 14$  shifts, where  $m$  – the number of accounting batches per shift.

Variant 2 -  $T_2 = \frac{|Q|}{X_2 \cdot m} = \frac{8}{0.1 \cdot 3} \cong 27$  shifts.

Similarly to the first iteration, we shall estimate the number of shifts necessary for registering the NM theft.

2.1. One shift –  $\Sigma X_1 = 0.6$  kg ( $\Sigma X_2 = 0.3$  kg) – because three "accounting batches" are dealt with during one shift

$$|ID| = \pm t_{\lambda,k} \frac{\Delta_{random}}{\sqrt{n}} = 4.3 \frac{0.25}{\sqrt{3}} = \pm 0.62 \text{ kg} > 0.6 \text{ kg} (> 0.3 \text{ kg})$$

i.e., theft of 0.6 kg (0.3 kg) is not registered during one shift.

2.2. Two shifts  $\Sigma X_1 = 0.1.2$  kg ( $\Sigma X_2 = 0.6$  kg).

$$|ID| = \pm t_{\lambda,k} \frac{\Delta_{random}}{\sqrt{n}} = 2.571 \frac{0.25}{\sqrt{6}} = \pm 0.26 \text{ kg} < 1.2 \text{ kg} (< 0.6 \text{ kg}).$$

Thus, during two shift-long observation, it can be stated with certitude that theft of more than  $X_i = 43$  g from each "accounting batch" is recorded reliably.

As we can see from the comparison with the first step of iteration, the last variant renders higher numerical guarantees against NM theft for a shorter period of time. Therefore, if we consider that this guarantee level is acceptable for us, we assume  $X_i = 43$  g as a significant amount of material from an "accounting batch" from the standpoint of possible theft to be ( $X_i \leq 43$  g). Let this value be termed "criterion of material attractiveness" (or significance of a single theft)

( $|X_i|$ ); but preference will be given to such strategy of measurement in MBA, for which  $|X_{i+1}| < |X_i|$ , provided that financial expenditures for the organization of such measurements would be less than such losses.

Thus, in order to define the limits of MBA with material in bulk-form present, and for the preparation of the input information on this MBA, strategy of NM measurement must be chosen, capable of ensuring the solution of the safeguards problem and attaining the objective of NM management at Federal level.

Both tasks are solved through the ensurance of standardized accuracy and reliability (sufficient confidence levels) of the quantitative estimations of NM in regular inventory taking procedures, as well as in each transfer of NM.

The estimation technology, including that of measurement intended to ensure this requirement must be based on the following complex of activities:

1. Estimation of NM quantity present in an assumed MBA which passes through this area during the material balance period.
  2. Defining the "criterion of material attractiveness" which could result in an accumulation of critical amount of NM only during the material balance period.
  3. Defining the numerical size of "accounting batch" for the given NM.
  4. Defining periodicity of analysis of measuring "accounting batches" of NM, which could ensure meeting the chosen numerical characteristics of safeguards for non-diversion of NM.
- Notes: 1. For two consecutive MBAs along the NM flow, with different values of their parameters such measurement accuracy, production output, quantity of nuclear material, duration of material balance period, etc.), the choice of "accounting batch" size must be agreed upon.
  - 2. In order to ensure the uniformity of measurements and possibility of uniting the estimates of NM at Federal level, the information on NM quantity available in MBA with the material in the form of "items" also must be presented as "accounting batches, as well as for the adjacent MBAs with NM in bulk form.

## CONCLUSIONS

Summarizing the discussion on strategy for bulk form NM measurement for MBA, the following statements can be made:

1. Concepts have been defined such as "Shipment batch", "Technological batch", and "Accounting batch".
2. It has been found out that Shipment and Technological batches should be formed through the arrangement of group of measured Accounting batches.
3. The strategy for NM measurement based on the Accounting batch is shown to give a possibility to use the above advantages for the accounting purposes:  
Ensure safeguards of non-diversion of NM at quantitative (numerical) level, which is a higher grade of safeguards compared to the systems of accounting and control now in force in the US and Euratom.  
Ensure a guaranteed accuracy and reliability (confidence level) when making up NM balance in MBA and at Federal level, which has been realized only in part in the NMC&A systems listed above.
4. Strategy of NM measurement for MBAs containing NM in bulk form has been proposed.

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## **ANALYTICAL MEASUREMENTS FOR SAFEGUARDING LARGE REPROCESSING FACILITIES**

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### **Abstract**

Measuring samples taken at large reprocessing plants poses a particular challenge to an analytical laboratory. The size and complexity of these plants imply a high number of samples and a great variety of sample types. For meeting the safeguards goal of detecting the potential diversion of a significant quantity, strict accuracy requirements are imposed on these measurements. Consequently, criteria like accuracy, laboratory time, waste production, resource allocation and analysis costs determine the development and implementation of suitable analytical procedures. Analytical concepts have been developed at the Institute for Transuranium Elements for verification measurements at large reprocessing plants. These concepts take into account the specifics of the individual plant (feed materials, sample types and numbers) and the boundary conditions mentioned above.

Active and passive radiometric measurement techniques such as K-Edge densitometry (KEDG), X-ray fluorescence analysis (XRF), high resolution gamma spectrometry (HRGS) and neutron coincidence counting (NCC), either used individually or in combination, have become the primarily applied methods. Specifically, spent fuel solutions are measured by the Hybrid KEDG/XRF techniques, plutonium product solutions by KEDG, plutonium oxide and mixed oxide samples by NCC/HRGS, and uranium solutions by KEDG/HRGS. Intensive investigations have considerably contributed to improve the accuracy of these measurements. Additionally, development activities were carried out to further facilitate the use of the instrumentation and to increase the sample throughput by automation and unattended operation.

The application of methods based on chemical principles, due to their potential for superior accuracy, is shifting to specific fields like quality control i.e. to check the results obtained by physical methods and provide the reference basis for radiometric techniques. Accordingly, isotope dilution mass spectrometry and titration are used for the element and isotope assay in spent fuel solutions and products.

## 1. Introduction

Safeguards aims at the verification of the non diversion of fissile material from its intended and declared (peaceful) use. Therefore a reliable nuclear material accountancy has to be established by the plant operator. An at least evenly reliable system of verifications is the responsibility of the safeguards authority in charge.

Safeguarding nuclear material involves the quantitative verification of the accountancy of fissile material based on independent measurements. These measurements consist of a bulk measurement, hence a combination of mass and/or volume determination and the analysis samples taken from this bulk. The sampling procedure should guarantee that the sample is indeed representative of the bulk. It has furthermore to be ensured that the sample is not tampered with on its way from the sampling station to the measurement laboratory /11/.

A number of analysis strategies are currently being applied in parallel:

**On-line analysis** is limited to the monitoring of typically a single parameter by a non destructive measurement method. This technique is rather applicable for checking material flows.

**In-field analysis** requires the transport of mobile measurement equipment to the facility and the analysis of samples on the spot. This technique is only applicable to Uranium samples, as the transportation of Pu contaminated instrumentation poses practical problems. In another modification, in-field analysis uses equipment which is permanently installed at a certain facility, but it used and operated only by the inspectorate. This requires analysts to travel frequently.

**Off-site analysis** is the 'classical' way of performing verification measurements. Samples taken by an inspector are dispatched to an analytical laboratory which will then perform the measurements on request of the inspectorate. This enables the laboratory to operate under optimal conditions and with instrumentation permanently installed.

**On-site laboratories** represent a new concept which is based on the operation of a specialized laboratory working under the responsibility of the safeguards authority at the site of the facility to be inspected and analyzing material exclusively for safeguards purposes. Its independence and the confidentiality of the results produced have to be guaranteed by appropriate measures.

Depending on the plant size, material throughput and material type, an efficient measurement scheme has to be established, making use of one or more of the above mentioned possibilities.

The latter three analysis strategies do rely on the measurement of samples taken by an inspector. This paper focuses on the analysis of such samples as taken in large reprocessing plants and their treatment and measurement.

## 2. Sample Types

Depending on the nature of the plant, different types of samples are obtained. This includes different chemical compositions, different physical appearance and different handling techniques. The key elements of the nuclear fuel cycle immediately determine the types of samples to be expected. The sample spectrum in a reprocessing plant typically covers spent fuel solution, uranium products (uranyl nitrate solution, uranium oxide) and plutonium products (plutonium nitrate solution, plutonium oxide). Whenever a MOX fuel production plant is on the same site, MOX powders and pellets need to be analyzed as well.

### **2.1. Reprocessing**

Irradiated nuclear fuel can be reprocessed after appropriate cooling time. Most of the reprocessing processes are based on a liquid-liquid extraction for the separation of the valuable materials uranium and plutonium. The most widely used technique is the so called PUREX process. Therefore, in a first step the fuel needs to be dissolved. The solution (**reprocessing input solution**) is stored in the input accountancy tank. Samples of the solution are taken from this tank. These samples are analyzed for uranium and plutonium content and for the respective isotopic compositions. Samples of input solutions contain apart from the fissile elements also the fission products and some activation products. Because of this and due to the intense radiation such samples are delicate to handle and difficult to analyze.

The separation of uranium, plutonium and the fission products results in concentrated, rather pure **solutions of U and Pu**. Samples from these 'product' solutions are subject to measurements of the concentration of the respective element.

The product solutions are used as base material for oxide powder production.  $\text{UO}_2$  and  $\text{PuO}_2$  samples are again subjected to measurements of the fissile isotope and element content.

### **2.2. Fuel fabrication**

The civil fuel cycle knows two major categories of fuel: uranium oxide fuel and U/Pu mixed oxide fuel (MOX).

MOX fuel is manufactured from uranium and plutonium base materials. Depending on the production process U and Pu solutions or  $\text{UO}_2$  and  $\text{PuO}_2$  powders are used as starting materials. These however, are usually not measured (for safeguards purposes) in the fuel fabrication plant as this is already done at the reprocessing plant. In contrast to that the products, i.e. the MOX pellets are intensively verified. These samples have to be analyzed for uranium and plutonium element content as well as for their isotopic composition. The  $^{241}\text{Am}$  content provides useful information on the last plutonium purification.

Metallic fuels or fuels of high initial  $^{235}\text{U}$  enrichment are not commonly used in commercial reactors for electricity generation. These fuels therefore represent only a marginal fraction of the whole fuel production. Samples of these types of fuel are nevertheless of high relevance to Safeguards Authorities. They are analyzed in the laboratory with great care.

## **3. Information Requested**

Depending on the sample characteristics and on the facility, the analytical requirements are set. As already indicated in the previous chapter, a variety of samples of different chemical and physical properties have to be analyzed. The information requested usually focuses on the one hand on the uranium isotopic composition, where the  $^{235}\text{U}$  isotope abundance is the most relevant information for safeguards purposes. The uranium content in a sample also needs to be determined. The combination of the latter with the mass of the bulk and the  $^{235}\text{U}$  abundance allows to calculate the total amount of fissile uranium. On the other hand, information on the plutonium element content is required, additionally the plutonium isotopic composition needs to be known. The combination of the sample analysis results with the information on the bulk results in the total amount of plutonium.

The knowledge of the  $^{241}\text{Am}$  concentration, relative to the amount of Pu, allows to draw conclusions on the last plutonium separation.

Table 1 summarizes the plant types, material types and analysis types typically encountered in the fuel cycle. The third column specifies the sample sizes taken for verification measurement purposes. They are specified such that the uncertainties arising from sampling are kept to a minimum /1/. It has to be emphasized that the amount of material actually required for a measurement can be considerably lower (compare table 2).

**TABLE 1. Simplified overview of plant categories, material types, desirable sample sizes and analyses**

Plant Type	Material	Sample Size /1/	Analysis
Enrichment	UF <sub>6</sub>	4-8 g	U conc., U iso.
Fuel Fabrication	Solution: UO <sub>2</sub> <sup>2+</sup>	10 g	U conc., U iso.
	Pu	1-5 g	Pu conc., Pu iso.
	Powder: UO <sub>2</sub>	10 g	U conc., U iso.
	PuO <sub>2</sub>	2 x 0.5 g	Pu conc., Pu iso., Am
	Pellets: UO <sub>2</sub>	7-20 g	U conc., U iso.
	MOX	2 x (5-10) g	U conc., U iso. Pu conc., Pu iso., Am
Reprocessing	Solution: Spent Fuel	1-5 g	U conc., (U iso.) Pu conc., Pu iso.
	UO <sub>2</sub> <sup>2+</sup> Nitrate Soln.	10 g	U conc., (U iso.)
	Pu Nitrate Soln.	1-5 g	Pu conc., Pu iso.

#### 4. Sample Analysis Methodology

For any of the quantities to be determined as mentioned in table 1, a selection of analytical techniques is available, each of them could be applied to attain the desired goal. The choice of the measurement method to be applied for the determination of a certain parameter depends on a number of criteria, such as:

- sample composition
- available amount of material, which may be limited due to:
  - activity or dose rate restrictions
  - sample transport regulations
  - sampling procedures at the plant
- desired measurement uncertainty
- instrumentation and manpower available
- tolerable measurement delay
- creation of (secondary) waste

Whatever the method of choice might be, there are always advantages and disadvantages. Methods enabling a higher accuracy might require higher investment and/or running costs or might be more demanding in terms of operator skills and analysis time. This evaluation should in any case

be done in the light of the analytical needs, the available resources and the desired degree of "fitness for purpose". The list of methods presented below is neither exhaustive nor is it intended to describe a preference in any form. However, it covers the techniques currently being used or analyzing samples taken at large reprocessing plants.

#### 4.1. *Element Assay*

The determination of the content of uranium and/or plutonium is of key importance for establishing the material balance in a plant, for accountancy purposes and for the verification of the latter. It is therefore essential to have a method at hand that allows to determine the respective element concentrations in samples taken at some place in the plant. Classical chemical methods compete with methods based on physico-chemical or purely physical principles. Whatever principle is applied, the analytical goal is the quantitative determination of the amount of uranium or plutonium in a sample. The different techniques may require different ways of sample conditioning (e.g. dissolution, dilution, special geometry).

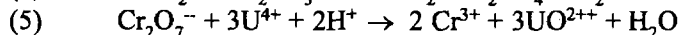
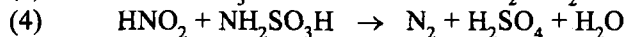
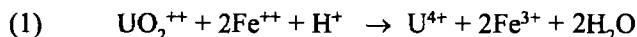
##### 4.1.1. *Titration*

Titrimetric measurements are carried out by determining the volume (or mass) of a solution of accurately known concentration (the titrant) which reacts quantitatively with the solution of the substance to be determined (the titrand). The point at which the reaction is just complete is called the equivalent point or stoichiometric end-point.

This end-point can be indicated by

- the potential between an indicator electrode and a reference electrode, **potentiometric titration**
- the change in electrical conductivity, **conductometric titration**
- the current which passes through the titration cell between an indicator and a reference electrode, **amperometric titration**
- the change in absorbance of the solution, **spectrophotometric titration**

Uranium can be determined by potentiometric titration using the so-called "Davies and Gray" method /2/. It is based on the reduction of uranium(VI) to uranium(IV) in concentrated phosphoric acid solution, in the presence of sulfamic acid, by reaction with iron (II) sulfate. The excess of iron(II) is subsequently oxidized by nitric acid in the presence of molybdenum. The uranium(IV) is determined by mass titration with standardized potassium dichromate solution to a potentiometric end point.



Interferences are to be expected from bromide, iodide, chromium(III), silver(I), tin(II) and vanadium(IV) and (V).

Plutonium can also be determined by potentiometric titration, using the so called "silver oxide" method /3/. First the plutonium is quantitatively oxidized with silver(II) oxide. The excess of silver is destroyed by adding sulfamic solution. The plutonium(VI) is then reduced to plutonium(IV) with an excess of iron(II) sulfate. The excess is titrated with potassium dichromate solution.

Interferences are observed from V(V), Mn(II), Am and Np.

Titration techniques, also others than potentiometric titration, are widely applied for verification measurements in pure products, i.e. in U and Pu solutions and in the respective oxides.

#### 4.1.2. Coulometry

Coulometry is considered to be a reliable method for the determination of uranium and/or plutonium. This method does not require a reference material for calibration, as it measures electrical charges and time. Coulometry is consequently a "primary method of measurement". It has furthermore the potential of being highly precise and accurate. However, reference materials are required to verify the proper working of the instrumentation.

The uranium determination by controlled potential Coulometry calls upon the reduction of uranium(VI) to uranium(IV) at a mercury electrode in sulfuric acid. A potential of -0.325V is applied for the reduction reaction. The amount of uranium is calculated from the number of electrical charges (Coulombs) required to complete the reaction. The end-point of the reaction is reached when the residual current (background) is as low as some  $\mu\text{A}$ . Corrections have to be applied for the blank current and the background. Interferences may arise from impurities such as copper, iron and manganese.

The determination of plutonium applies the oxidation/reduction of plutonium between its oxidation states +3 and +4 in sulfuric acid. Reduction is performed at a potential of +0.270 V, while the oxidation step requires +0.670 V. Interferences may arise from iron present in the sample solution.

Coulometry is only applied in a few laboratories for routine verification measurements on U and Pu product solutions or dissolved oxides.

#### 4.1.3. Gravimetry

The gravimetric determination of uranium and plutonium is based on the assumption that calcination of a (rather pure, solid, oxidic) sample of either element will lead to a (stable) compound of defined stoichiometry. This compound is then easily weighable, from the difference in sample masses observed before and after calcination, the element content in the initial sample can be calculated. Corrections have to be applied for impurities contained in the sample, as they will cause systematic errors. Hence, gravimetry always requires an impurity determination. The latter may be achieved glow discharge mass spectrometry (GDMS), spark source mass spectrometry (SSMS) or inductively coupled plasma mass spectrometry (ICP-MS).

Gravimetry is also a "primary method of measurement" according to the BIPM definition. As it requires only weighing data and information on the sum of impurities, its potential for precision and accuracy is unsurpassed. Additionally, the amount of secondary waste produced is very small. This method is therefore of growing interest to safeguards laboratories.

Uranium is heated in air or oxygen at 950°C in order to obtain  $\text{U}_3\text{O}_8$ . If prepared under these conditions, no deviations from stoichiometry are to be expected.

Similarly, Plutonium is heated in air or oxygen at 1250°C in order to obtain  $\text{PuO}_2$ . If prepared under these conditions, the latter compound has been demonstrated to be stable and stoichiometric.

#### 4.1.4. K-Edge Densitometry

Uranium or plutonium can be determined in a sample by K-Edge Densitometry (KED). The method /4/ uses a highly collimated X-ray beam passing through a solution sample of well defined path length. Its transmission is measured as a function of energy in critical energy regions. The underlying measurement technique is the K shell absorption edge spectrometry, colloqui-

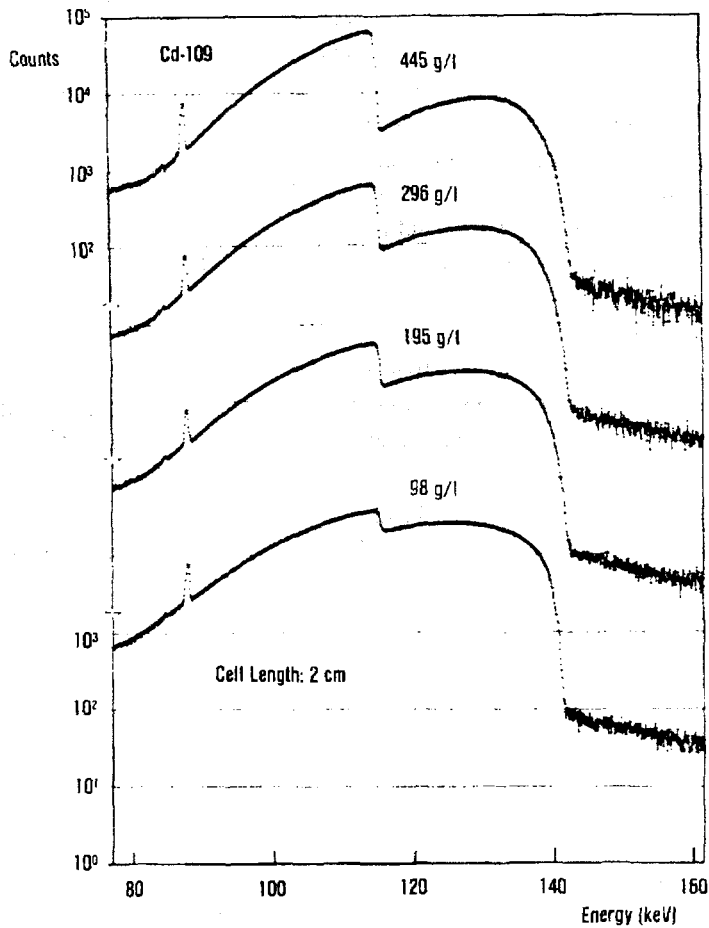


Fig. 1a K-Edge jump for different uranium concentrations

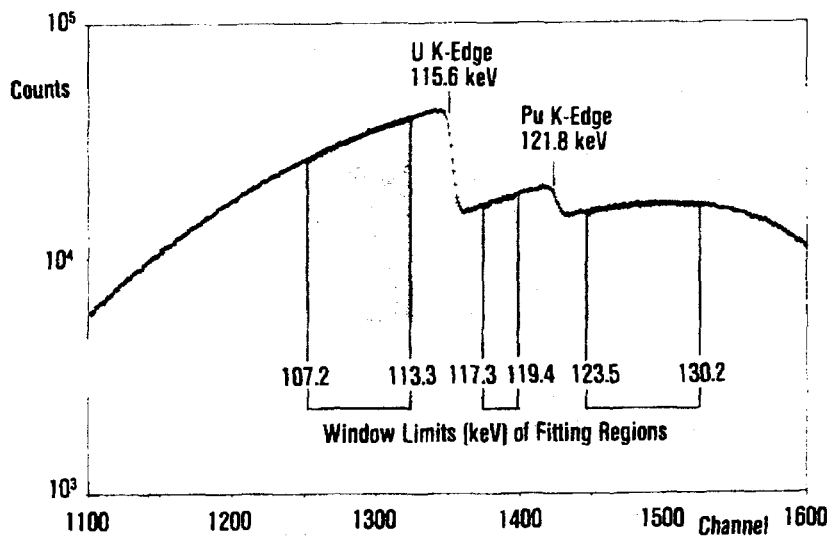


Fig. 1b X-ray transmission spectrum for a solutions with a U/Pu ratio of about 3

ally called K-edge Densitometry. The abrupt change of the transmitted X-ray intensity at the K absorption edge is a measure of the uranium or plutonium concentration in the sample as can be seen from fig. 1. The K-edge instrument requires a series of carefully characterized solutions of uranium and/or plutonium for establishing a calibration curve.

K-edge Densitometry can be applied to uranium or plutonium solutions from 25 g/L up to saturation (approx. 400 g/L). Interferences arise only from elements having their K absorption edge in the same energy region as the element under investigation. Consequently, the simultaneous presence of uranium and plutonium needs to be taken into account and an appropriate correction has to be applied.

K-Edge Densitometry delivers measurement results as volume concentration, i.e. in milligrams per milliliter or grams per liter. Most other techniques deliver results in mass concentration, i.e. in milligrams per gram. Consequently, one needs to determine the density of the solutions under investigation in order to be able to compare the measurement results obtained by the different techniques.

K-Edge is most advantageously applied to rather pure solutions of uranium and plutonium. If U and Pu are simultaneously present in a solution, the Hybrid K-Edge technique needs to be applied (see 4.3.2)

#### 4.1.5. X-Ray Fluorescence

An X-ray beam of larger divergence stimulates the emission of characteristic X-rays from uranium and plutonium (X-ray fluorescence, XRF). The intensities of the induced X-rays may be used for the determination of the U/Pu ratio in a sample or, after appropriate calibration, for the absolute determination of the respective amounts of element. In the first case the U/Pu ratio is derived from the net peak areas of the  $UK\alpha_1$  and the  $PuK\alpha_1$  X-rays. The latter case calls upon the peak area measurement of the  $K\beta_{1,3}$  line of uranium and plutonium.

Interferences are to be expected from any X-ray or soft  $\gamma$  lines of similar energy. Furthermore the self-absorption in the sample has to be taken into account. Measurement uncertainties of about 0.7% have been demonstrated.

#### 4.1.6. Isotope Dilution Mass Spectrometry

Isotope dilution mass spectrometry (IDMS) is based on the addition of a known amount of enriched isotope (called the "spike") to a sample /5,6/. After equilibration of the spike with the sample, mass spectrometry is used to measure the altered isotopic ratio(s). The concentration of the element under investigation can be derived from the change(s) in isotope ratio(s). Consequently, only weighings (of sample and spike mass) and measurements of ratios (of ion beam intensities) have to be performed. The actual measurement is performed after equilibration of spike and sample and chemical separation of the element of interest. This assures the removal of isobaric interferences and a smooth ionization process.

Uranium samples are usually spiked with  $^{233}\text{U}$ , an isotope which is not present in the sample. Therefore a single measurement allows to simultaneously determine the uranium concentration and the isotopic composition of the sample. Also enriched  $^{235}\text{U}$  or  $^{236}\text{U}$  may be used as spike isotopes, this however requires independent measurements of the ratios in the unspiked and the spiked sample.

Plutonium samples can be spiked with  $^{244}\text{Pu}$ , an isotope which is usually not present in the sample. Due to its very limited availability, the use of this isotope has been restricted to exceptional cases. Mostly, plutonium is spiked with enriched  $^{242}\text{Pu}$ . Furthermore, the application of  $^{239}\text{Pu}$  or  $^{240}\text{Pu}$  has been demonstrated successfully /12/.



IDMS is a highly selective method. It has the potential for high accuracy and precision. Also IDMS is a "primary method of measurement". Thermal Ionization Mass Spectrometry (see section 4.2.1) is mostly applied as isotope ratio measurement technique. However, also ICP-MS may be used, however allowing somewhat higher uncertainty. IDMS has found wide application in safeguards measurements and is applied in almost all analytical laboratories. Due to its high selectivity and its potential for high accuracy, IDMS is the method of choice for analyzing samples of spent fuel in reprocessing plants.

## **4.2. Isotope Assay**

Besides the determination of the element concentration, the measurement of the isotopic composition of uranium and plutonium is of interest. This is due to the fact that not the elements uranium or plutonium are fissile, but their isotopes of uneven mass numbers. Despite this fact, plutonium is regarded as fissile material, irrespective of its isotopic composition. In contrast to that, safeguards authorities pay particular attention to the uranium isotopes  $^{235}\text{U}$  and in special cases to  $^{233}\text{U}$ .

However, the accurate determination of the isotopic composition of U or Pu is of prime importance for verification purposes. Different measurement techniques based on different measurement principles are available for this purpose. The choice of the method depends on the requested accuracy, the nature of the material and other factors as discussed already earlier.

### **4.2.1. Thermal Ionization Mass Spectrometry**

Thermal Ionization Mass Spectrometry (TIMS) is widely applied for isotopic measurements. A sample preparation step prior to the actual measurement is required; this consists of the separation of the element of interest from other elements (e.g. matrix materials or impurities). The sample is then deposited onto a filament from which it is evaporated once being introduced in the mass spectrometer. These "vapors" are then atomized and at a hot surface ionized, from which the name "thermal ionization" is derived. The species  $\text{U}^+$  are subjected to an acceleration by applying a high voltage and subsequent mass separation (e.g.  $^{234}\text{U}^+$ ,  $^{235}\text{U}^+$ ,  $^{236}\text{U}^+$ ,  $^{238}\text{U}^+$ ) by means of a magnetic field, an electrostatic field or a quadrupole.

An appropriate detection system allows the measurement of ratios of ion beam intensities. The isotope abundances are derived from these ratios. TIMS relies on chemically purified samples in order to avoid isobaric interferences. TIMS is therefore very selective and has proven to be highly precise and to have a potential for being very accurate. This method is widely applied in nuclear analytical laboratories and suitable for practically all types of material.

### **4.2.2. Gamma Ray Spectrometry**

Also radiometric methods can be applied for isotope assay. These methods, however, are limited to non stable isotopes emitting either  $\alpha$ -particles or gamma rays. The most prominent is certainly gamma ray spectrometry. It uses the characteristic gamma lines, or more precisely the energy of the gamma rays emitted from a particular isotope. Their intensity is a measure of the number of atoms present in the sample. It is applied in a variety of instrumental and software modifications. Detectors of different geometries (planar, coaxial, dwell) and prepared from different materials (NaI(Tl), Si(Li), Ge(Li), high purity silicon, high purity germanium, CdTe) are in use. Depending on the application, the desired spectral energy resolution, the efficiency and the useful energy range, the type of detector has to be selected. Furthermore, a number of computer codes have been developed for spectral deconvolution, for data reduction and evaluation.

One of the codes used is the so called MGA code, developed at Lawrence Livermore Laboratory /7/. The Multiple Group Analysis (MGA) allows to determine the isotopic composition of plutonium samples without external efficiency calibration. Its major drawback arises from the fact that  $^{242}\text{Pu}$  does not show useful gamma rays. This isotope abundance has therefore to be estimated using isotope correlation techniques. However, it is a non destructive technique which has proven its capability in field application /8/ as well as under laboratory conditions.

The determination of the  $^{235}\text{U}$  enrichment is typically done by measuring the intensity of the 186 keV line of this isotope. A particular geometry and calibration is required as the other isotopes can only insufficiently be determined. One of the major advantages is the possibility to use cheap NaI(Tl) detectors of relatively low resolution. A more sophisticated approach is the application of the MGAU code. Again, no external calibration is required.

Gamma spectrometric methods have considerably improved in performance over the last years. They are widely applied for accountancy and verification measurements.

#### 4.2.3. Alpha Spectrometry

Alpha spectrometry uses the discrete energy of the  $\alpha$ -particles emitted by certain radioactive isotopes for the identification and quantification of the respective nuclides. It requires the careful preparation of thin layers of analyte. The resulting  $\alpha$ -spectrum allows to determine the activity ratio of the  $\alpha$ -active isotopes present in the sample. The isotope abundance ratios can be derived from that. Its application in safeguards is limited to the determination of the  $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$  ratio. This information is complimentary to the mass spectrometrically obtained information on the  $^{238}\text{Pu}/^{239}\text{Pu}$  ratio which sometimes suffers from isobaric interference of  $^{238}\text{U}$ .

### 4.3. Combined Methods

It is evident that the methods discussed so far can to some extent be combined for simultaneous use. Powerful measurement methods may result, often enabling increased sample throughput, reduced operator radiation doses and more efficient laboratory work. Combined methods usually rely on physical measurement principles which are applicable at the same time.

#### 4.3.1. COMPUCEA

The combined product uranium concentration and enrichment assay (COMPUCEA) calls upon a combination of gamma ray spectrometry for measuring the  $^{235}\text{U}$  abundance and K-edge Densitometry for the uranium element concentration measurement. The methods involved have been discussed in some detail in the sections above. Applications of the instrument are in verification measurements at enrichment plants, at fuel production plants and for the uranium product streams of reprocessing plants. Due to its compactness, the instrument is suitable for mobile use. The Euratom Safeguards Directorate has successfully been applying a COMPUCEA for in-field measurement campaigns at a large reprocessing plant and at Low Enriched Uranium fuel fabrication plants.

#### 4.3.2. Hybrid K-Edge / K-XRF Densitometry

The combination of K-edge Densitometry with X-ray fluorescence resulted in the so called Hybrid K-Edge (HKED). This instrument is using a single X-ray source for both parts of analysis, the K-edge absorption and the fluorescence excitation. It has proven to be an extremely useful analytical tool in the verification of reprocessing input solutions. It is also applicable to mixtures of uranium and plutonium. The combination of the two techniques allows the simultaneous and quantitative determination of uranium and plutonium. This can even be done directly from samples of highly radioactive input solutions.

The number of such instruments in use and being installed is increasing, as the experience with existing instrumentation demonstrates the reliability of the results. At each of the two large European reprocessing plants, the Euratom Safeguards Directorate installed a hybrid K-Edge instruments. They have been operated by ITU staff for many years. Several hundred samples of spent fuel solution have been analyzed each year providing a solid experience in the operation of this type of instrument. The results are usually reported to the inspectors within a few days after sample arrival.

#### *4.3.3. Neutron-Gamma Counting*

Neutron coincidence counting relies on the spontaneous fission on  $^{240}\text{Pu}$  and the neutrons produced with each fission process. The neutron count rate is a measure of the amount of  $^{240}\text{Pu}$  present in the sample. However, accurate information on the isotopic composition of the sample is required in order to:

- correct for neutronic contributions from other Pu isotopes ( $^{238}\text{Pu}$  and  $^{242}\text{Pu}$ )
- calculate the total amount of plutonium

If applied in combination with high resolution gamma spectrometry, a complete plutonium assay in solid samples (MOX or  $\text{PuO}_2$  powder) is possible /9, 10/. The Euratom Safeguards Directorate has installed a neutron/gamma counter at a MOX production facility and gained positive experience in measuring small samples (individual pellets, powders). Such instruments will be used as primary tools for verification of Pu containing oxides in the on-site laboratories at the two large reprocessing plants.

### **5. Analysis of Samples for Verification Purposes**

The analysis of samples for verification purposes needs to meet certain requirements, such as:

- accuracy of results
- laboratory delay
- efficient use of resources
- sample throughput

These requirements are intended to ensure an efficient and effective safeguarding of nuclear material. On the laboratory level measures have to be taken to make sure that these requirements can be met. These measures comprise organisational, infrastructural and scientific/technical arrangements. This can best be illustrated using a concrete example:

Inspectors take approximately a thousand samples per year in a large reprocessing plant. As mentioned earlier, these samples may be analysed in a specialized laboratory after having been transported. Seen the high number of samples and the delays going along with the transport arrangements, EURATOM decided to install On-Site Laboratories at the two large reprocessing plants in Europe. Already earlier, a hybrid K-edge instrument had been installed at each plant for verification measurements on spent fuel solution.

#### *5.1. Analysis in the laboratory*

The analysis of samples in the laboratory (be that off-site or on-site) represents certainly the ideal case from the measurement point of view. Optimal measurement conditions can be achieved, profiting from a well developed infrastructure and technical support. A selection of the measurement techniques described above can be applied.

In the on site laboratories the following measurement techniques and instrumentation are routinely applied for verification sample analysis:

- thermal ionization mass spectrometry

Finnigan MAT sector field mass spectrometers are available

- isotope dilution mass spectrometry

sample preparation is performed in a glove-box by laboratory robots (Zymark)

- neutron coincidence counting combined with gamma spectrometry

a specially developed instrument is used

- K-Edge Densitometry

several instruments are available

- Hybrid K-Edge

several instruments (equipped with sample changers) are attached to the hot cells

- alpha spectrometry

several devices from Canberra and Ortec, some of them modified for use by the robot in a glove-box, are available

- COMPUCEA

an instrument is available in a dedicated glove-box (uranium only !)

#### *5.1.1. Infrastructural Considerations*

These instruments are operated by local computers (personal computers or work stations). The computers are connected via a local area network. A specifically developed software manages the analyses, collects the data, calculates the results, applies an integrated quality control and allows communication with the inspectorate.

Table 2 summarizes the amounts of material typically for a particular technique, required to carry out a measurement.

## **5.2. In-Field Measurements**

In-field measurement campaigns using mobile equipment are carried out exclusively on uranium samples to avoid the transport of Pu contaminated equipment. In-field analysis comprises titration, COMPUCEA, thermal ionization mass spectrometry and gas source mass spectrometry. This spectrum of analytical techniques covers the needs arising in uranium handling facilities. It is perfectly suitable for verification measurements during physical inventory taking (PIT) by the inspectors. Hence, in-field measurements are an excellent tool for near real time verification measurements.

Another important area of in-field measurements covers the verification analyzes of reprocessing input solution samples. These measurements are carried out using equipment which is permanently installed (Hybrid K-Edge instrument) in the plant operator's laboratory, but exclusively used for safeguards measurements. More than six hundred input samples have been measured in 1997 by ITU staff members applying the HKED instrument at one of Europe's biggest reprocessing plants.

Table 2. Typical amounts of sample needed to perform a measurement

Method	Typical amount of element required for a single measurement (excluding sample preparation) <sup>a</sup>			Unit
	U	Th	Pu	
Titrimetry	20 - 100	50 - 100	5 - 50	mg
Coulometry	2 - 20	-	2 - 10	mg
Gravimetry	2 - 20	0.5 - 1	0.2 - 3	g
X-ray fluorescence	0.1 - 30	0.1 - 30	0.1 - 30	mg
Isotope dilution mass spectrometry	10 - 1000	10 - 1000	1 - 1000	µg
Spectrophotometry	20 - 500	5 - 250	b	µg
Fluorimetry	2 - 500	-	-	ng
Alpha counting	2 - 250	-	0.1 - 1	µg
K-edge	0.3 - 1	-	0.3 - 1	g
Gas mass spectrometry	20	-	-	mg
Thermal ionization mass spectrometry	1 - 1000	10 - 1000	1 - 1000	µg
Gamma spectrometry	0.1 - 1	-	-	
Alpha spectrometry	-	-	0.1 - 1	µg

a) The sample preparation is not considered except for the spiking and chemical treatment in isotope dilution analysis; b) Spectrophotometry direct measurement at 830 nm Pu(VI): 0.1-10 mg

## 6. Summary

Verification analysis of samples taken at large reprocessing plants can be performed off site after shipment of the samples to a specialized laboratory or, more advantageously in terms of cost and timeliness, on site. The latter may be achieved either by using permanently installed equipment which is operated by an inspector or in fully equipped on-site laboratory. Analytical techniques suitable for determining uranium and plutonium isotopic compositions as well as the respective element concentrations, are applied. Experience with a number of these techniques has shown that effective analytical support in safeguarding large reprocessing plants can be provided to the safeguards authorities.

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## NDA SYSTEMS TO SUPPORT NMC&A IN SPENT FUEL REPROCESSING PLANTS

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### ABSTRACT

Reprocessing operations on BNFL's Sellafield site encompass the handling of uranium metal and oxide fuels. A variety of installed NDA systems have been provided to support all aspects of nuclear materials control and accountancy (NMC&A) and safeguards within the Magnox reprocessing complex and the Thermal Oxide Reprocessing Plant (Thorp). These instrument systems range from confirmatory monitoring of fuels whilst in temporary storage and immediately prior to dissolution, in-process hold-up measurements, and waste inventory monitoring through to monitoring systems to support the storage and eventual export of plutonium and uranium products.

Detailed descriptions of a number of instrument systems relating to accountancy and safeguarding of plutonium operations and storage on Thorp are provided. The systems described include the Plutonium Inventory Measurement System (PIMS), used to provide Near Real Time Materials Accountancy (NRTMA) information within the Thorp plutonium finishing area; the Product Can Contents Monitor (PCCM), used to verify can weight measurements and isotopic composition and; the In-Store Plutonium Verification Monitor, used to provide in-situ measurements of plutonium in cans whilst they are in their storage channels. These NDA systems are necessarily combined with other physical security, surveillance and identification arrangements for the handling and storage of plutonium product cans. Nonetheless, they constitute essential elements of the overall control and accountancy arrangements currently implemented at Sellafield.

### INTRODUCTION

BNFL operates a large number of discrete plants on its Sellafield site, ranging from storage and dismantling of irradiated fuel assemblies, through reprocessing operations to eventual product finishing and waste management. An essential component of the control and safeguarding methodologies employed to support these operations is the use of special purpose instrumentation. Generally, this instrumentation is integrated into the fabric of the plant to provide on-line, near real-time information for key aspects of the plant operating conditions. This paper describes some of the instrumentation systems that have been developed by BNFL Instruments to support accountancy and safeguards activities on the Sellafield Thermal Oxide Reprocessing Plant (Thorp). In particular, details are provided of those systems relating to plutonium finishing and subsequent product storage.

The Thorp Product Finishing Line (TPFL) is an area of Thorp dedicated to conversion of plutonium into chemical and physical forms suitable for long term storage. Within TPFL, freshly separated plutonium nitrate solution from the main reprocessing and product purification plant

is converted into plutonium oxide powder using an oxalate precipitation process. The particle size and surface properties of the powder are carefully controlled within a series of filters and furnaces. Blending of the powder is performed to ensure product homogeneity before it is dispensed, via a hopper and screwfeeder arrangement, into a product can. Following further packaging and monitoring (for contamination or faulty welding) the filled product cans are loaded into a buffer store. The buffer store reduces the number of man-entries required into the main long term store enabling the cans to be transferred into long term storage in batches.

As the TPFL plant is dealing with large quantities of separated plutonium it is of obvious interest to the safeguards authorities. The plant operates a Near Real Time Materials Accountancy (NRTMA) system comprising computerised interrogation of on-line instrumentation, including feed tank level systems, weigh stations and a suite of installed NDA instrumentation systems. Three special radiometric instruments developed by BNFL Instruments are used to support the accountancy and safeguarding aspects of this overall finishing and storage process, these being:

Plutonium Inventory Measurement System (PIMS). The PIMS is an installed, non-intrusive NDA measurement system capable of determining the quantity and distribution of plutonium process materials throughout the operating plant. This information provides the operators with a near real time indication of the distribution of plutonium in the plant, allowing early identification of plant abnormalities, such as blockage occurrence, spillage, etc. The PIMS data is also used as an input to the main materials accountancy systems for the plant and for nuclear safety control of the plant operations. For safeguards purposes, the mass estimates produced by the TPFL PIMS are available for monitoring by Euratom.

Product Can Contents Monitor (PCCM). The PCCM provides an accurate, independent, NDA measurement of filled and sealed product cans to verify the declared plutonium mass and isotopic composition. This measurement is in addition to conventional item accountancy involving a gross weight measurement of the plutonium and its containment. The PCCM incorporates data branching to a separate Euratom data logger for independent verification of plant declared accountancy information.

In-Store Pu Verification Monitor. Plutonium will normally be catalogued for plant control and safeguards purposes on entry into and exit from a store using the PCCM described above. However, there remains the necessity to ensure that material is not being diverted from within the store. Modern stores are equipped with multiple layers of containment and surveillance in an attempt to minimise the risk of undisclosed material diversion. Usually material is stored in purpose designed sealed containers which are uniquely identifiable, e.g. through the use of a permanent bar code system. The Thorp In-store Plutonium Verification Monitor meets the requirement to confirm the presence of material within the containers, whilst they are within the storage channels, to guard against unauthorised replacement.

The design and operational features of all these systems are reviewed, demonstrating their potential applicability to a wide range of plutonium storage facilities.

## INSTALLED INSTRUMENT DESIGN PHILOSOPHY

BNFL Instruments have supplied many installed radiometric measurement systems to the BNFL Sellafield site, with over 100 instruments being installed within Thorp alone. These systems incorporate a range of techniques including active and passive neutron counting and both high



and low-resolution gamma spectrometry. BNFL Instruments has developed a modular approach to system design, building confidence in the operation of the individual components and therefore the instrument system as a whole.

This approach has been evolved over many years of successful implementation of such measurement systems and is, wherever possible, based on the use of available, well proven measurement techniques. Where such techniques are inadequate due, for example, to new or more challenging measurement requirements, this approach usually calls for intensive internal development following through to the proving of a new technique in a form entirely compatible with its implementation in key plant instrumentation. In this way the risk of disruptions to plant construction, operation and decommissioning programmes can be minimised.

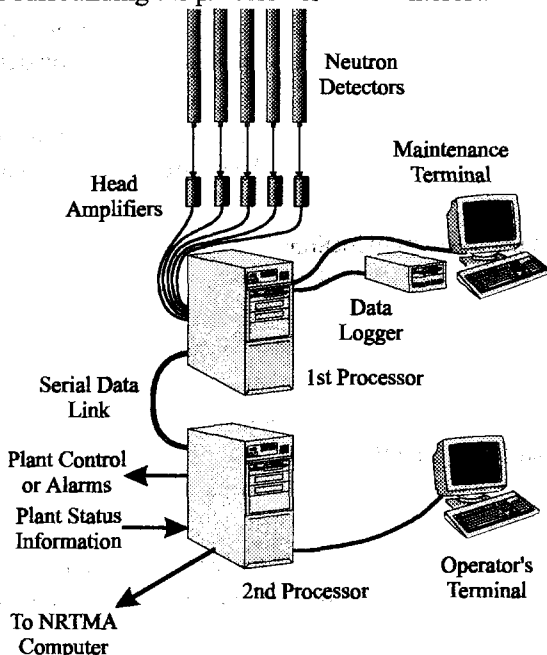
Many years of commissioning and operations support activities by BNFL Instruments' staff, with additional feedback via the close links established with plant managers has provided the company with an extremely detailed knowledge of both plant and systems operation and performance. This feedback has enabled a continuous improvement in system design and operation, ensuring that future generations of systems are simpler to operate in addition to having improved performance, reliability, or operability.

## **THORP PLUTONIUM FINISHING LINE**

### ***Plutonium Inventory Measurement System (PIMS)***

PIMS systems are currently installed within four plutonium finishing lines on the Sellafield complex. These applications include the Magnox plutonium oxide finishing line (FL5), a plutonium metal finishing line (now shut down) and the Thorp product finishing line (TPFL) with the latest system currently being commissioned within the Sellafield Mixed Oxide Fuel Fabrication Plant (SMP).

Figure 1 shows a block diagram of the PIMS. The PIMS comprises a number of neutron proportional counters, generally mounted within polythene moderating modules, positioned at strategic locations throughout the plant. The neutron detector modules are positioned either singly or in arrays surrounding the process vessels of interest.



**Figure 1 - PIMS Block Diagram**

A BNFL Instrument designed head amplifier processes pulses from each detector. The pulse acquisition and data processing electronics comprise a number of standard commercially available electronics modules in addition to proprietary electronics systems.

The system measures the total neutron count rate from each of the neutron detectors and mathematically unfolds the count rate and detector response functions to calculate the neutron emission from each process area.

Achieving optimal accuracy in PIMS operation relies on the positioning of the detectors relative to the plant process line. Typically, the PIMS comprises between 30 and 150 detector modules positioned around the process vessels and gloveboxes throughout the plant. Each vessel / glovebox is modelled, either experimentally or using Monte Carlo N Particle (MCNP) transport codes to determine the optimum locations of the neutron detectors. The criteria for siting the detectors are:

1. to maximise the detection efficiency of the detector (or array) to the process area being monitored;
2. to minimise any system response variation over the process area being monitored, due to different source positions;
3. to minimise the detection efficiency of the detector (array) to adjacent areas of plant;
4. to avoid dead-time problems due to excessive count rates.

In order to meet each of these criteria it is essential that the PIMS is integrated into the plant design at an early stage, enabling the optimum detector locations to be incorporated into the plant design.

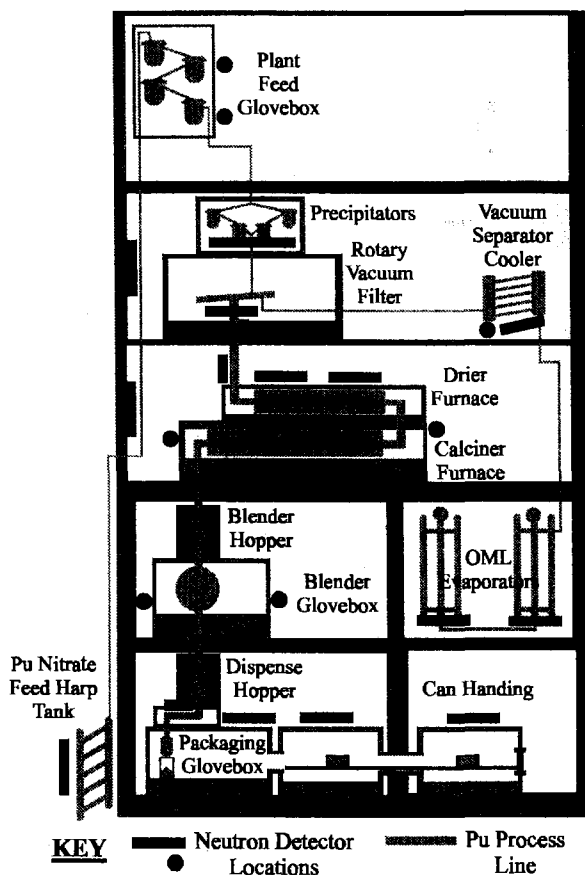
Many of the neutron detectors have been installed within polythene / cadmium moderating modules in order to harden their response to epithermal and fast neutrons (ideally, the module would only be sensitive to neutrons above 1 MeV). The neutrons emitted from fissions and (alpha, n) reactions within the plutonium process material have energies typically greater than 1 MeV. Scattering of these neutrons within the glovebox shielding or concrete structure of the plant quickly reduces the neutron energy. If the PIMS detector package could be designed to only detect those neutrons having energies above 1 MeV the system would be able to reject any neutrons not originating directly from the area being monitored, effectively addressing criterion 3 above. The use of proprietary designed polythene moderating modules enables this fast neutron detection package to be achieved without the need for expensive, unreliable or inefficient neutron detectors (i.e. helium-4 detectors).

Figure 2 shows a simplified sketch of the THORP Plutonium Finishing Line (TPFL) layout including the main process vessels, and identifies the typical positions of the neutron detectors that form the PIMS.

Once the detectors have been positioned in their optimum locations it is necessary to calibrate the system. The calibration involves moving an intense radioisotopic neutron source, usually  $^{252}\text{Cf}$ , throughout the plant process line. The calibration source is ideally located within process vessels in order to best simulate the position of process material during normal plant operations.

The sensitivity of each detector to this "point" source in each of the calibration positions is accurately measured and used to build a calibration matrix, defining the response of each detector to neutron emitting material within each process area being monitored. The calibration matrix is used to interpret the count rate data from the detectors and to estimate the neutron emissions from, and hence the plutonium masses in, each of the areas.

The calculation of neutron emissions uses an iterative weighted least squares technique. The large quantities of data processed for each measurement require the use of sophisticated matrix mathematical procedures.



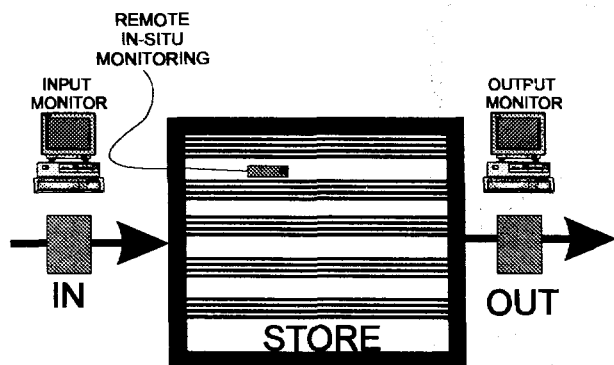
*Figure 2 – Key PIMS Detector Locations within the Thorp Product Finishing Line*

The overall performance of PIMS varies since each system is designed to meet specific plant designs and requirements. The PIMS system installed in FL5 has been fully characterised and employed as an experimental test bed for development of the PIMS technologies. In an extended trial of this system a comparison of the PIMS data against normal book accountancy procedures showed agreement to within 5%.

## THORP PLUTONIUM PRODUCT STORE

### **Background**

Safeguarding a plutonium store requires an accurate account of plutonium within the store and a methodology to demonstrate that plutonium is not being diverted. Such a regime usually comprises a mix of containment and surveillance, nuclear material accountancy and routine inspection by the safeguards authorities. Store operators must report the contents of their store to a good level of accuracy and provide evidence of this to the satisfaction of the safeguards authorities. The information required by safeguards authorities includes form and isotopic composition of material within the area, as well as the quantity of material. In general, plutonium stores can contain a large variety of different materials varying from oxides and metal to solutions and residues (i.e. less than 50% Pu with respect to weight). The Thorp Store, however, only contains plutonium oxide originating from the Thorp reprocessing plant. The necessity of conversion / stabilisation of some or all of this material to allow long-term storage presents additional safeguards challenges, particularly with the physical combination of items originally accounted for separately.



*Figure 3 - A model of NDA systems and methodology of store safeguards*

A simple model of plutonium store safeguards methodology is shown in figure 3. An accurate and verifiable determination of material entering and leaving a store is required as well as a means of identifying the contents of the store. Since the storage canisters are sealed and opaque non-destructive assay (NDA) measurements of the items entering or leaving the store must form an integral part of both the store loading and unloading operation and the safeguards regime.

Inventory taking and verification are activities undertaken by the operators and the safeguards authorities respectively. These activities occur on a regular basis and require considerable inspection effort, a major factor of store safeguards methodology. Verification could, in principle, require the store emptying and re-measurement of a significant proportion of the store contents. This is, however, undesirable in terms of cost, dose uptake and time. In-situ monitoring of the material is clearly preferable, demonstrating the presence of plutonium in the items in the store, and checking this against the declared inventory of the store.

The long term product store on Thorp is the only area covered directly by IAEA safeguards (all other areas being covered by Euratom). The large size, high capacity and high dose rate of the store are not suited to regular manual inspection. The highly automated nature of the store does, however, lend itself to the use of installed instrumentation systems to assist in this verification. Two such instrument systems have been provided for Thorp.

### **Product Can Contents Monitor**

Within TPFL the main accountancy system for filled product cans is a high accuracy accountancy weigh station, coupled with unique bar coding and eddy current labelling of the cans. On route from the accountancy weigh station to the main product store, however, the can passes through a buffer store where it is effectively concealed from view.

In order to confirm that cans have not been switched within the buffer store (from a safeguards point of view) or to confirm that the correct can has been withdrawn by the automated machinery (from an operator's point of view) an accurate, independent NDA measurement is used to verify the declared plutonium mass and isotopic composition of material within the can. Such a measurement is carried out routinely for all material entering and leaving the main product store and is integrated into the operation of the store. The Product Can Contents Monitor uses a combination of time correlated neutron counting and a high resolution gamma spectrometry (HRGS) measurement to determine the plutonium content of a product can. It is an automatic, remote, non-intrusive assay system that is normally installed on the access and egress route to and from a product store so that all cans loaded into or removed from the store are measured.

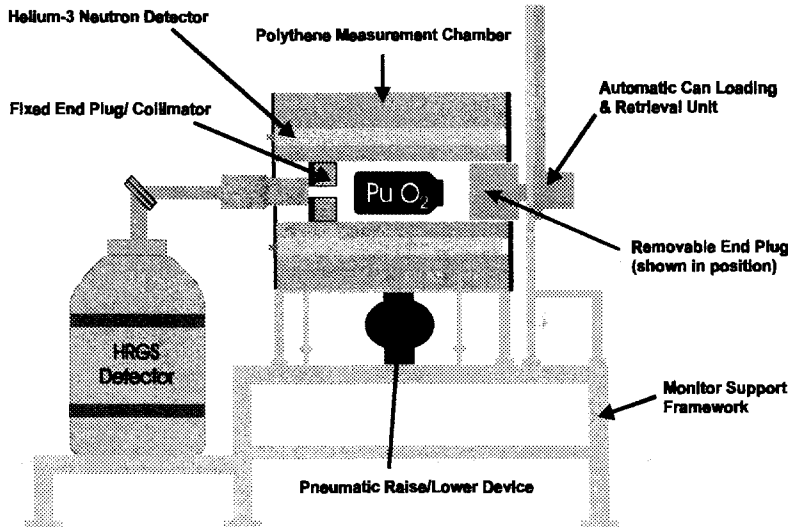


Figure 4 - General Arrangement of the Product Can Contents Monitor

This system uses a cadmium lined polythene measurement chamber containing 18 helium-3 filled neutron detectors. One end of the measurement chamber forms a collimator for the HRGS detector, the other end consists of a removable plug for automatic can loading and unloading that is reproducibly positioned during measurements. Figure 4 shows the general arrangement of the Product Can Contents Monitor.

The measured coincident neutron count rate or “reals” rate is corrected for environmental background, dead time, and, multiplication using the Krick-Ensslin method to determine plutonium-240 equivalent mass. The plutonium isotopic composition, defined as the percentage Pu-240 equivalent content, is calculated using the relative abundances of Pu-238, Pu-240 and Pu-242 determined from the HRGS measurement. The plutonium mass in the product can is then calculated by dividing the plutonium-240 equivalent mass by the HRGS percentage Pu-240 equivalent content.

The design of the THORP product Can Contents Monitor was optimised using a combination of experience with other similar systems, mathematical modelling, an extensive range of commissioning measurements and by the measurement of the initial plutonium product cans. For example, optimisation of the HRGS measurement was achieved by the elimination of measurement biases due to neutron capture events within the germanium detector crystal. Figure 5 shows the results of a check of the HRGS calibration by plotting the measured percentage Pu-240 equivalent content against destructive analysis results. The low irradiation results shown on the figure were derived using laboratory standards to demonstrate the system’s applicability to a wide range of isotopic compositions.

Figure 6 shows the corrected coincident reals neutron count rate for the first two campaigns through THORP against the declared Pu-240 equivalent mass (calculated from mass spectrometry results). This demonstrates the linearity of the calibration, giving additional confidence in the operation of the system.

The example of a Product Can Contents Monitor described above is specific to the Sellafield THORP plant with a pure plutonium oxide product from commercial reactor operations. The system can be configured to meet a wide range of challenges for the measurement of plutonium products.

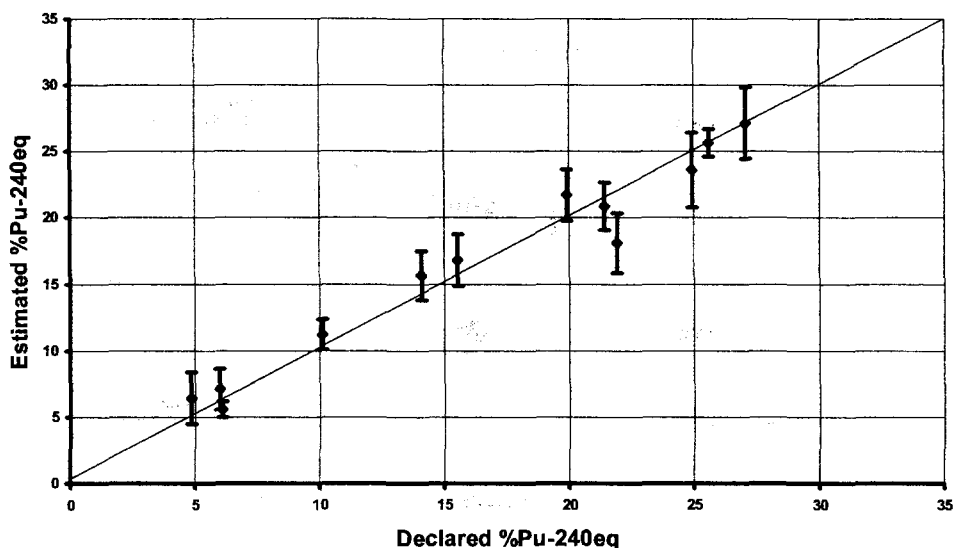


Figure 5 - Calibration check of HRGS %Pu-240 equivalent content measurement

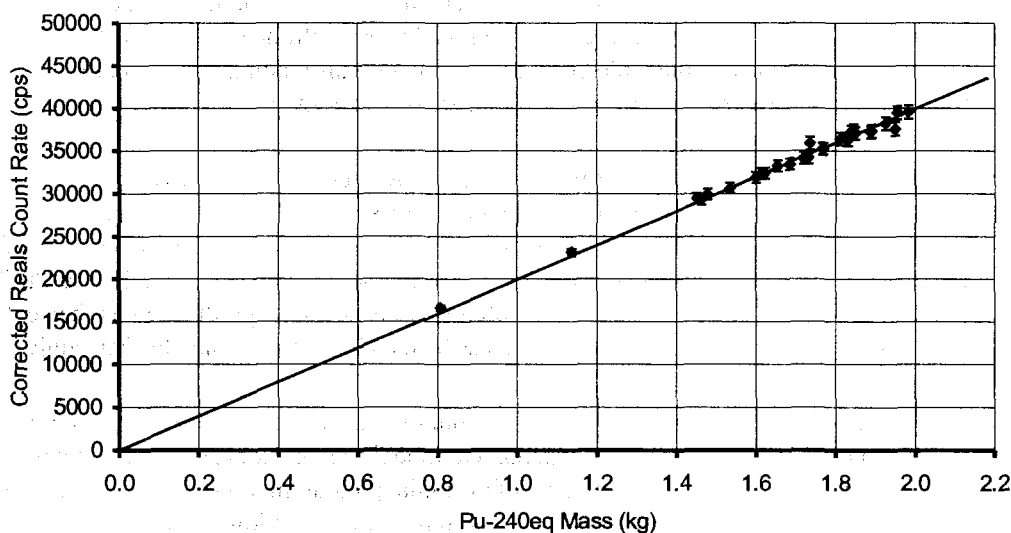


Figure 6 - Calibration check of the Neutron Coincidence Counting system

Future development of Product Can Contents Monitors will provide an improvement in the system accuracy. The Thorp Product Can Contents Monitor plutonium mass measurement uncertainty is better than  $\pm 5\%$  at  $3\sigma$ . In addition, multiplicity counting can be used to correct for multiplication where this can be shown to reliably achieve significant improvements to the measurement accuracy for samples with a range of multiplications and  $(\alpha, n)$  reaction rates particularly impure samples, inconsistent measurement geometries, etc.

#### ***In-Store Plutonium Verification***

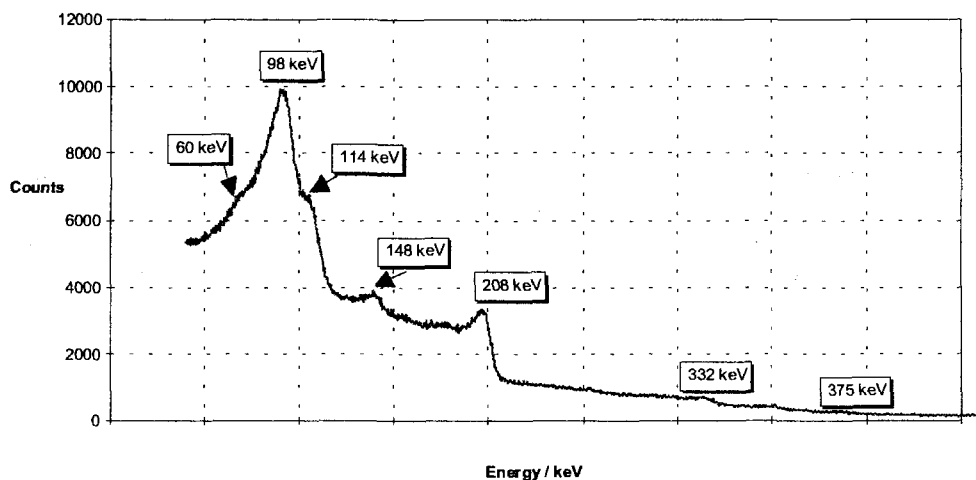
Plutonium will normally be catalogued for plant control and safeguards purposes on entry into and exit from a store using a device similar to the Product Can Contents Monitor. There remains the

necessity to ensure that material is not being diverted from within the store, therefore, modern stores are equipped with multiple layers of containment and surveillance. Usually material is stored in purpose designed sealed containers which are uniquely identifiable, e.g. through the use of a permanent bar code system. The requirement might exist to confirm the presence of material within the containers to guard against unauthorised replacement. This can be done using indirect measurements of temperature, however, the most transparent method is using a direct measurement of the characteristic gamma-ray emissions from each container. The measurement challenges posed using this method are the neutron and gamma ray background from other containers, the significant temperatures reached by the containers, the practical aspects of monitoring in a restricted space and time environment, and the requirement to minimise disruption to normal operations in the store.

A prototype system is currently under test on Thorp in collaboration with IAEA and Euratom Safeguards authorities. This system allows an in-situ gamma-ray measurement of plutonium within storage cans and uses a purpose built, compact, room temperature semiconductor detector based on cadmium-zinc-telluride (CdZnTe). The detector has an "on-board" amplifier allowing the analogue signal to be driven along large lengths of cable enabling the detector to be operated remotely. The temperature within the store channels is reduced using forced air cooling systems which allows a detector to operate at a temperature significantly lower than that at the surface of the cans.

The Thorp plutonium store has been designed with a narrow channel beneath the plutonium cans large enough to allow access for a small, purpose built, instrumentation trolley. The CdZnTe detector is mounted on such a trolley allowing direct measurement of individual cans. It is anticipated the combination of this system with a bar-code or eddy current reader will give the safeguard authorities confidence that the recorded listing of can location, identification and contained material is correct. In addition, a temperature sensor is used to record the surface temperature of the product cans for plant control purposes.

Figure 7 shows a typical plutonium spectrum obtained using the prototype system. The 59.5 keV, 148 keV and 208 keV peaks can be easily identified and are characteristic of plutonium of the type produced at Sellafield. The spectrum shape, with the large low energy "tails" is a consequence of



*Figure 7- Plutonium spectrum obtained with the prototype In-Store Verification Monitor*

using a CdZnTe detector which has low charge carrier mobility (particularly for holes) and enhanced trapping and recombination effects within the crystal. CdZnTe detector developments are currently focused on overcoming this limitation, including work on coplanar-grid electrode detectors<sup>3</sup> which have been shown to have improved resolution without the disadvantages of low energy tailing.

Demonstration of this technique on the Thorp store has been completed successfully and the system has been successfully used for inventory verifications. Further developments to improve mechanical handling, system efficiency and energy resolution are being resolved for the future. This approach for in-situ monitoring, because of the robust and compact nature of CdZnTe detectors, is easily transferable to other stores which have been designed to provide a remote monitoring capability.

## SUMMARY

Three instrument systems currently being used on Thorp for safeguards or accountancy purposes have been described. These instruments form part of a family of products that have been designed by BNFL Instruments to meet specific requirements of the operational plants on the Sellafield site. Further development of the systems has progressed since the Thorp plant was constructed allowing BNFL Instruments to offer these instrument systems or techniques into the wider nuclear market.

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## THE ROLE OF ERRORS IN THE MEASUREMENTS PERFORMED AT THE REPROCESSING PLANT HEAD-END FOR MATERIAL ACCOUNTANCY PURPOSES

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### Abstract

Nuclear materials accountancy methods at reprocessing plants are based on measurements performed on input solutions (at the head end) and on products (at the product storage).

One of the most common procedures used in determining the amount of nuclear material contained in solutions consists of first measuring the volume  $V$  and the density  $\rho$  of the solution, and then determining the concentration  $C$  of this material. The mass of nuclear material present in the solution is then obtained as the product of these three quantities:

$$M = V \rho C$$

Volume and density measurements are made on the process line. Concentration measurements are performed on samples taken from the solution in a laboratory.

This presentation will focus specifically on errors generated at the process line in the measurement of volume and density. These errors and their associated uncertainties can be grouped into distinct categories depending on their origin:

- those attributable to measurement instruments,
- those attributable to operational procedures,
- variability in measurement conditions,
- errors in the analysis and interpretation of results.

Possible errors sources, their relative magnitudes, and an error propagation rationale are discussed, with emphasis placed on biases and errors of the last three types (often called systematic errors).

### 1. Accountancy at the head-end of the reprocessing plant

Accountancy at the head-end of the reprocessing plant is particularly important for safeguards, since this is the first point at which nuclear material is measured again after its fabrication, and it is the first point at which plutonium formed in the reactor is measured.

Accountancy measurements are made on the fuel in liquid form (e.g. a nitric solution) after it is dissolved following receipt at the reprocessing facility. The nuclear content (inventory) of the fuel is obtained from measurements of the volume  $V$  and density  $\rho$  of the liquid in the process tank, together with a measure of its plutonium or uranium content (concentration)  $C$ . The mass of the nuclear material in the liquid (plutonium or uranium) is then computed as the product of these three measurements:

$$M = \rho V C$$

Volume and density measurements are made at the tank on the process line by engineers and operators, using instruments and techniques that pertain to classical engineering (scales, manometers, thermometers, etc.). Concentration measurements are performed on samples taken from the solution by destructive analysis methods, by chemists, in a laboratory using "chemical" techniques.

In spite of its apparent simplicity, this procedure is quite complex because measurements (i) are performed on liquids that are not directly accessible and (ii) involve many steps, including the use of a variety of measurement techniques applied under numerous operating constraints. Temperature variations can have a significant effect on measurement results because the liquids and the systems used to measure them are both highly sensitive to small changes in temperature. Errors are also generated during the solution sampling process, a step required for obtaining the samples for chemical analysis. All of these factors and others affect measurement results, either in the form of increased error or increased uncertainty.

The interpretation measurement results obtained under these conditions is correspondingly complex, and delicate analyses may be required to detect the small differences that are significant for accountability purposes. Computations typically involve a substantial amount of data processing and statistical evaluation based upon appropriate physical models.

All of the factors involved in the collection and analysis of inventory data are, in turn, reflected in material balances, either in the form of biases (nonzero differences) or in the form of increased uncertainty. The nonzero difference is called "Material Unaccounted For (MUF)" in the international safeguards community or "Inventory Difference (ID)" in other environments. It is a matter of particular concern when this difference is large relative to the estimated uncertainty.

These errors and their associated uncertainties can be grouped into distinct categories depending on their origin:

- those attributable to measurement instruments
- those attributable to operational procedures
- variability in measurement conditions
- errors in the analysis and interpretation of results

Uncertainties of the last three types are generally of a systematic nature, and therefore in some sense "more dangerous" than measurement errors because they do not compensate each other nor self-compensate over the long periods. In this paper, we focus on the "systematic" effects of measurement activities at the process line.

## 2. Effect of data uncertainty on the Material Balance

Material Balance evaluations at bulk handling nuclear facilities are based on measured or estimated data, both of which are subject to uncertainty. For this reason, the Material Balance of these facilities never closes to zero. Errors in procedure and variations in measurement conditions, such as those identified in the previous section, all contribute to the Material Balance in the form of increased bias or increased uncertainty.

Evaluating the Material Balance at a reprocessing plant is complicated not only by factors identified in the previous section, but also by the fact that the material is rarely accessible. A reprocessing plant is such a vast and complex structure that small amounts of material may easily remain hidden and go unnoticed by the operator (e.g.: plated on metal surfaces, ....). However, experience has shown that it is possible to make a very good accountancy at a reprocessing plant, provided that the operator is fully aware of the possible sources of uncertainty and is either capable of eliminating them or, otherwise taking them appropriately into account.

It is possible to reduce or control uncertainties in a number of ways, for example, by: (i) improving the quality of the measurement instruments, (ii) improving the knowledge and the control of the process, (iii) improving the measurement procedures and (iv) improving the data interpretation models. It is, for example, important to stabilize or standardize measurement conditions as much as possible to ensure that measurements made at different times are in fact comparable. The use of computerized systems can help to eliminate or reduce procedural errors. In some cases, appropriate models can be used to compensate for differences that cannot be completely controlled (e.g., temperature variations). In any event, the goal from the accountability perspective is to obtain accurate inventory difference estimates and to control, insofar as possible, the factors that contribute to the uncertainty of these estimates.

### **3. Why it is important to know the uncertainty sources**

Since the material balance exercise does not, in principle, yield a material "balance", one might be tempted to conclude that the operator should be content with whatever result(s) he obtains. This conclusion would however be naively dangerous and if this were indeed the case, there would be no basis for an accountability program. Indeed it is essential to evaluate the contribution to overall uncertainty of the various factors which affect the material balance calculation. A measure of total uncertainty is essential for (i) evaluating what magnitude of difference is significant relative to the capability of the measurement process, and (ii) identifying components of the process that can be improved if uncertainty estimates prove to be too large.

It is clear that, in addition to measurement uncertainties, there are a large number of other potential causes for a large or increased MUF/ID for a given Material Balance. Examples include:

- Human error in preparing the Material Balance, such as
  - forgetting to record one or more accountancy data
  - recording the same datum twice
  - mistaking one datum for another
  - making the wrong measurement
  - misreading the measurement instrument
  - incorrectly transcribing the data
- Intentional falsification of the Material Balance (prompted, for example, by the desire to cover operational or management errors, illicit traffic of material, sabotage), such as:
  - failing to record some data
  - repeatedly recording the same data
  - recording data for non-existent measurements
  - recording deliberately falsified data
- Loss of control of the material by the operator, such as:
  - theft
  - un-noticed removal or addition of materials
  - inadvertent or unreported loss of materials

- unawareness of the whereabouts of some material (e.g. material hidden in the process line, or material forgotten in a little used store).

This list is non exhaustive, but it does illustrate that it is in the best interest of the operator to know whether the MUF(ID) of his Material Balance is caused by “naturally” occurring (random) measurement errors or is attributable to negligence or wrong doing of his personnel.

Unfortunately, there are no methods which allow the operator to discriminate with complete certainty whether a certain MUF/ID result is attributable to measurement errors or is due to other causes. The only recourse is to predict the expected value and uncertainty range for the MUF/ID when the process is in control and variation is due entirely to random measurement error. Fortunately, there do exist good methods /1/ for predicting the range in which the MUF/ID generated by a certain measurement system is expected to fall. These methods are based on a statistical analysis of all error components known to have a potentially significant effect on the measurement process /2/. With knowledge of the characteristics of the measurement system used by the operator, a statistical analysis can be used to predict the expected probability distribution of the MUF(ID) for any given Material Balance. The variance (or standard deviation) obtained from this distribution can then used to define acceptance criteria for the MUF(ID): a potential problem is indicated if the MUF(ID) exceeds the established acceptance limit. These acceptance criteria can also be used to judge the “quality” of the various measurement procedures and, with a view toward improving process management, optimizing the measurement process.

Because there are no methods which allow the operator to discriminate with complete certainty whether a certain MUF (ID) result is attributable to measurement errors or is due to other causes, Safeguards Authorities generally employ additional means to understand whether or not the MUF(ID) declared by the operator is justified entirely by measurement errors. These means include:

- Verification of the operator’s book accountancy values
- Physical verification of the End Inventory and of selected Receipts and Shipments
- Comparison of Receipts with shipper’s declarations and of Shipments with receiver’s declarations.

It should be kept in mind that although these verification procedures are apt to detect most of the abnormal events listed above, they do not provide absolute assurance that, after verification, the remaining MUF(ID) is solely due to measurement errors. There is in fact no verification system that can provide absolute assurance because it is necessarily based on measurements, and all measurements are subject to error.

#### 4. Errors in the head-end measurements

This report provides hints on the nature and magnitude of the systematic errors made in the measurements performed at the head-end of the reprocessing plant. The measurement process considered here applies to a tank equipped with “dip tubes” for determining liquid content. With such a system, a measure of the pressure exerted by a column of liquid in the tank is used to determine liquid content. Pressure-based measurement systems are widely used in the nuclear industry and they have often been described in the literature /3/, /4/, /5/, /6/. Although the reader is certainly familiar with the use of a dip tube system to measure volumes, the basic measurement principles are outlined in this paragraph.

The purpose of the measurement process is to determine the content (volume) of liquid in the tank, given a measure of the pressure it exerts at the tip of a submerged dip tube, for use in determining its nuclear content (e.g., mass of Pu). This step requires a calibration which relates

the response of the tank's measurement system (pressure) to the liquid content (volume) of the tank. The calibration-measurement process for determining amounts of nuclear material has several distinct components:

- **Tank Calibration.** The tank used for accountability measurements is "calibrated". Calibration consists of those steps undertaken to determine the relation between some independent measure of tank volume and the level of liquid it contains, as determined from the response (pressure) of the tank's measurement system. The calibration process involves adding carefully measured increments of some calibration liquid to the tank and observing the corresponding measurement system response. The calibration liquid must be some liquid whose density is known or can be determined with sufficient accuracy at all measurement temperatures to produce sufficiently accurate measures of liquid height. After they are properly standardized to compensate for variations in measurement conditions, the data from several calibration runs are used to develop a "calibration curve" that relates liquid height to tank volume.

- **Determination of Process Liquid Volume.** During normal operation, and under conditions similar to those prevailing at the time of calibration, the operator determines the level of liquid in the tank and derives the corresponding volume from the calibration curve. An accurate measure of liquid density is required to make the height determination. If the tank is equipped with two or more dip tubes (of differing length) it is possible to accurately determine (calibrate) their separation (difference in lengths) with a liquid of "known" density. The separation can, in turn, be used to estimate the unknown density of the process liquid.

- **Determination of Nuclear Content.** As indicated in Sec. 1, the volume and density of the process liquid are multiplied by a measure of concentration to determine the amount of nuclear material in the liquid (or tank).

Errors can occur at various points in this measurement process and they may be of various types. Errors may be classified, for example, according to whether they are due to instrumentation, operational or procedural activities, or to the data analytical model. Moreover, they may be random, affecting only single measurements, or they may be more systematic in nature, affecting several measurements (e.g. those involving a particular instrument). Some of the principal errors are identified here.

Instrumentation errors can be divided into those due to instruments:

- at the process line. These include biases in the balance used to weigh the liquid additions during the calibration stage, in the manometer used to measure the liquid content (level) of in the tank, and in the manometer used to measure the density of the (calibration or process) liquid in the tank.

- in the laboratory. These include errors attributable to the densitometer, the mass spectrometer.

Operational or procedural errors can arise from inadequate process control practices or from instrument management practices. Errors due to inadequate process control include

- employing different measurement conditions during calibration than during operation
- making incomplete additions of calibration liquid (water) to the tank
- measuring the tank liquid (height) before the calibration liquid has had sufficient time to drain into the tank

- inadequate homogenization of the tank liquid with respect to concentration and temperature, with the result that samples are not representative

## Session 5

- inadequate data collection, including the recording of incorrect variables and the incomplete recording of required data, or the failure to record vital information (e.g., temperature and density)
- inappropriate handling and storage of samples that results in deterioration between the time of collection and analysis.

Possible errors in instrument management include:

- using different instruments during calibration than during operation
- unnecessarily recalibrating an instrument between the time it is used for calibration and during operation
- operating an instrument under different procedural conditions for calibration than during operation

Errors of a calculational or data analytical nature include:

- failure to take account of temperature variations, both within a set of calibration measurements and between measurements for calibration and operation /4/ /7/. The most important consequence of ignoring temperature variations is a bias in the calculation of liquid heights resulting from the use of an incorrect density.

- neglecting the effect of air buoyancy in the calculation of mass in measurements both for calibration and operation

- neglecting the effect of other factors, such as
  - bubble formation and surface tension at the tip of the dip tube,
  - the weight of the air column in the pneumatic lines between the manometer and the tip of the dip tube /8/, and differences in the weight of the air column between calibration and operation

- flow resistance in the pneumatic lines

- failure to develop an adequate calibration model. An inadequate calibration model can result in

- inaccurate volume determinations

- incorrect estimates of variability for volume determinations. In particular, if run-to-run variations in a set of calibration data are not properly modeled, the resulting variability estimates may seriously underestimate the true volume measurement variability /9/.

The errors that affect individual measurements must either be eliminated procedurally, or their effects must be quantified and included in the overall MUF/ID determination for the facility. This propagation exercise should take possible covariances or correlations into account (when positive, these covariances have the effect of reducing error estimates relative to those obtained when covariances are ignored). Although apparently simple, propagation models turn out to be rather complicated. First, a large number of individual measurements are involved, each of which can be quite complex. Moreover, the use patterns for the various instruments over a period of time may be quite complicated, resulting in many possible combinations of "systematic" effects. It is therefore important to determine the relative magnitudes of the errors that prove to be significant in a particular situation, and it is important to have good computer models to perform the necessary calculations.

### **5. Relevance of possible errors**

The relative magnitudes of the errors in the volume measurement process strictly depend on the layout of the plant, the measurement instruments, the measurement and process control procedures, and variations in ambient measurement conditions. Nevertheless it is possible to make some general comparative remarks about the various types of errors that affect the measurement process.

**Random errors.** Random errors due to variations in individual measurements are mainly of instrumental origin, and they tend to compensate throughout a series of measurements (although their effect variability may not). The precision of modern instruments (scales, manometers, densitometers and mass-spectrometers) is very good, so errors of this type are generally small relative to those of other types. The variability of measurements (e.g., MUF/ID measurements) when all errors are assumed to be random is the baseline against which the significance of all other errors is evaluated.

**Systematic errors of instruments.** The specific performance of a particular measurement instrument tends to effect a series of measurements in a systematic way. The effect of a systematic error typically appears in the form of a bias. The accuracy of modern instruments is very high. Therefore systematic errors tend to be relatively small, provided that proper calibration, maintenance and operating procedures are maintained.

**Operational errors.** Operational errors can have a very important effect on measurement results, and they are generally not systematic. For this reason, their effect can be very difficult to quantify. Incomplete additions of water during calibration is reflected in an error in the determination of U and Pu that is of the same magnitude as the volume measurement error. The effect of non-representative samples (due for instance to incomplete fuel dissolution or incomplete homogenization of the solution) is also reflected in the determination of U and Pu as an error of the same magnitude as the lack of representativeness, and this error can be very difficult to quantify. Loss of water from the solution during the sample taking operation may increase the concentration in the sample by a significant amount. Poor sample preservation can result in precipitation of nuclear material, with the result that concentration measurements are biased. The magnitude of these errors cannot be estimated *a priori*, but experience has shown that operational errors can have a total effect of at least several tenths of a percent, and have the potential to be much larger. The best way to deal with operational errors is to minimize or eliminate them through the use of good process control procedures and standardized measurement techniques.

**Calculation errors.** Calculation and modelling errors can have a very significant effect on both the reliability and comparability of process measurements. Neglecting the temperature variations during calibration or not using the correct density to determine liquid height are potentially large sources of error. The density of water varies by approximately 0.3% between 20°C and 30°C, so use of a constant value for density can cause errors (biases) of the same magnitude when liquid temperatures actually vary over this range. If uncorrected, errors due to differences in temperature between calibration and operation can be even larger. Neglecting the air buoyancy in weighing operations implies an bias of about 0.12 % in the corresponding mass determination.. Neglecting the weight of the air columns in the pneumatic lines leading to the manometers may imply biases of up to 0.1%, depending on the altitude of the manometers over the solution.

Finally, errors result from the use of an incomplete or inadequate statistical model can lead to unacceptably large errors. If a straight line is fit to the calibration data from a tank which does not have a constant cross-sectional area at all elevations, the fitted model will produce biased volume predictions. The error depends on variation in the cross-sectional area of the tank, but errors in the range of 1%-5% are possible for reasonable process tanks. Moreover, if data from several calibration runs exhibit significant run-to-run variation and this variation is not taken into account, the resulting variance estimates will seriously underestimate true measurement variability.

## Summary and Conclusions

Random measurement errors are the baseline against which to evaluate the significance of a particular MUF/ID result. Other errors, especially of a procedural sort, serve to increase the MUF/ID relative to that expected (zero) when the process is in control and errors are random. Therefore, a (statistically ) significant MUF/ID is an indication of some procedural problem. When a significant MUF/ID is obtained, the operator must initiate an investigation to determine the nature of the problem, such as, for example, an instrument failure, a procedural error, or a diversion, and then take the necessary steps to correct the problem.

A realistic assessment of baseline measurement capability is vital for establishing and maintaining a nuclear materials accountability program. This assessment requires a characterization of the in-plant performance of the various measurement instruments used in the measurement process, together with a good data interpretation model. Variability in measurements due to factors of a procedural or operational nature should be eliminated. If elimination is impossible, the factors should be controlled as much as possible by the use of standardized and systematic operating procedures. Uncontrollable variations in ambient conditions at the time of measurement should be taken into account in the computational model. and that all measurements are made under the same set of operating conditions. The goal is to ensure that all measurements are made under the same set of operating conditions.

A good data analysis model and sound computation procedures are required both to assess baseline capability and to obtain accurate volume estimates. At a minimum, this model should

- employ correct values for liquid density, i.e. employ measurements of the density of the liquid at its measurement temperature for each measurement of liquid height
- include an air buoyancy correction for all determinations of mass
- take account of run-to-run differences in a set of calibration data.

As with operational procedures, standardization procedures and computational models should be standardized whenever possible. A suitable standardization procedure and statistical model for tank calibration and volume measurement are given in /6/ and a forthcoming set of international standards based on /6/. The standardization procedures presented therein have been implemented and are available in a computer code /10/ developed for the International Atomic Energy Agency.

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## CONTRIBUTION TO THE RMTC IN THE FIELD OF TANK CALIBRATION AND MEASUREMENTS – THE TAMSCA LABORATORY

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### Abstract

This paper describes the status of activities currently underway in providing a training facility dedicated to upgrading the methodology within the Russian Federation in the application of mass/volume measurement techniques for the enhancement of safeguards in Radiochemical Plants. The objective is to set up, install and commission a **T**ank **M**eaSurement and **C**alibration laboratory (**TAMSCA**) in IPPE, Obninsk, Russia. The TAMSCA design has been completed and is well advanced, consisting of some basic equipment for e.g.

- 3 tanks of dimension and shape as encountered in radiochemical plants in Russia
- a precision balance for providing accurately weighed increments of liquid
- high level precision pressure measurement sensors
- instrumentation for measuring ambient parameters, temperature, atmospheric pressure, humidity;
- instruments to monitor solution temperature, air flow rates
- capacitance probes
- all instruments to be connected via RS485 serial communication protocols connected to a computer for data acquisition and process control inclusive of the necessary software
- software provision for carrying out dedicated exercises and demonstrations for process monitoring as well as for tank calibrations
- dedicated portable measurement equipment inclusive of software and computer.

In order to familiarise IPPE personnel with TAMSCA three separate training courses have already been given to IPPE personnel at Ispra in the TAME (Tank Measurement) facility utilised for training of inspectors from the IAEA and Euratom Safeguards Directorate.

### Introduction

The design of the Russian Methodological and Training Centre (RMTC) on nuclear material control and accounting (NMC &A) is being developed in accordance with Minatom Order No 244 of July 10, 1995. The RMTC is being established for training of personnel from the various Russian and CIS nuclear facilities organisations in the control and accountancy methods utilised in Euratom and the IAEA, and in addition in the R & D of methodological approaches.

The European Commission is coordinating a TACIS project oriented to establish in the RMTC a system for Nuclear Safeguards and Nuclear Material and Accountancy (NMC&A). The RMTC is located in Obninsk State Scientific Centre, Institute of Physics and Power Engineering (IPPE).

Two locations have been identified for the siting of the RMTC namely, the 4th floor of the Physical and Engineering Building (PEB) and the Experimental Nuclear Safety Building (ENSB). Under this project equipment and support will be provided in a number of areas, namely containment and surveillance, training, passive/active neutron assays and mass/volume methodologies. For the latter a mass/volume measurement laboratory i.e a Tank Measurements and Calibration Laboratory (TAMSCA) is being set-up in IPPE, Obninsk. The goal is to upgrade the methodology within the Russian Federation in the application of mass/volume measurement techniques and render a facility suitably adapted to carrying out training courses with specific orientation for the nuclear inspectors and operators of nuclear facilities for nuclear accountancy and control.

### **The TAMSCA Laboratory**

The TAMSCA Mass/Volume Laboratory is located on the 4<sup>th</sup> floor of the Physical and Engineering Building (PEB) inclusive of administration rooms, working rooms and classes for training without nuclear materials, i.e. basic course work, computerization of nuclear materials accountancy, weighing balances and mass/volume methodology. The hardware and operations of TAMSCA are carried out in a room 19.2 square meters with a ceiling height of 3m.

The TAMSCA Laboratory for measurements of solutions mass and volume is intended for :

- the preparation and carrying out of training courses for specialists in MINATOM facilities and for Russian inspectors in GAN on the topic of solutions mass and volume control techniques utilised in radiochemical facilities.
- carrying out scientific and technical support for implementation of the techniques of solution mass and volume control at MINATOM facilities
- the development and testing of new technical tools applied for the nuclear materials solution measurements based on dip tube technology.

The work identified for the laboratory concerns:

- the preparation of training courses, including working materials and necessary equipment
- the organisation of the training courses for vessel calibrations, taking into account different operating parameters, employing similar size or scale models of typical plant vessels and using simulants of representative solution, either distillate product or easily soluble non-toxic salt providing rather high solution density.
- Training courses on data treatment including evaluation with modern software application packages.
- Enhancing the methodological approach for carrying out on-site inspections
- Examining the possibility for adaptation of measurement techniques and implementation particularly on the scientific and technological impact involved in upgrading measurement methods in existing nuclear facilities.

### **TAMSCA Design**

The design of the TAMSCA laboratory is analogous to the mass/volume laboratory existing in the "Mini-TAME " (Tank Measurement) laboratory at the JRC Ispra site of the European Commission in Italy. This laboratory has been operational for a number of years carrying out training courses for both Euratom and IAEA inspectors. In addition R & D work carried out in association with the larger 1:1 scale facility namely "TAME," has resulted in the development of various monitoring equipment for inspectors use under diverse application conditions, for example,

portable compact equipment for quick checking on pressure measurement readings and unattended long term monitoring systems. Such up-to-date modern technological equipment described later will be incorporated in to the TAMSCA facility.

A contract has been setup to cover the installation work for TAMSCA and the NDA laboratories in the PEB and the ENSB locations respectively. These laboratories will be equipped with instrumentation purchased under a call for tender according to the TACIS rules and regulations and installed according to the designs elaborated by the JRC in conjunction with IPPE. The contract established IPPE to:

- elaborate the designs and technical requirements necessary for reconstruction of the existing facilities (PEB and ENSB existing facilities) which will house the above two laboratories.
- carry out all the necessary modifications to the PEB and ENSB locations (structural, mechanical and electrical) in relation to the approved designs.

### **Project Definition**

The proposed work was divided into the following phases.

#### **PHASE I:**

- completion of the final design and technical requirements for the reconstruction of the facility housing the TAMSCA laboratory (room 409 of the PEB location), including the design of the 3 tanks to be constructed with specifications.
- completion of the final design and technical requirements for the reconstruction of the facility in ENSB in order to satisfy the needs of the NDA laboratory (ENSB location).

#### **PHASE II:**

(i) reconstruction, modification and preparation of the PEB facility to house the TAMSCA laboratory as defined in PHASE I and in relation to the needs of the mass/volume laboratory specified by the JRC in the call for tender of equipment to be purchased in order to set-up the laboratory. In particular this phase included:

- fabrication of 3 tanks (cylindrical, annular and tubular vessels) to be utilised in the Mass/Volume laboratory according to the designs and layout elaborated and specified in Phase I of the project.
- the mechanical installation of the hydraulic and compressed air supply lines in the laboratory.
- the electrical supply for conventional equipment, instrumentation and informatic lines.

(ii) reconstruction, modification and preparation of the facility to house the NDA laboratory in ENSB. In particular this operational phase shall include:

- Equipment for nuclear materials storage.
- Ventilation System, nuclear materials storage area
- Alarm System
- Communication System
- Fire Warning
- Security Alarm System of NDA Laboratory
- Power Supply Equipment
- Equipment of Lighting System

### **PHASE III:**

- Commissioning and acceptance tests of both the TAMSCA and NDA laboratories of RMTTC to meet with JRC approval.

### **Mass/Volume Laboratory Equipment**

The laboratory contains the following equipment:

- rack mounted water storage tank of 140 litre volume with level-meter;
- metering liquid tank of 15 litre volume with liquid level meter sensor to place on the electronic balances.
- three testing tanks of up to 1.2 m height on the moving platform. The first tank consists of glass tubes of 100 mm inner diameter each and of 30 litre volume; the second cylindrical stainless steel tank of 400 mm diameter and 100 litre volume. A third circular cone shaped stainless steel-tank of 700 mm outer diameter, 150 mm of circle width, 100 litre volume . These tanks are shown schematically in Figure 1.

The tanks are connected by stainless steel pipes and soft polyethylene tubing with both computer controlled valves and manual controlled valves. A pump is also included in the combined hydraulic system to transfer water from the testing tank and external source to the water storage tank. The general system design has been specified by the JRC Ispra. Also included in the laboratory set-up are:

- gas-air supply system which includes the nitrogen cylinder connected by pressure and gas flow regulators with valves;
- rack-mounted devices for flow rate, pressure, gas mass and liquid level control;
- data acquisition via the computer;
- table with computer and electronic unit and printer. Total power consumption does not exceed 4 kW.

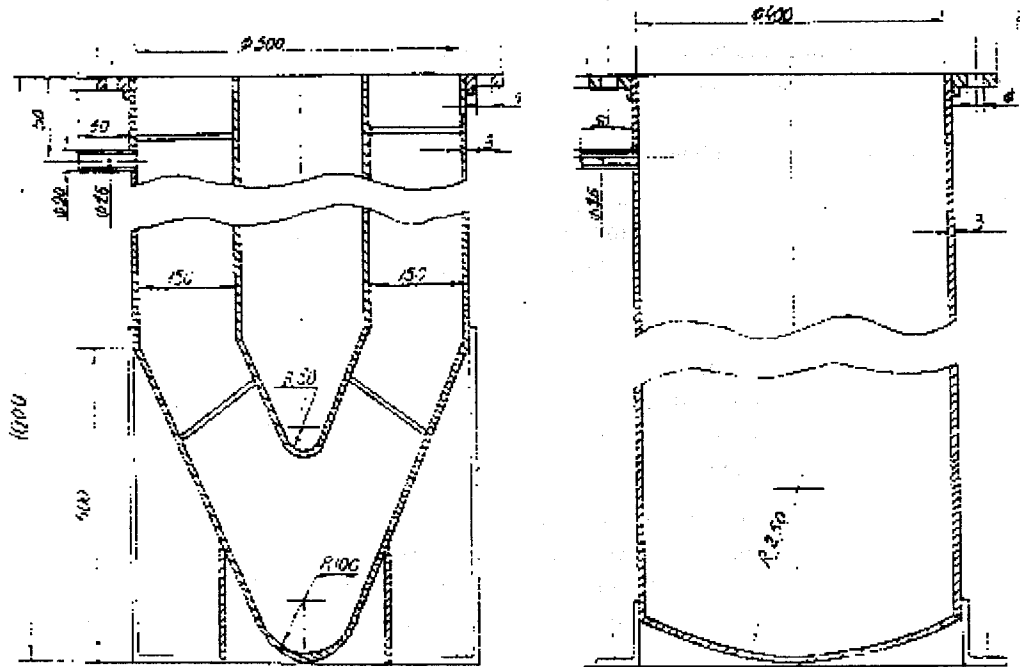
The laboratory will be provided with a power supply board of 6 kW power consumption with circuit-breaker on 380 V and circuit-breakers on 220 V.

Four Euro-standard and two ordinary socket-outlets and four Euro-standard socket-outlets multiplier for the equipment rack to be provided. For the balances three Euro-standard and two ordinary socket-outlets and three Euro-standard socket-outlet multipliers on each table.

### **Level Measurement Unit**

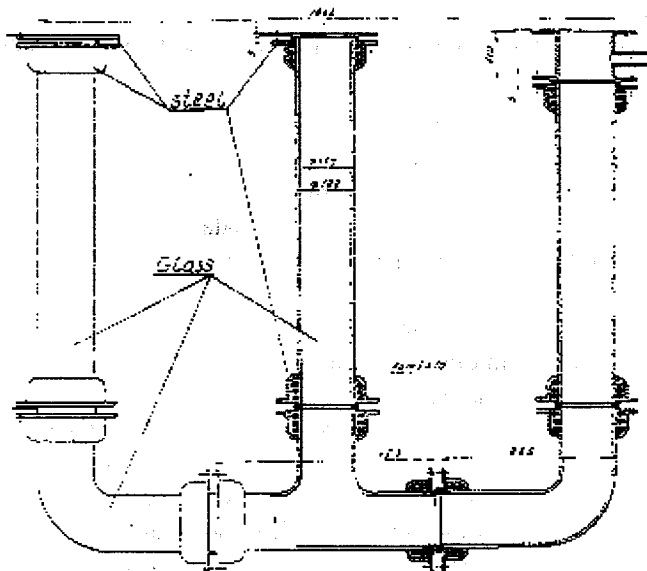
The Level Measurement Unit (LMU) shown in Figure 2 is a complete measurement station dedicated for a new installation with dip-tube technology.

The complete unit comprises very accurate pressure transducers (0.01% full scale), air flowmeters and RS485 input/output protocol. Several of these units can be connected together. The application software runs under Windows 95 and Windows NT and is dedicated for calibrations and monitoring of tanks. During calibrations for example the weight values are corrected according to buoyancy calculations. Results are presented in graphical and tabular form and are also available for further data analysis. Quick verification checks are also performed on the hardware, whilst software is protected by an electronic key.



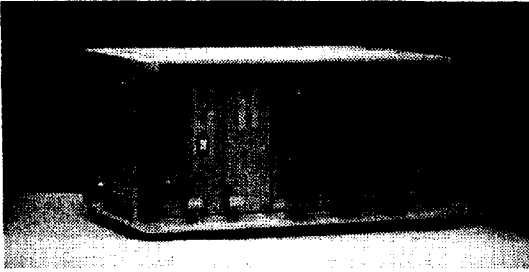
**Circular Cone Steel Tank**  
Volume 105 litres

**Cylindrical Steel Tank**  
Volume 115 litres



**Glass Tube Tank, Volume 41 litres**

Figure 1. Schematics of the Three Tanks Installed in TAMSCA



*Figure 2. Level  
Measurement Unit*

### **Conclusions**

The laboratory is functional for demonstration purposes and undergoing commissioning verification as and when the remaining instrumentation is delivered. Continued training of IPPE personnel is planned, the next course foreseen for January 1999.



## MEASUREMENT INSTRUMENTS DEVELOPED FOR ATTENDED AND UNATTENDED TANK MONITORING AND CALIBRATIONS

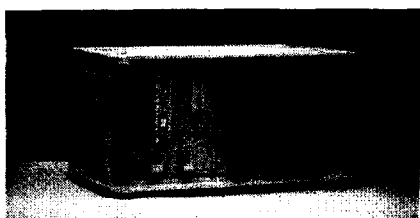
B. A. Hunt, D. Landat, M. Caviglia and L. Silvapestana

European Commission JRC, Ispra, Italy

### Abstract

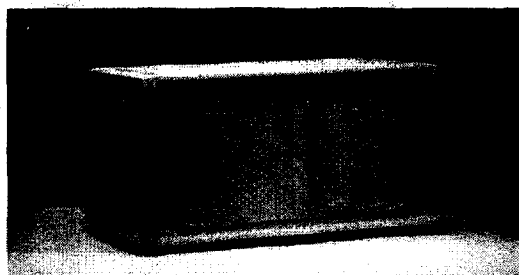
Radiochemical facilities invariably have a number of tanks handling process liquids, which from both the operator's and inspector's side have to be monitored for process control and product quality as well as for nuclear material accountancy. The techniques used for monitoring liquid level in tanks can vary from installation to installation and indeed between countries, however one common aspect is the fact that each tank must be initially calibrated. Modern instruments having serial communication protocols render the systems to be easily linked via a computer for data acquisition and analysis. Software application packages are designed to collect such data either during a calibration exercise of a tank, or during normal plant operations. Systems have been developed and tested at Ispra with the goal of providing for the inspectorates, portable measurement systems, either for short term verification of operator's readings or for long term unattended monitoring.

### *Level Measurement Unit (LMU)*



*Front View*

### *Rear View Pressure and Electrical Connections*



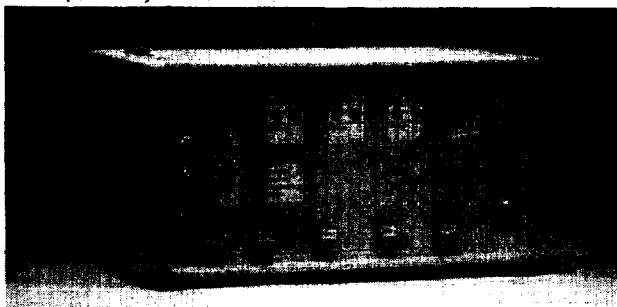


A technique most commonly applied in nuclear installations is based on the classical pressure measurement readings of air flowing through pipes inserted to various depths in tanks. A demonstration of a portable pressure measurement system and applied software is given, similar to that which will be installed in the TAMSCA facility of the RMTC, IPPE in Obninsk. Although retrofitting of equipment may not always be a practical alternative in nuclear installations, it may be possible to adapt the instrument's signal to be registered and elaborated via similarly applied software. The overall scope will be to increase the methodological approach in the enhancement of safeguards as applied to the treatment and bulk handling of liquids.

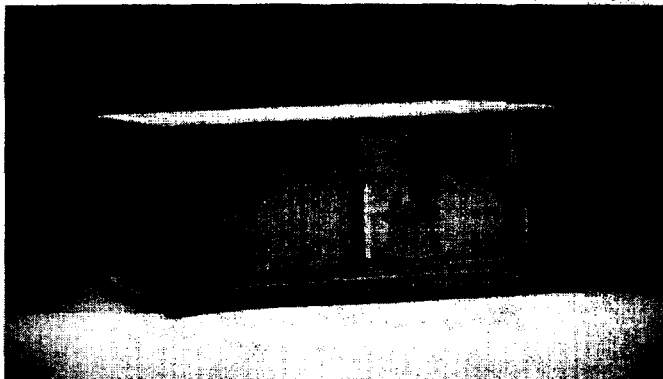
*Volume Long Term Monitoring Unit (VLTM)*



*Portable Pressure Measurement Device (PPMD)*



*Unattended Volume Measurement System (UVMS)*



**Summary of the Main Characteristics of the Instruments and Software Packages Developed at the JRC Ispra**

<b>HARDWARE:</b> Level Measurement Unit	Complete measurement station for new installation with Dip-Tube technology. A complete unit comprising very accurate pressure transducers (0.01%0 air flowmeters and a Pt100 gauge for the temperature measurement. Use of the RS485 I/O protocol. Several units can be connected together.
<b>HARDWARE:</b> Portable Pressure Measurement Device (PPMD)	Modular pressure measurement device very simple to install. Up to 4 pressure transducers per unit. Several units can be connected together. Fast data acquisition system, with up to 8 acquisitions per second. Closed doors on the front and rear panels to avoid unintentional events.
<b>HARDWARE:</b> Unattended Volume Measurement System (UVMS)	Tamper proof unit dedicated for long term monitoring. The UVMS is a typical inspector unit which comprises the same characteristics and advantages as the PPMD and the VLTM. The front and rear doors can be sealed.
<b>SOFTWARE:</b> Volume Calibration and Monitoring (VOLCAM)	This application software runs under Windows 95 and Windows NT and is dedicated to the calibrations of a tank, yet is capable to simultaneously carry out monitoring campaigns on 10 tanks. All measured data are reported to a reference temperature. During calibration weight values are corrected according to the buoyancy calculation. Results are presented in both graphical and tabular form and readily available for further data analysis. VOLCAM also carries out verification checks on the hardware. The software has electronic key protection
<b>SOFTWARE:</b> Volume Long Term Monitoring (VLTM)	This is a user friendly application software to be used with the VLTM and UVMS units. This software is only used to configure the VLTM and to download the data after a measurement campaign. Results are presented and compatible with readily available commercial application tools (e.g. Excel)

**REMOTE DEVICE TO DETECT THE NEUTRONS AND GAMMA-RAY ACTIVITY**

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**ABSTRACT**

Two options of the device (neutron and gamma) were developed to detect and register in memory the results of measurements. Devices are battery powered and could perform monitoring for the long time which was achieved by very low energy consumption. For the gamma option of the device this level is approximately equal to 1 mA and not less then 1 year of the remote monitoring without batteries changing. There is build in processor and 512 Kb memory so the device is capable to put into the memory not only results of the measurements but the date also and perform the necessary processing of the information The information could be transferred out throw infrared interface with RS232 protocol.. Devices have high level of the IP protection. The gamma device could be used under water and for the neutron device the protection is IP-65. The devices could be used for remote monitoring under heavy climatic conditions where external power is not available. It could be used for example as radiation seals at some kind of nuclear materials storage or the possible routes of the transfer of materials or some other way.

**The goal of the development and field of implementation**

The goal was to develop monitoring devices with extremely low power consumption capable to detect radioactivity exceeding the background; (gamma and neutrons). The Goal was achieved by using special new technical solutions and modern electronics materials.

The devices could be used as follows:

- Detection of unauthorized movement (removal) of the nuclear material and/or radioactive sources from the facility. As an example it could be long term storage of the materials or something like that. Usage of the devices together with some kind of seals could be considered as dual surveillance measures. The main goal of such a surveillance is to confirm the continuity of knowledge of the previous verification of the materials.
- Monitoring of the radioactivity for different purposes in places with harsh climatic conditions and where the usual power supply is not available.

**Technical features of the device**

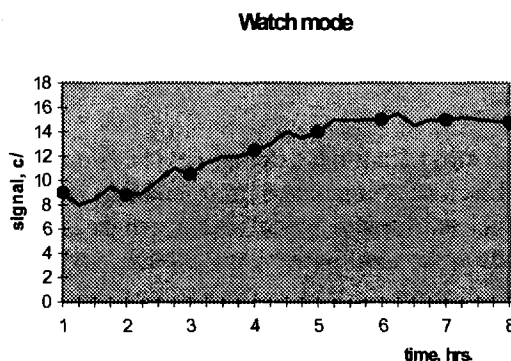
Devices are battery powered and intended to be used for long autonomous functioning. Devices have enclosure with high level of the environmental protection and could be used in wide range of the temperature conditions - from minus 20°C to +60°C and at moisture level up to 100%. The main technical characteristics are as follows:

	<u><math>\gamma</math> device</u>	<u>neutron device</u>
The type of the registered radioactivity	$\gamma$	neutrons
Counters	CTC-6 (GM)	CH-19H (He-3)
Sensitivity	310 (c/s)/(MBq/s)	1.4 c*cm <sup>2</sup> /n
Build in memory	512 Kb	512 Kb
Periodicity of background measurement	1 hour	1 hour
Saved information	month, day, hour, minute, value of the activity	
Output information:		
type of the output channel	optical, infrared	
bout rate	9600 bits/s	
maximum time of information transfer	20 minutes (for 512 Kb )	
Device for reading of information	IBM - compatible PC with RS -232	
Power supply	4 elements LR20, (6 V)	
Time of functioning from one kit of batteries ("DRACELL" type, T°C>0)	≥ 1 year	≥ 3 month
The level of environment protection	IP68	IP65

### Functioning of the device

After initiating the device is functioning the following way:

#### Mode 1. Monitoring (watch mode):



The regime of the device operations is regulated by the timer. Once in the hour (00 min 00 s) the device is measuring the activity ( $t=256$  s for gamma and  $t=64$  s for neutron device ). The value is going to the memory and is used to calculate the threshold value ( $bkg + n \cdot \sigma$ ). The threshold value is stored in the special electronic circuits. Further the device is working in "sleeping regime". Only a few circuits are in operation. The main of them is the circuits which performs quick measurement of the activity ( $t=16$  s for gamma and  $t=8$  s for neutron device) and comparison it

with the threshold. Processor during that operation is switched "OFF". If the count rate is less than threshold value than the operation of the quick measurement and comparison to the threshold is repeated and so on. Once in the hour (00 min 00 s) the device is wakened up. All circuits are in operation but only for the time a little bit more than 256 s for gamma and 64 s for neutron device. During this time the measurement and processing of the information by processor is performed. The measured information and data are going to the memory. After that the main circuits of the device are switched "OFF" and device is going to the sleeping mode during which only quick measurements and comparison to the threshold is performed. During such a mode the slow processes (change in the activity during 16s. is less than threshold value) are monitored with periodicity - once per hour (Fig.1). Marked points - the data to memory.

#### Mode 2. Events detection:

Different situation could occur if there is fast change of the activity. (Release of the activity, movement of the guarded material, etc.). Fast change means that the change of the activity during 16 s (8 s for neutron device) is more than the threshold value.

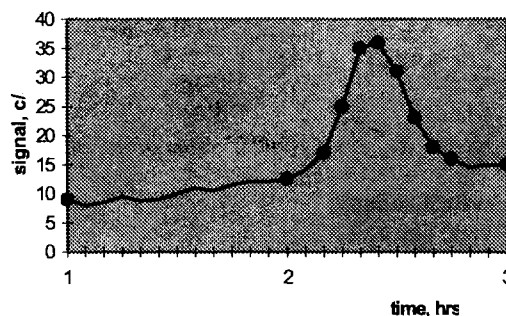
**Events detection**

Fig. 2

In that case the device is functioning the following way. First of all it is wakened up so all circuits are in operation and stay in the operation till the data exceeds the threshold. The main part of the device begins to work in this mode. It is a MICROCOMPUTER, nonvolatile memory and their framing elements. The measurement time is 64 s. The comparison to the threshold is performed as well. If it less of the threshold than processor is simply self disconnected (device moves over to the watch mode), otherwise the information on the value of the signal and time of the event is brought in the nonvolatile memory. The next event

could be registered after passing at least 5 minutes. It is performed to safe power. Nevertheless there is option of the software to register events each 8 s. In such a mode even short changes of the activity could be registered (Fig.2.). Marked points - the data to memory.

Both modes of measurement and processing are characterized by one general particularity: The main part of the time (64 s and 256 s) is used to measure the signal. Only a few ms are used for processing of the information by processor. In that case it becomes possible to use the processor (87C51) in the half sleeping mode when only part of this processor connected with measurements are working. Because of this measures the power consumption in this mode becomes 4-5 mA. Otherwise it would be much more.

**Mode 3. Data transfer:**

Mode of the data transfer is initiated on the request from the external device through the optical channel interface. In this mode the entire block of accumulated information is removed from the memory of the device and transferred through the infrared optical interface to the external PC. After this the device is reinitialized. This mode is the most power consumption mode. The current consumption is - 16 mA.

**Components of the devices**

The device consists of:

- GM counter (CTC-6) for GM device and He<sup>3</sup> counter for neutron device
- Low power consumption high voltage power supply
- Watch dog circuits in sleeping regime (looking for increasing of the activity)
- Timer (10 mA power consumption)
- Processor and memory (512 Kb)
- Infrared interface
- Enclosure (shallow underwater usage)

Node of feeding of counter presents itself a special stabilizing converter of voltage with independent excitement on transistors. Input voltage of node is (4...6)V, output - (380...400)V for GM device and 1400 V for neutron device. Stabilization of output voltage is realized by pulse amplitude stabilization on the primary winding of transformer. Power consumption of this node in sleeping mode is 30-35 mA for GM device and no more than 1 mA for neutron device.

System timer is intended to keep the real-time and hour's marks for switching "ON" the processor for the background measurement. Access to the timer is produced through the consequent

channel I<sup>2</sup>C, consisting of the 2 buses: bus of the data (SDA) and bus of synchronizing (SCL). Emulation of cycles of channel is produced by software. In view of importance of information saved in the timer its feeding are back upped: under normal operation the timer gets power supply from the main source (main batteries). but when there are changing of the batteries it gets power supply from back up elements. Mode of operation is selected such a way that at the presence of the main battery, back up elements are unplugged and are not discharged, i.e. term of their service is sufficiently long (design value is not less than 5 years).

Main elements of the microprocessor unit (MPU) are a one crystal micro-COMPUTER with the internal memory of programs and external nonvolatile memory of 512 Kb. The main part of the time the MPU (as it was indicated above) is switched "OFF" and does not consume energies. After switching "ON" the micro-COMPUTER begins to execute a program from the internal memory.

Infrared interface consist of 2 parts: receiving module on and transferring module. Program of output of data quantifies data, accumulated for a time from past sensing, consecutively removes them in the channel and reinitializes a system with the initial background measurement.

Enclosure for the gamma device was constructed as a tube so it could stand the operation under the shallow water. For the neutron device there is not sense to use it underwater so it was constructed such a way to stand at least under heavy rain and snow. It should be noticed that there is a moderator inside of the enclosure to increase the sensitivity of the device to fast neutrons.

## Results of tests

### Detection of the Pu sample

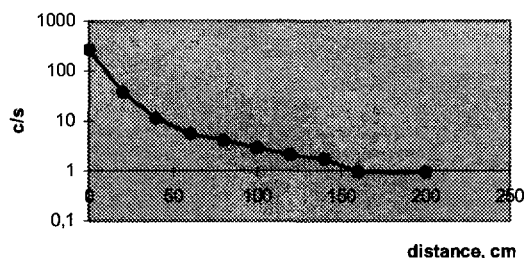


Fig. 3

All technical features of the device including duration of the operating without changing of the batteries very tested under laboratory conditions according to the program of testing. Devices successfully passed all the tests. Because of routine procedures and considerable volume of information the data are not presented on the report. Below there are results of a couple of experiments that show the ability of the devices to detect some specific sources and nuclear materials.

Estimation of the sensitivity of the gamma device "Mouse\_γ" to the detection of the Uranium sample (2.1 g of U<sup>235</sup>, 11.9 g of U<sup>238</sup>). The background count rate was 0.9 c/s. The sample was detected on the distance of 10 cm - count rate = 1.9 c/s. The sensitivity could be increased in next generation of the device using a number of GM tubes used in parallel. Additionally the

### Detection of the neutron source

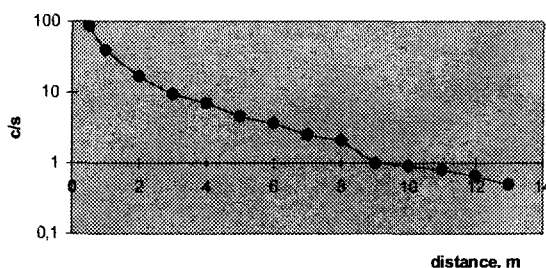


Fig. 4

sensitivity of the device to the plutonium sample (4.5 g) was tested as well. The background count rate was the same - 0.9 c/s. The results are presented in the Fig.3.

The sensitivity of the neutron device to detect the neutron source of the 10<sup>6</sup> n/s was checked as well. The results are presented in the Fig.4. The threshold was installed as 5\*σ but not less as three

neutrons for 8 s. It means that the threshold value was about 0.3 c/s. The distance of the detection was more than 10 meters.

Additionally the sensitivity of the neutron device to detect the plutonium sample of 4.5 g was checked as well. The threshold was installed as above -  $5 \cdot \sigma$  but not less as three neutrons for 8 s. The sample was detected on the distance of 25 cm or less.

### **Conclusion**

The device for monitoring of gamma activity and neutrons with the following advantages was successfully developed and manufactured:

Extremely low power consumption which allow the long time of monitoring (up to one year or even more for GM device).

1. High level of the intellect (build in processor)
2. A big memory to remember results (512 Kb) including the date of events (min, hour, day, month, year)
3. Extremely simple and highly automated mode of operating
4. Infrared interface to pass information to the external computer
5. High level of the IP protection
6. Wide range of the possible application



## NPP FUEL CYCLE AND ASSESSMENT OF POSSIBLE OPTIONS FOR LONG-TERM FUEL SUPPLY

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During a half-a-century period of its development nuclear power has proved its viability and necessity actually in all the developed countries.

Exaggeration of the hazard from new technologies, especially nuclear technologies can be come across very frequently among different polititions and not well-informed part of the society. The formers express this statement in their political speculations due to their unsatisfied political ambitions, the latters express it due to their inertial way of thinking, i.e. everything new and vague is dangerous. Currently there are no alternatives to nuclear power technologies but we cannot develop nuclear power (though we are sure in the necessity of its development) without a comprehensive analysis of its potential capabilities.

Since the middle of 60s, to be more precise after the third Geneva conference (1964), the economic advantages of nuclear power have become obvious.

In 1975 in the European Nuclear Power Conference it was shown that one kilowatt-hour of nuclear electric energy is twice as cheap as one kilowatt-hour of electric power generated with organic fuel.

Today this difference is not as much first of all due to expenditures for NPPs safety, but everywhere, may be with the exeption of the west of USA where power coal is very cheap, the nuclear energy is cheaper than the energy generated with other types of energy resources.

Under the conditions of normal operation if we compare potential capabilities of alternative energy technologies the nuclear power differs in having some ecological advantages, because its consumption of the earth resources is much lower.

Out of the total amount of mineral resources mined in the world 85% fall on the organic fuel and it was a very fast development of power generation in support of industrial development in the end of the last century that resulted in slowing down the Earth entropy decrease and then in its growing, thus indicating the loss of balance in the Earth biosystem.

The purpose of this paper is to present some results of the analysis of the possible options for Russian NPPs fuel supply.

In the classical consideration these are four fuel cycles:

- Uranium cycle based on natural uranium.

This cycle has several economical advantages with the use of "CANDU" type reactors with a heavy-water moderator.

- Uranium cycle based on enriched uranium.

It is the basis for the current and future nuclear power.



- Uranium-thorium fuel cycle with capabilities which are very promising but unfortunately difficult to implement in practice;
- Plutonium - uranium cycle.

In terms of its potential capabilities it is an excellent option, but it is extremely difficult to implement it in practice due to a high activity and toxicity of nuclear materials under recycling.

All these things should be taken into account when developing both short-term and long-term predictive options of future nuclear power development.

It should be stressed that for Russia due to its climatic peculiarities and a vast territory the nuclear power is very crucial for the progressive development.

Any branch of industry is based on this or that raw material.

For nuclear power this material is uranium. And if nuclear power could be developed only in the direction of its peaceful application, i.e. for the electric power generation, everything could be rather clear. The classical fuel cycle could be implemented:

- ore uranium;
- enrichment in  $^{235}\text{U}$ ;
- fabrication of FEs and FSAs;
- NPPs.

It would be followed by the fuel reprocessing at the NPP sufficient capacity (60-80 GWt(e)) with sending the recovered nuclear materials back into the NPP fuel cycle.

Only a rather big radiochemical plant (1-1,5 thousand tons of uranium per year) is capable to provide acceptable economical parameters of the NPP closed fuel cycle. That was proved long ago (in the 70-s) and for the conventional nuclear technologies used and accepted to be used in the nearest future (i.e. fuel application in the form of fuel sub-assemblies, FSA, with a periodical, about once a year, refueling and reprocessing based on the PUREX technology) the most part of expenses falls on the waste management and a relative portion of these expenses goes significantly down when increasing the plant uranium productivity.

An extremely intensive development of military programs on highly-enriched uranium production and Pu build-up in industrial reactors with the following reprocessing of irradiated uranium resulted not only in accumulation of large stockpiles of weapons-grade Pu and U but also to large stockpiles of natural uranium in the form of  $\text{U}_3\text{O}_8$  and metal, meant to be used as fuel for industrial reactors, as well as recovered uranium with a little bit less content of  $^{235}\text{U}$  but quite applicable for the purpose of enrichment and fuel fabrication for NPPs.

The fuel factor cost in the cost of NPP electric energy is mainly determined by three links in the cycle: natural  $\text{U}_3\text{O}_8$  production, isotope separation and fuel fabrication.

As far as natural  $\text{U}_3\text{O}_8$  production is concerned, it should be stressed that in Russia actually there are no resources with high concentration of uranium (more than 1%) so there is no way out, but to develop the ore deposits with the average uranium content of 0.1%-0,2% and to construct large mining and processing plants with a rather expensive mining activity at the level of deep underground horizons and expensive reprocessing with application of the most efficient hydrometallurgical technologies.

The ore uranium price in Russia is significantly higher than the prices on the world market and even predicting the growth of world market prices we can say that the Russian uranium could hardly be competitive with uranium from Canada, Africa, Australia.

No doubt, we need to make certain efforts to develop our own reliable raw base of nuclear power but at the same very time we need to be very reasonable when distributing limited resources.

The uranium isotope separation in Russia is implemented based on currently the most efficient gas-centrifugal technology. This technology needs to be maintained and developed without losing the steady position in selling enriched uranium on the world market.

The gas-centrifugal technology makes it possible to profitably process slag-heaps of isotope-separation plants. These heaps accumulated in the past can meet the demand of Russian NPPs of the up-to-date level of capacity for the next - 10-15 years.

Investigation and analysis of uranium raw base allow the resources to be classified in the following way:

Uranium in the bowels of the earth	The resources are limited with rather high expenses
Uranium in the form of $U_3O_8$ in the storage	Residual stocks are limited
Uranium in the form of $UF_4$ , $UF_6$ , $UO_2$ , $UO_2$ pellets	Stocks are limited in the storage
Uranium in ingots and products	The most convenient for a long-term storage with the least expenses as a strategic stock. Stocks are significant
Slag-heaps accumulated in the past on isotope - separation plant sites	They are used in the uranium enrichment process for nuclear power. Stocks are significant
Weapons-grade uranium	Options of application: - for a long-term storage as a strategic stockpile - for sale abroad The domestic use currently is ineffective due to the availability of sufficient separation capacities and low cost of separation
Uranium reprocessed from industrial reactor spent fuel	It is used in the production of enriched uranium for nuclear power. The stocks are considerable and are replenished in the course of operation of three industrial reactors.
Uranium reprocessed from VVER440, Propulsion units, BN, research reactors spent fuel	Reprocessing is on the limited scale. The accumulated stocks are insignificant. Serious restrictions in application
Uranium in submarine cores subject to disposal	Potential stocks are insignificant

The diagram of NPP fuel supply is given in Fig. 1.

In Russia there are certain stockpiles of weapons-grade Pu in non-dismantled nuclear war-heads, in salts and oxides formed in the result of industrial reactor fuel reprocessing and certain amount of reactor-grade plutonium accumulated in the course of RT-1 operation.

The weapons-grade plutonium is currently used in MOX-fuel of pilot FSA in BN-reactors.

The reactor-grade Pu (with a high background) is supposed to be used in the future when fuel fabrication remotely operated technologies are available.

All the NPP fuel cycle expenditures should be incurred by the nuclear power industry which sells the final product of nuclear fuel-power complex.

However, evidently not only the power industry should be responsibility for the previous history of its development.

For instance, when the production output goes down the operation of excess fuel production capacities results in the growth of expenses, however the value of these expenses must not decrease NPP capabilities to compete successfully on the power market.

Moreover only large NPP capacities which can be achieved at high NPP competitiveness make it possible to implement profitably the closed fuel cycle with the reprocessed reactor-grade U and Pu being involved in it.

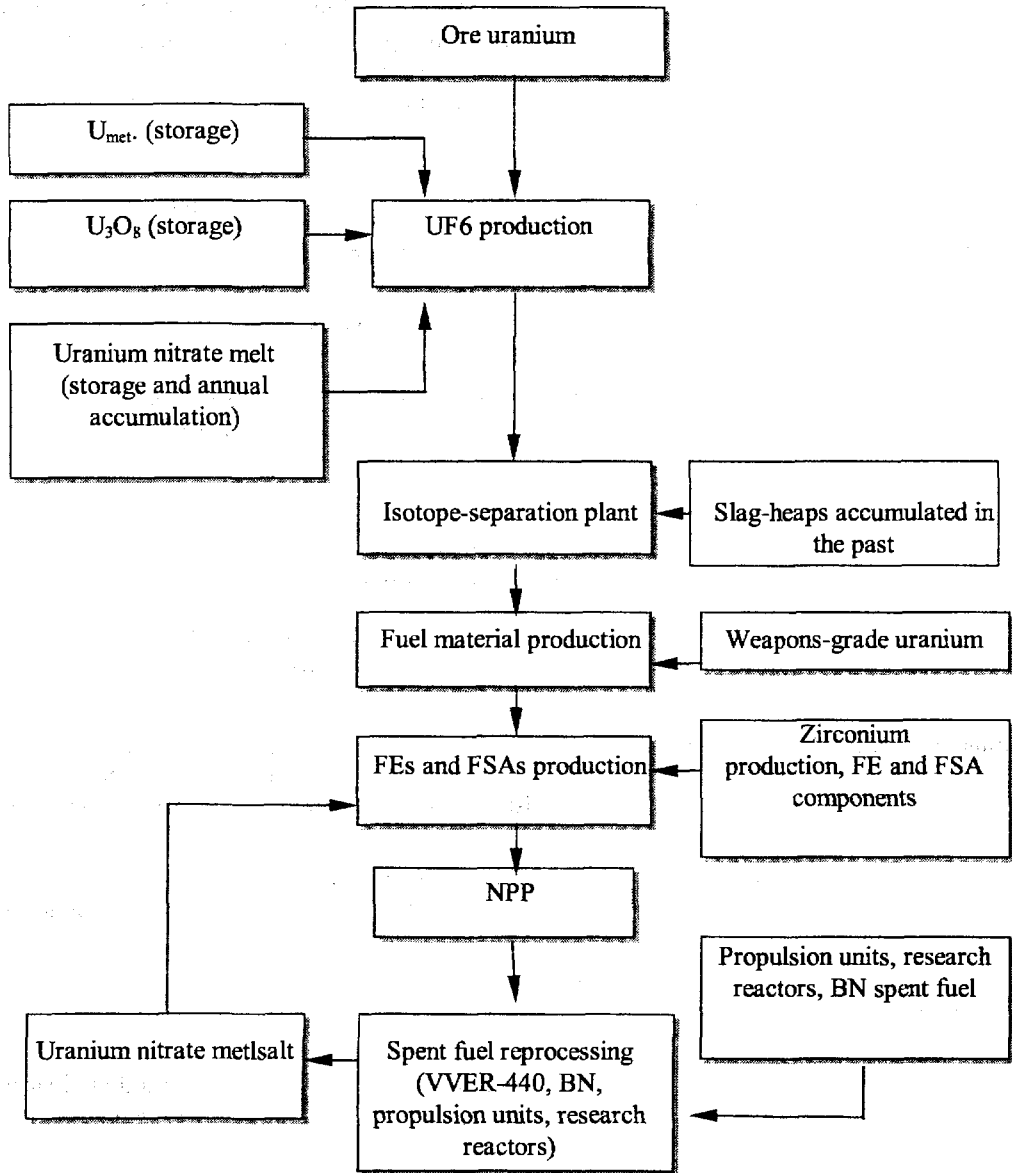


Fig. 1 NPP fuel supply

In view of that the nuclear power of Russia is currently aimed at using the cheapest fuel resources, that is first of all, uranium reprocessed from industrial reactor fuel and slag-heaps accumulated on the past in isotope-separation plant sites. These resources are enough for the Russian large-scale nuclear power to be developed.

In parallel the research, development and design work must be performed on developing safe and efficient technologies for using reprocessed reactor-grade U and Pu. However big investments into the commercial implementation and practical use of these technologies will be justified only at the stable operation (loading) of a rather big radiochemical plant. The RT-2 plant will be of that type (i.e. the reprocessing capacity 1000 - 1500 tons per year which corresponds to the NPP capacity of ~ 60-90 GWT(e)).

We think that the practical realization of NPP closed fuel cycle must be based on complex universal production plants which reprocess all types of fuel and fabricate fuel from various types of raw material. It requires the development of reliable remotely operated technologies at the stages of nuclear material treatment.

Besides, it is extremely important not to allow the equipment to be contaminated with hazardous isotopes above the permissible level as early as at the stage of technology testing.

With a very scarce budget available the NPPs of Russia cannot pay for the construction of a large reprocessing plant which currently costs not less than 2-3 billion dollars. So it is necessary to get investments from the potential foreign customers interested in fuel reprocessing. And for this purpose we need contracts based on the principle: "the customer pays" as it was done in France and Great Britain when constructing large radiochemical plants.

But evidently nowadays it is extremely difficult to realize this principle in Russia.

The NPP closed fuel cycle allows ecological problems of nuclear fuel use to be solved most reliably with implementing the tasks of radioactive waste fractionation and conditioning. But all these problems and tasks can be solved only by the large-scale nuclear power which at the same time must be reliable and economically attractive.

The steady raw material base of the future large-scale nuclear power is provided with a combination of thermal and fast reactors in its structure with the use of both uranium and MOX uranium-plutonium fuel in the closed cycle. In order to create this base certain funds are required. They can be obtained as a result of decrease in energy production cost, fuel cost included.

The use of thorium-uranium fuel cycle requires even more expenses related to the necessity to develop a new raw material base and to apply very expensive remotely operated technologies in all the parts of the cycle where uranium reproduced from thorium is available.

**SYSTEM TO DETECT NUCLEAR MATERIALS BY ACTIVE NEUTRON METHOD**

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**ABSTRACT**

The report presents the results of the development of the system to detect nuclear materials by active neutron method measuring delayed neutrons. As the neutron source the neutron generator was used. The neutron generator was controlled by the system. The detectors were developed on the base of the helium-3 counters. Each detector consist of 6 counters. Using a number of such detectors it is possible to verify materials stored in different geometry. There is an spectrometric scintillator detector in the system which gives an additional functional ability to the system. The system could be used to estimate the nuclear materials in waste, to detect the unauthorized transfer of the nuclear materials, to estimate the material in tubes (e.g.  $UF_6$  at some facilities), etc.

**The goal of the development and field of implementation**

The goal was to develop highly intellectual industrial system to detect fissile materials by active neutron method measuring delayed neutrons. Possible application of the system could be as follows:

- Detection of illegal transfer of shielded fissile materials (e.g. Uranium in container in track)
- Measurements of fissile materials contents in different samples, including different kind of waste (enrichment must be measured by spectrometry).
- Different kind of usage in scientific laboratory for different kind of investigations.

**Main Components of the SYSTEM**

The system consist of:

- **Central station** including industrial PC card, display, counters cards, special cards to get information from the detectors and to sent signals to the neutron generator, power supply modules, software, etc
- **Neutron detectors**
- **Neutron generator**

**PRINCIPLE OF ACTION**

The principle is very simple. The target should be surrounded by the set of neutron detectors to achieve the necessary sensitivity. On command from central station the neutron generator produces the shot of neutrons. The neutrons penetrate the target and cause fission in fissile material in target. As a result of fission there are delayed neutrons which are registered by the neutron detectors. Signals go to the central station for processing and making decisions. The information are displayed on the screen. Example of the decay curve of delayed neutrons registered by the system is presented on Fig. 1.

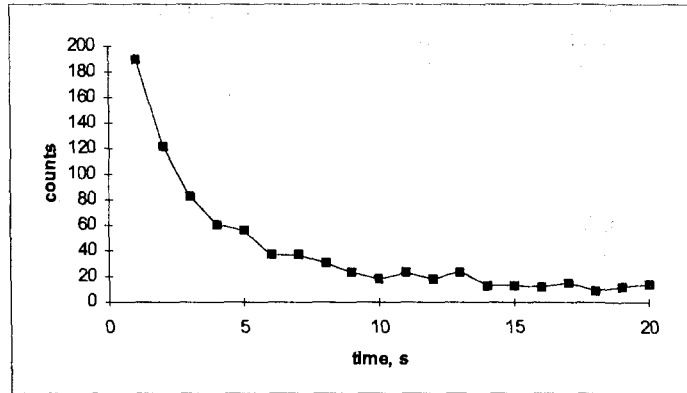


Fig.1

The simplified algorithms of functioning is as follows:

- The signal (command) from Central station to the neutron generator
- The shot of the neutron generator (one or a number depending on the command)
- The pause for time more than live time of the neutrons in the moderator
- Delayed neutrons measurements
- Information proceedings
- Go to stage # 1 if necessary to repeat this cycle as many times as necessary to achieve the required sensitivity of the system in the specific geometry of measurements.

## Technical features of the SYSTEM

### 1. Central Station:

Type of registered activity

(any) detector dependence

Type of input signals:

pulsed signal

Polarity

negative

duration, msec

from 5 to 15

frequency, Hz

from 0 before 100000

amplitude, V

from 1.5 before 15

Amount of input channels (detectors)

up to 12

Channels power:

output voltage, V

from 11 before 15

current on each channel, mA

no more than 80

Amount of output analog signals

4

Ranges of output analog signals, V

(0...+10), (0...+5), (-5...+5)

Amount of output pulsed signals:

1

polarity

free

duration, msec

15 $\pm$ 30%

amplitude, V

15 $\pm$ 30%

## Poster Session

Built-in minicomputer:	IBM-compatible, ISA
Volume of solid state disks SSD1, Kb	512
Volume of solid state disks SSD2, Kb	128
Communications	RS-232
Display	monochrome, 640x400
Control	keyboard AT
Voltage needed	network (170-230 )V, (50-60) Hz
Power ,consumption, Wt	no more than 50
Level of the IP protection	IP32
Dimensions, mm	370x350x330
Mass, kg	no more than 6

### **2. Neutron detector:**

Type of He-3 counters	SNM-18
Number of counters in the detector	6
Type of output information	pulsed sequence
Polarity of output pulses	negative
Voltage of output pulses, V	not less than 2
Duration of output pulses, msec	$5 \pm 10\%$
Sensitivity, neutron*cm <sup>2</sup> /s	82
Voltage of power supply, V	12...16
Current of consumption, mA	no more than 50
Level of protection from surrounding ambiance	IP66 EN 60529
Dimensions, mm	405 x 410 x 120
Mass, kg	15
Length of communication link, m	100

### **3. Neutron generator:**

The type of generator	Pulse Neutron Generator-01 standard
Neutrons per shot	$10^7$ neutrons
Maximum number of shots per s	30

## **Description of the SYSTEM**

### **Central Station**

Central Station is designed for receiving and processing of the information from sensors (detectors) and presenting information for the Operator on the screen of the display. Additionally it sends command signal to the neutron generator to make shots of neutrons.

Central Station is intended for non-stop operation and can be used within the range of temperatures from minus 10°C up to +40°C and at moisture up to 90%.

Central station is based on Micro PC technical means of the Octagon Systems company (USA). This system is based on the IBM PC architecture. It uses a number of controlling and peripheral elements (cards), united by 8-bit ISA-a bus.

Functional scheme of the central station is presented on the Fig. 2.

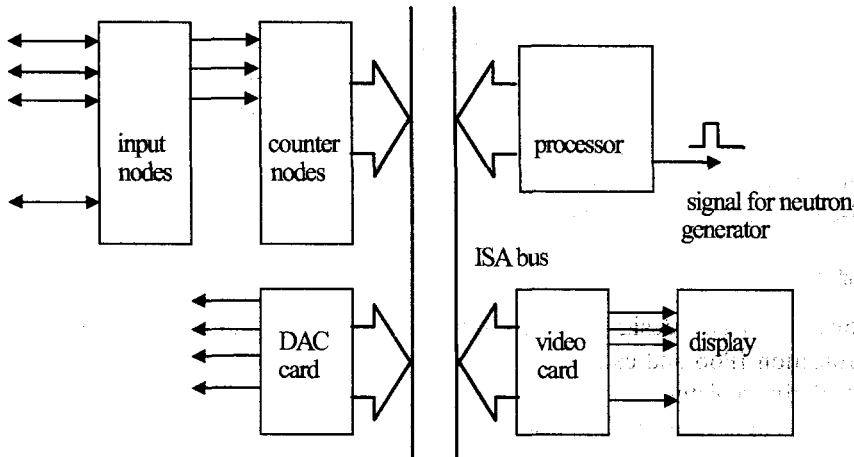


Fig 2.

Information from sensors (detectors) is going through "input card" to the timer-counter Card which is connected to ISA bus and throw it to the processor. "Input card" are additionally used to supply power to the sensors (detectors). Both signal (pulses) and power supply to the sensors are physically united in one coaxial cable. Processor performs control of the hardware (connected to the ISA bus), receives and processes information and puts it on the display.

Central station consist of:

- processor (5025A Control Card);
- video card (5420 Super VGA Card);
- display (Planar EL 640.400-CD3);
- 2 counter/timer cards (5300 Counter/Timer Card);
- digital-analog converter (5750 4-channel DAC Card);
- 2 input cards;
- modules of primary power supply.

Processor Card represents itself a PC on the base of processor 80386SX-25 MHz with 1 Mb operative memory (RAM). There are built-in disc subsystem on the base of solid state disks - microcircuits ROM (ROM) or RAM. Card can have up to 3 such disks:

- SSD0: ROM disk, containing in its composition BIOS and built-in DOS 6.0.
- SSD1: ROM disk up to 1 Mb on the base of the programmable by user memory.
- SSD2: ROM disk up to 1 Mb on the base of the programmable by user memory or RAM disk up to 512 Kb on the base of RAM with the battery support (or without it).

Video Card could support a number of the different type of displays. Some adaptation to the different types of displays could be performed using reprogrammable flash-memory. Different options of BIOS were delivered with this card as well. Card supports a majority of VGA modes.

Counter-timer Card (5300 Counter/Timer Card) is software-operated by the processor. There are 3 three-channel microcircuits of counters/timers 8254 and one microcircuit of parallel port 8255 on the card. The card in the system is used with six pulse channels on 8254 and one pilot channel on 8255.



Digital-analog converter (5750 4-channel DAC Card) is a 4 channel DAC. Each of channels of DAC realizes 12-bit transformation and works regardless of the others. By means of jumpers output voltage of each channel can be adjusted on ranges (0...5)V, (0...10)V or (-5...+5)V.

Input card is an original development for the device. Card consist of a converter of voltage 5 V/ 12 V for power supply of the sensors(detectors) and 6 channels for receiving of the information.

Central station is manufactured in plastic body with "door type" cover. On the cover ("door") are installed: display, speaker, regulator of brightness. Connectors of external cables and tumbler of switching "ON" a power supply are situated in "pocket" on back surfaces of body. Connection of keyboard is realized through the hole in the lateral wall. During operation the enclosure is installed on the stand, ensuring slopping 75 degrees for the horizontal plane. Inside of the body is installed 8-card framework and other auxiliary elements.

### Neutron detector

Neutron detectors are designed for continuous (non-stop) operation. It is manufactured at the level of protection IP66 and can be operated under temperature range from minus 20°C up to +50°C and relative moisture up to 80%.

Neutron detector consist of six helium type counters (SNM-18) and electronics circuits. Electronics circuits performs amplification, discrimination and shaping of the signal. There is a stabilization of the input voltage. The power supply to the counters (SNM-18) is realized by a high-voltage converter.

Neutron detector is located in the plastic body with 405x 400x120mm dimension. There is a connector of type CP50-73 BO on one of the sides to connect a communication link.

Electronic cards, counters and paraffin moderator are made as united construction, which is fixed to the body of enclosure by 6 screws.

### Results of tests

All technical features of the system were tested under laboratory conditions according to the program of testing. System successfully passed all the tests. Because of routine procedures and considerable volume of information the data are not presented on the report. It might be mention that the system has high stability and additional error due to the temperature is about (or less) 0.03%/°C.

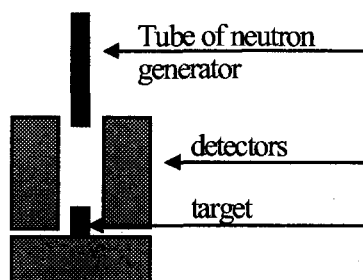


Fig. 3

As to the sensitivity of the system - it strongly depends on the geometry of the measurements, amount of the neutron detectors and number of neutron generator shots. Some experiments were performed in the geometry as it shown on the Fig. 3.

A number of small uranium samples were used in such geometry as a target. It was estimated that uranium emits approximately 0.95 delayed neutrons per g of U235 and 0.33 delayed neutrons per g of U238 per 30 shots of the neutron generator (neutron generator emits fast neutron).

In another experiment some plutonium samples were measured in passive mode (no shots from neutron generator) in the same geometry. Results are presented in the table and Fig. 3.

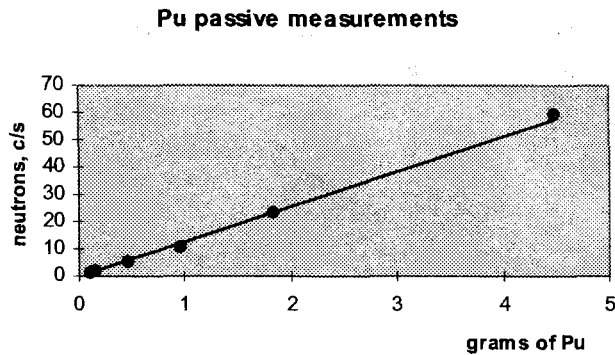


Fig. 3

Plutonium, g	Static measurements, c/s
0, background	0.2
4.48	59.7
1.83	23.5
0.96	11.2
0.47	5.8
0.16	2.5
0.11	1.6

Time of measurements - 5 min

### Conclusion

The System to detect the fissile materials by active neutron method (by delayed neutrons) was successfully developed, manufactured and demonstrated. The advantages of the System:

1. Ability to detect the fissile materials when others passive methods can not do a job.
2. High level of the intellect, PC card.
3. Openness of the system.
4. Wide range of the possible application.



## **CASE STUDY OF INVENTORY DIFFERENCE (ID) COMPUTATION AND ANALYSIS BASED ON THE RADIOCHEMICAL PLANT MODEL**

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Inventory Difference (ID) computation and analysis is an urgent task of high priority in the field of nuclear material control and accountancy (MC&A). In this paper this task is considered from the point of view of studying different practical cases (case study) in order to upgrade qualification of MC&A specialists.

Training courses which are regularly held in RMTC, SCI training center and discussions with specialists during these courses confirm how urgent this task is. Analysis of practical cases, close to the real conditions of particular plants, is usually of great interest for students. When trying to solve the tasks by themselves specialists have the chance to compare their own techniques for ID calculation and analysis with the methods and techniques given in the case study.

Conventionally the model of low-enriched uranium fuel fabrication plant [1] was used in the training courses as the basic case because it was considered the most developed in the domestic and international practice. In this paper the model of radiochemical plant [3] is considered, for this case the practical tasks and solutions have been developed.

The extraction technology forms the basis for the irradiated fuel reprocessing flow sheet. The structural diagram of the process of principal components extraction from the irradiated fuel and their purification is given in Fig.1. The extraction technology consists of a set of typical operations: extraction, scrubbing, backwashing, evaporation. This flow-sheet presupposes a joint U&Pu recovery from  $\text{HNO}_3$  in the inert diluent. After scrubbing the solvent product the partition of U and Pu is carried out by means of a selective backwashing of Pu with diluted nitric acid in the presence of a reducing agent. Uranium which stays in the organic phase is washed back by the diluted nitric acid solution. The further purification is performed separately in uranium and plutonium branches of the process by means of recycling.

Nuclear material flows for the given model of radiochemical plant are described in the following way:

1. The radiochemical plant for NPP nuclear fuel reprocessing received 350 batches with nuclear material during half a year. On average, each batch contains 3.6 kg Pu.
2. 170 batches of plutonium nitride were received during the same period (half a year), each batch contains on average 7.19 kg Pu.
3. The received material in the form of NPP fuel sub-assemblies (FSA) was reprocessed and resulted in a certain amount of liquid waste. The given radiochemical plant has 9 flows with 185 batches per a flow. Each flow contains about 2.15 kg Pu.
4. Solid radioactive waste is also formed in the course of reprocessing. During half a year 1650 containers were filled. The total amount of solid waste is equal to 8.5 kg Pu.

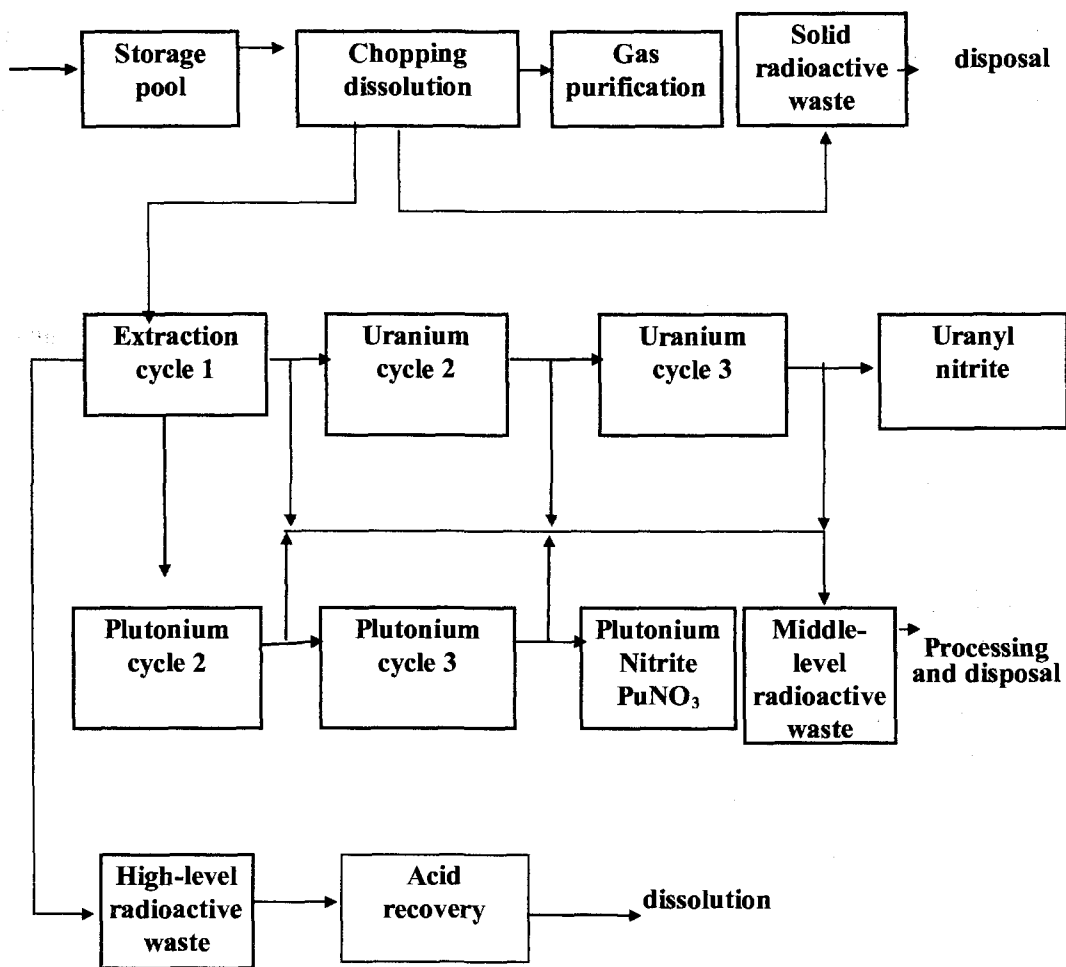


Fig. 1. Technological flowsheet of radiochemical plant model

5. The physical inventory taking (PIT) of accountancy tanks is based on the readings of those devices which are installed on them. In the radiochemical plant there are 10 accountancy tanks, which contain on average 1.23 kg Pu per a tank as the beginning inventory.
6. In the course of initial PIT all the residues (holdup) in the process equipment have been measured. The total amount of Pu is equal to 0.85 kg Pu.
7. The ending inventory of accountancy tanks was equal to 1.25 kg Pu per tank
8. The ending holdup inventory in the equipment was equal to 0.82 kg Pu.

The following measuring procedures are used in the given technological process in order to achieve MC&A goals:

- volume measurement;
- sampling and sample analysis;
- NDA
- indirect measurements.

The input accountancy tank volumes; output volumes with plutonium nitrate, liquid waste, process tank volumes are measured during the PIT. Samples for chemical analysis are taken from

the above-mentioned tanks. NDA techniques are used to measure Pu content in solid waste. The material holdup in the equipment is determined by indirect methods of measurement. Relative values of measurement errors are shown in Table 1.

All the instruments which serve for measuring volumes are not recalibrated during the material balance period. Analytical measurement systems are calibrated every month.

The task is as follows:

1. To calculate inventory difference
2. To determine a standard relative deviation (SRD) for the inventory difference
3. To find a confidence interval
4. To make the conclusion about the ID value in terms of NM accountancy for the given technological process.

### **SOLUTION**

1. Calculation of the inventory difference (in terms of Pu)

- 1.1 Input

$$350 \cdot 3.6 = 1260 \text{ kg Pu}$$

- 1.2 Output (plutonium nitrate,  $\text{PuNO}_3$ )

$$170 \cdot 7.19 = 1222.3 \text{ kg Pu}$$

- 1.3 Output (liquid waste)

$$9 \cdot 2.15 = 19.35 \text{ kg Pu}$$

- 1.4 Output (solid radioactive waste)

$$8.5 \text{ kg Pu}$$

- 1.5 Beginning inventory

Input accountancy tanks

$$10 \cdot 1.23 = 12.3 \text{ kg Pu}$$

Holdup in the equipment

$$0.85 \text{ kg Pu}$$

Ending inventory

Input accountancy tanks

$$10 \cdot 1.25 = 12.5 \text{ kg Pu}$$

Holdup in the equipment

$$0.82 \text{ kg Pu}$$

$$\text{ID} = 1260 - 1222.3 - 19.35 - 8.5 + 12.3 + 0.85 - 12.5 - 0.82 = 9.68 \text{ kg Pu}$$

The case study given in the paper is only the first version of ID calculation and analysis for a radiochemical plant. The author will be very much obliged to everyone who will express critical remarks or proposals about the improvement of practical cases which will be used for the next training courses.

TABLE 1. Measurement errors

Type of measurement	Object under measurement	Random error	Systematic error	Recalibration random error
1. Volume	Input tank	0.0035	0.0017	0.002
	Output tank (PuNO <sub>3</sub> )	0.0035	0.0015	0.002
	Liquid waste	0.055	0.03	0.03
	Physical Inventory	0.05	0.013	0.015
2. Sampling	Input tank	0.003	0.002	-
	Output tank (PuNO <sub>3</sub> )	0.005	0.002	-
	Liquid waste	0.065	0.065	-
	Physical Inventory	0.03	0	-
3. Analytical analysis	Input Isotopic Dilution Technique	0.0085	0.0025	0.0025
	Output Titration	0.0055	0.0025	0.003
	Isotopic Dilution Technique for Liquid Waste			
	NDA for Solid Waste	0.015	0.0035	0.004
	Isotopic Dilution Technique for process tanks during PITs	0.45	0.065	0.065
		0.015	0.0025	0.0025
4. Indirect measurements	Holdup in the equipment	0.075	0.075	-

TABLE 2. Calculation of a systematic error contribution

Type of measurement	Object under measurement	Systematic error, $\sigma_s$	Mass, M (kg)	Error contribution $M^2\sigma_s^2$
1. Volume	Input tank	0.0017	1260	4.5882
	Output tank	0.0015	1222.3	3.3615
	Liquid waste	0.03	2.15	$9(0.03)^2(2.15)^2=0.0374$
	Accountancy Tanks (physical inventory taking)	0.013	1.23-1.25=-0.02	$10^4(0.013)^2(-0.02)^2=0.0000006$
Sum = 7.9871				
2. Sampling	Input tank	0.002	1260	6.3504
	Output tank	0.002	1222.3	5.9761
	Liquid waste	0.065	2.15	$9(0.065)^2(2.15)^2=0.1758$
Sum = 12.5023				
3. Analysis	Input tank	0.0025	1260	9.9225
	Output tank	0.0025	1222.3	9.3376
	Liquid waste	0.0035	19.35	0.0046
	Solid Waste	0.065	8.5	0.3053
	Accountancy Tanks (physical inventory)	0.0025	12.3-12.5=-0.2	0.0000002
4. Indirect measurements	Holdup in the equipment	0.075	0.85-0.82 =0.03	0.000005
Sum = 19.57				
Total sum = 40.0594				

TABLE 3. Calculation of random error contribution

Type of measurement	Object under measurement	Random error	Mass, M (kg)	Number of measurements, N	Error contribution $\sigma_e M/\sqrt{N}$
1. Volume	Input tank	0.0035	1260	350	0.0556
	Output tank	0.0035	1222.3	170	0.1077
	Liquid waste	0.055	19.35	1665	0.0007
	Accountancy tanks (beginning inventory)	0.05	12.3	10	0.0378
	Accountancy Tanks (ending Inventory)	0.05	12.5	10	0.0390
	Sum = 0.2408				
2. Sampling	Input tank	0.003	1260	350	0.0408
	Output tank	0.005	1222.3	170	0.2197
	Liquid waste	0.065	19.35	1665	0.00095
	Accountancy tanks (beginning inventory)	0.03	12.3	10	0.0136
	Accountancy Tanks (ending Inventory)	0.03	12.5	10	0.0141
	Sum = 0.2892				
3. Analysis	Input tank	0.0085	1260	350	0.3277
	Output tank	0.0055	1222.3	170	0.2658
	Liquid waste	0.015	19.35	1665	0.00005
	Solid Waste	0.45	8.5	1650	0.0089
	Accountancy tanks (beginning inventory)	0.015	12.3	10	0.0034
	Accountancy Tanks (ending Inventory)	0.015	12.5	10	0.0035
	Sum = 0.6094				
4. Indirect measurements	Holdup in the equipment (beginning inventory)	0.075	0.85	1	0.0041
	Holdup in the equipment (ending Inventory)	0.075			0.0038
	Sum = 0.0079				
	Total Sum = 1.1473				

TABLE 4. Calculation of recalibration error contribution

Type of measurement	Object under measurement	Random error of recalibration	Mass, M (kg)	Error contribution $\Sigma \sigma_p^2$ , kg <sup>2</sup>
1. Volume	Input tank	0.002	1260	6.3504
	Output tank	0.002	1222.3	5.9761
	Liquid waste	0.03	2.15	$9(0.03)^2(2.15)^2=0.0374$
	Accountancy tanks (physical inventory taking)	0.015	$12.3-1.25=-0.02$	$10(0.015)^2(0.02)^2=0.0000009$
	Sum = 12.3639			
2. Analysis (on a monthly basis)	Input tank	0.0025	210	$6(0.0025)^2(210)^2=1.6538$
	Output tank	0.003	203.72	$6(0.003)^2(203.72)^2=2.2411$
	Liquid waste	0.004	3.225	$6(0.004)^2(3.225)^2=0.001$
	Solid waste	0.065	1.42	$6(0.065)^2(1.42)^2=0.0511$
	Accountancy tanks (beginning inventory)	0.0025	12.3	$(0.0025)^2(12.5)^2=0.00098$
	Accountancy tanks (ending Inventory)	0.0025	12.5	
	Sum = 3.9499			
Total sum = 16.3128				

Total variance =  $40.0594 + 1.1473 + 16.3128 = 57.5195$

Root-mean-squar error (standard deviation) = 7.5842

Confidence interval =  $-15.1684 \leq ID \leq +15.1684$

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## METHOD AND ALGORITHM OF THE ATTRIBUTE VERIFICATION OF NUCLEAR MATERIALS

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### Abstract

The report presents the results of the development of the software for the attribute tester of the nuclear materials. The software is oriented for the using of a scintillator as an detector. The development was performed on the basis of the IAEA research contract. The developed algorithm is capable to identify U-235 and plutonium. Additionally it is possible to estimate the enrichment of the uranium and Pu-241 and Am-241 ratio (separately) to the Pu-239. Using this algorithm it is not necessary to perform any kind of calibration except usual energy calibration. The method has very week sensitivity to the geometry of measurements and forms of the nuclear materials. The algorithm was tested using IAEA samples at Seibersdorf laboratory.

### The Goal of the development and field of implementation

For the IAEA Safeguards or for the State System of accountancy and control of the nuclear materials there are needs to perform the verification of the nuclear materials. Usually it is performed by the identification of plutonium or uranium - specific peaks. The procedures use in most cases the presence of one gamma-peak but not all the information contained in the gamma-spectrum. This simple approach makes it difficult to use detectors with limited resolution to verify mixtures of plutonium and uranium or these isotopes in the presence of different background radiation and under different geometry of measurements.

The goal is to develop such algorithms so it could be possible to recognize the Uranium and/or Plutonium and make some estimation of the enrichment using LOW RESOLUTION (NaI) DETECTOR. It could be very important if such an estimation could be done without preliminary U235 calibration and could be more or less independent on the geometry of measurement, time of measurements and on the form of the materials. The development was performed under IAEA Research Contract /1-4/. Possible application of the software could be as follows:

- Attribute verification of the nuclear materials by IAEA inspectors
- Verification of the nuclear materials by state inspectors
- Using by Operator to control and identify nuclear materials
- Using in some special cases of the nuclear materials control where the well known and most correct measures are not applicable because of the sensitiveness of the area.

## Description of the algorithm

## Identification of the Uranium and Plutonium

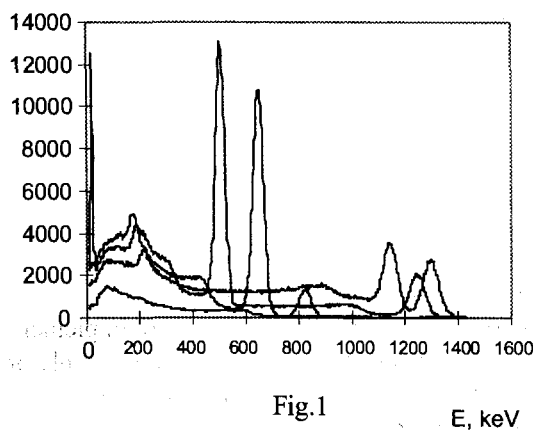


Fig.1

The simplest way to identify the U235 and Plutonium is to look for 186 keV peak for the Uranium and 208 keV peak for Plutonium. But first off all these peaks are very close (NaI) and secondly there could be back scattering peaks in the region from others nuclides. It means that proper identification sometimes could be not so easy as it seems to be. The energy of the back scattering peaks are:

- 171 keV for Na22,
- 185 keV for Cs137,
- 196 keV for Mn54
- 215, 223 keV for Co60

The example of these peaks is presented on the Fig.1. The back scattering peaks of the Na22, Cs137, Co60 (3 upper curves) could be very clearly seen.

To perform the identification in the automated mode by software it is worthwhile to use the wide energy region looking for the shape of the spectra. There are quite drastic difference between uranium and plutonium shapes (Fig. 2) of the spectra. Others nuclides have specific features as well. To identify the Uranium, the following features of the spectra are taking in the consideration by the algorithm:

- Existence of the 186 keV peak (very essential)
- Existence of the 766 and 1001 keV peak (essential but could be very small for the very high enriched uranium)
- Specific shape of the spectra in the energy region (156 - 338 keV). There are a number of U235 peaks in this region (Fig. 3). Some of them could be seen (NaI) only for high enriched uranium so some qualitative information on the uranium enrichment is there as well.

The same approach is used for plutonium identification:

- Existence of the 208 keV peak (very essential)
- Specific shape of the spectra in the energy region (156 - 430 keV). Especially important is the existence of the wide 3 heads peak in the region of the approximately 375 keV. (see Fig. 2)

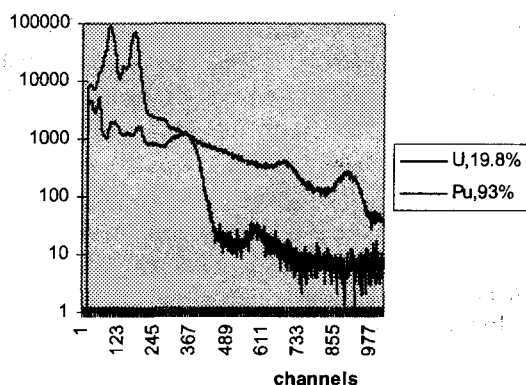


Fig.2

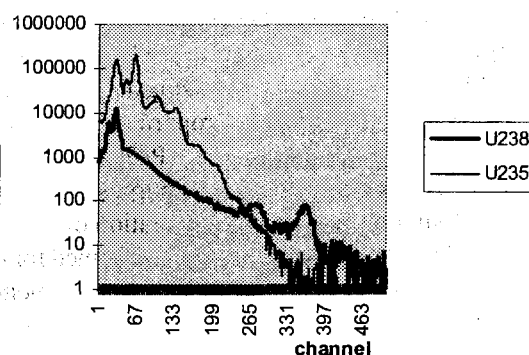


Fig.3

When we are speaking about the peaks, the program finds peaks and calculate the areas of it. The shape is investigated by a number of short energy region where the integral counts are used. In first steps the software gathers information on all features of the spectra: peaks and shape of the specific energy regions. The second step - analysis of the gathered information and making a decision:

- U235 is detected or
- Plutonium is detected or
- MOX is detected or
- U235 is not detected, Pu is not detected.

### ***Estimation of the U235 enrichment***

In principle it is possible to make estimation of the U235 enrichment using the ratio between 186 keV peak and 1001 keV peak /5/. This approach is used at this algorithm. To implement this method it is necessary to find out some constants which depends on the detector (mainly on the size of it). What is interesting that it becomes possible to perform rough estimation of the enrichment without special calibration. Only energy calibration is needed. But it could be done once by experts so the inspector will do very simple job - just press a button and get results. What's more interesting - with the same set of constants it is possible to make rough estimation of the enrichment for the samples with different matrix not thinking too much about geometry of measurements.

The area of the 186 keV peak is proportional to the U235 contents in the sample. The area of the 1001 keV peak is proportional to the U238 contents in the sample. Using the ratio of these areas the estimation of the enrichment could be made. The enrichment could be estimated by equation (1).

$$\text{Enr235} = 100 * b / (Z + b - a), \quad (1)$$

where: Enr235 - estimated enrichment, Z - ratio of the 1001 keV peak area to the 186 keV peak area. Estimated coefficients are (IAEA spectra):  $a = 0.0$ ;  $b = 0.0018$ .

In principle it is possible for the same purpose to use the ratio of the 186 peak area and the integral counts in the e.g. region of the (320-370 keV), which is proportional to the sum of the U235 and U238 contents taking with some coefficients. The same equation (1) could be used but with another set of coefficients.

Estimated coefficients for this case are (IAEA spectra):  $a = 0.01$ ;  $b = 0.01$ .

### ***Some analysis of Pu composition***

It is clear that real analysis of the Pu composition could not be performed using NaI detector. But some information still could be get even in that case. Approach is the same as for estimation of the Uranium enrichment using the ratio method. It is necessary to find out or calculate somehow the figure (area) which is proportional to the Pu239 contents; to find out the area (region) which is proportional in mixture to the Pu239 and another nuclide; make ratio; calculate the ratio of this nuclide to the Pu239. For this purpose the most interesting is the region (300-430 keV).

Using the data /6/ the calculation on the contribution to the 208 keV peak and wide peaks at the 378 keV and 662 keV were performed for the IAEA Pu standards. The contents of the nuclides were recalculated on the date of measurements. The relative contribution of the nuclides for the 378 keV wide peak is presented on the Table 1. In that calculation (for 378 keV peak) lines with energy less than 370 keV were not taken into consideration. First of all they are rather weak and their contribution is rather small especially because the software is using only right wing of the

378 keV wide peak to calculate the area. It could be seen from the Table # 1 that the main contribution to this peak makes Pu239, especially for samples with high contents of the Pu239. For the rough estimation (quite correct is not possible) we will neglect the Am241.

Table 1

	Am241 contents, %	Pu239 contents, %	Am241 contribution, %	Pu239 contribution, %
IAEA st. #1	3.97	62.66	10.6	89.4
IAEA st. #2	3.23	73.42	7.6	92.4
IAEA st. #3	0.610	84.39	1.3	98.7
IAEA st. #4	0.184	93.43	0.4	99.6

The relative contribution of the nuclides for the 208 keV peak is presented on the Table 2. It could be seen from the Table # 2 that the main contribution to this peak makes Pu241. There is considerable contribution of the Pu239, especially for samples with high contents of the Pu239. The contribution of the Am241 for these samples is about 4%. For the rough estimation (quite correct is not possible) we will neglect the Am241.

Table 2

	Am241 contents, %	Pu241 contents, %	Pu239 contents, %	Pu241 contribution, %	Am241 contribution, %	Pu239 contribution, %
IAEA st. #1	3.97	4.104	62.66	94.4	4.63	0.97
IAEA st. #2	3.23	3.350	73.42	94.1	4.6	1.3
IAEA st. #3	0.610	0.633	84.39	88.0	4.3	7.7
IAEA st. #4	0.184	0.136	93.43	65.9	4.5	29.6

The relative contribution of the nuclides for the 662 keV wide peak is presented on the Table 3. There is considerable contributions of the Pu239 and Am241.

We will use the 378 keV wide peak area as a representative of the Pu239. We will use the 208 keV peak area as a representative of the Pu241 and Pu239. We will use the 662 keV wide peak area as a representative of the Am241 and Pu239. Now we can estimate some ratios.

Table 3

	Am241 contents, %	Pu239 contents, %	Am241 contribution, %	Pu239 contribution, %
IAEA st. #1	3.97	62.66	91.5	8.5
IAEA st. #2	3.23	73.42	88.3	11.7
IAEA st. #3	0.610	84.39	55.5	44.5
IAEA st. #4	0.184	93.43	25.3	74.7

$$\text{Pu241/Pu231} = (Z - a) / b, \quad (2)$$

where: Z - ratio of the 208 keV peak area to the 378 keV wide "peak" area and estimated coefficients are (IAEA spectra, 1996 y., standard IAEA NaI detector): a = 0.15; b = 398.

For the Am241/Pu239 the same equation (2) could be used but with another set of coefficients. In that case Z is the ratio of the 662 keV peak area to the 378 keV wide "peak" area and estimated

coefficients are (IAEA spectra, 1996 y., standard IAEA NaI detector):  $a = 0.266$ ;  $b = 37$ .

## Results of tests

### Uranium enrichment estimation

The IAEA spectra were used for testing. Material  $\text{UO}_2$ , infinite. Detector - standard IAEA (NaI). The spectra were taken without shielding and with some steel absorbers placed between samples and detector. The energy region was above 1001 keV so this was used during calculations. Results are presented on the Table 4.

Another set of the IAEA spectra were used for testing. Results are presented on the Table 5. In

Table 4

Enrichment real, %	Enrichment measured, %	Remarks
0.708	0.99	no absorbers
0.708	0.87	4.5 mm. Steel absorber
0.708	0.74	10 mm. Steel absorber
3.105	3.06	no absorbers
3.105	2.68	4.5 mm. steel absorber
3.105	2.33	10 mm. steel absorber
18.82	19.71	no absorbers
18.82	16.81	4.5 mm. steel absorber

that case the energy region was about 512 keV so the 1001 keV peak could not be used for enrichment calculations. Instead of that the integral counts in the region of the (320 - 370 keV) were used as representative of the U238.

Table 5

Enrichment real, %	Enrichment measured, %	Remarks
0.72	0.88	$\text{UO}_3$ powder
4.46	4.87	$\text{UO}_3$ powder
26.305	23.51	metal
89.99	89.4	metal
0.475	0.47	metal

### Plutonium composition estimation

The spectra of the IAEA standards were used in calculations according the method described on the report. Results are presented on the Table 6 and Fig. 4.

Table 6

Pu239, %	Pu241/Pu239 real	Pu241/Pu239 measured	Am241/Pu239 real	Am241/Pu239 measured
62.66	0.0654	0.0657	0.0632	0.0655
73.42	0.0456	0.0456	0.0440	0.042
84.39	0.00746	0.0077	0.00723	0.0076
93.43	0.00146	0.00145	0.00193	0.002

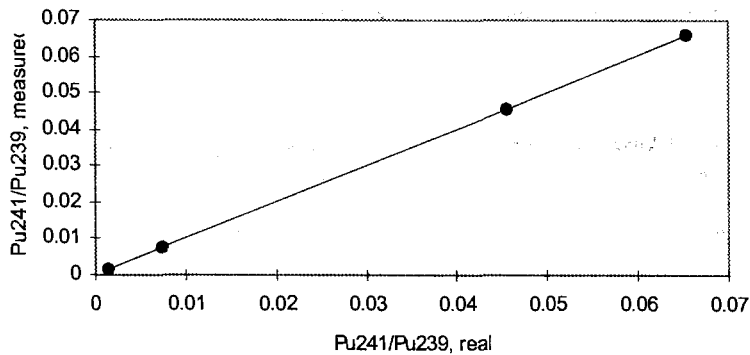


Fig.4

## Conclusion

A scope of investigation on getting maximum information from spectra taken by NaI detector was undertaken.

1. The algorithms for U235 and Plutonium identification in automated mode have been developed.
2. The algorithms for rough U235 enrichment estimation which is independent on the measurement time and low sensitive to others factors have been developed.
3. The algorithm for rough estimation of the Pu241/Pu239 and Am241/Pu239 ratios have been developed.
4. The test of algorithms was a performed on different sets of samples including IAEA standards.

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## NUCLEAR REACTOR FUEL CYCLE TECHNOLOGY WITH PYROELECTROCHEMICAL PROCESSES

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### Introduction

The current stage of nuclear power development is characterized by more stringent requirements to all the stages of fuel cycle in terms of their safety and impact on the environment. When nuclear materials (NM) of the military origin are involved into the fuel cycle there appear additional requirements to their non-proliferation and thus, additional requirements to the physical protection, control and accounting systems. Today oxide fuel is the main fuel type for the current nuclear power installations. Evidently in the nearest future any new fuel type will not be involved. In this connection the technologies which will make it possible to extend the potential capabilities of oxide fuel and to improve engineering and economical parameters on the whole may be of great interest.

In the NIIAR the basic principles of advanced closed fuel cycle (FC) have been formulated and experimentally justified by now. This FC is based on mutual compatibility of U-Pu recovery technologies and FE and FSA fabrication technologies. These principles are as follows:

- to use "dry" pyroelectrochemical processes in order to reprocess irradiated fuel and to dispose waste;
- to get the granulate of polydispersed composition with a high density of particles in the course of recovery;
- to use a vibro-packing technique in order to fabricate fuel elements (FE) from granulated fuel;
- to use remotely controlled automatic equipment in the course of fuel reprocessing and FE and FSA fabrication.

"Dry" methods make it possible to reprocess fuel with any burn-up values and holdup time covering a small number of technological stages. These processes do not require any moderator, thus allowing high concentrations of fissile and radioactive materials to be worked with. That is the principal reason why the "dry" processes are so efficient, compact and generate a low amount of waste making the MNC&A process much simpler.

The remotely controlled processes are very important in the FC technology and the additional expenditure related to the transfer from simple glove-boxes to the shielded glove-boxes are justified due to the following reasons:

- there is a feasibility to optimize fuel purification from fission products to the level of reactor requirements. It results in a certain decrease in expenses on additional fuel purification and processing additional waste volumes;

- it is possible to remove strict restrictions on the content of high-toxic isotopes and transplutonium elements in fuel;

- it becomes possible to develop new fuel cycle scenarios, for instance:

the fuel may be reused after its partial processing with the aim to remove gas and volatile fission products;

the reactor blanket fuel may be directly used in the BN reactor core;

the fuel from reactors with a certain level of enrichment may be burnt in reactors with a lower level of enrichment through partial fuel processing;

- their not very high purification makes NMs "less attractive", increases the level of their self-protection and allows all the NM movements to be easily controlled, thus decreasing the risk of their diversion.

The experimental and research complex (ERC) which comprises a granulated fuel fabrication facility serves as an experimental base for developing and refining the above mentioned principles. The fabricated FEs and FSAs are tested in BOR-60 and BN-600 reactors.

## 1. Pyroelectrochemical technology for fuel reprocessing

Any type of fuel (oxide, metal, carbide, nitride) can serve as a feed material for the pyroelectrochemical process. At the first stage it is dissolved in the molten salt, the chloride ones being the most convenient and well-studied. Fuel is extracted from the molten salt by means of electrolysis and methods of precipitation in the form of crystal oxides, e.g.  $\text{UO}_2$ ,  $\text{PO}_2$  or  $\text{UPuO}_2$ . As soon as the precipitate is ground and the crystal products are washed from the entrapped salt-solvent and other soluble impurities the granulate of a polydispersed composition is being formed. This granulate is used to fabricate vibro-packed fuel elements. The particles have the density not less than  $10.7 \text{ g/cm}^3$  and the size not bigger than 1 mm. The washing solutions are recovered by their evaporation which is followed by taking the dry precipitate back to the onset. The pyroelectrochemical process makes it possible to remove fission products from the fuel with the total purification factor more than 100, which is sufficient from the point of view of fast reactor physics.

The waste volume is minimum. Some valuable elements can be extracted from it, for instance, metals of the ruthenium subgroup. There are no strict requirements to the gas medium of the processes in this technology, that is why these processes are performed in the shielded glove-boxes filled with the usual air.

In terms of material control and accountancy the specific feature of granulated fuel fabrication technological processes consists in the fact that nuclear materials are concentrated in the solid phase and are reprocessed in portions with obligatory accounting operations (taking weight and analytical control) when receiving the input NMs and when moving them from one area to another.

The batches of fabricated fuel are transferred to the FE and FSA fabrication facility. Before being shipped the fuel batch is weighed and is accompanied with its passport with the indication of its mass, Pu or U concentration, isotopic composition, impurities, etc.

## 2. FE and FSA fabrication

Vibro-packing technology was always considered as one of the ways to fabricate fuel element cores which makes it possible to considerably decrease a nuclear reactor FE production cost and to improve FE performance. The main advantages of vibro-packing technology and FEs with vibro-packed fuel are:

- technological process simplicity and reliability due to a lower number of process and



control operations. It makes it easier to robotize and remotely control this technology thus allowing it to be used for FE fabrication in shielded glove -boxes. This advantage manifests itself to the most extent when dealing with highly-radioactive (recycled) fuel;

- it is possible to fabricate a fuel core with parameters which can be easily varied and based on multicomponent compositions;
- it is possible to use the granulate of any form, both of a homogeneous composition and in the form of mechanical blend;
- in comparison with the core of a pellet type, the vibro-packed fuel demonstrates a lower thermomechanical impact on cladding;
- the requirements to the FE clad inner diameter are not so stringent.

The process of vibro-packing consists in tight packing of powder particles of a certain granulometric composition under the impact of mechanical oscillations. Due to vibration the dispersed powder medium gets the property of quasi-liquid. In this case fuel particles gain mobility and capability to form an utmostly packed structure in accordance with particle sizes and their composition.

The feasibility to fabricate FEs by the vibro-packing technique was demonstrated in two options: manually in the glove-boxes and remotely in the shielded glove-boxes. A pilot-commercial implementation of remote methods of vibro-packed FE and FSA fabrication for BOR-60, BN-350 and BN-600 reactors was demonstrated in such facilities as "Oryol" (1977-1986), ERC (1989-1997) and "Kolibri" (1992-1997). Currently two chains of glove-boxes are in operation to fabricate FEs by vibro-packing. This is the place where different experimental programs are implemented and where FEs and FSAs for BOR-60 standard loading are fabricated. The 25-year experience in fabrication made it possible to formulate the main principles of FE production with vibro-packed oxide fuel, to develop and test the basic process conditions, to select the equipment with adequate specifications.

In the course of automatic line pilot operation the proper product output exceeded 98 %.

**TABLE 1. FE production with vibro-packed oxide fuel**

Fuel type	Reactor	Number of FSAs, pieces	Number of FE, pieces
UPuO <sub>2</sub> (HB)	BOR-60	426	15762
UPuO <sub>2</sub> (LB)	BN-350	2	254
UO <sub>2</sub> (recycled)	BN-350	7	889
UO <sub>2</sub> (recycled)	BN-600	6	762
UPuO <sub>2</sub> (LB)	BN-600	6	762
UPuO <sub>2</sub> (HB)	BN-600	4	508
UPuO <sub>2</sub> (LB, HB)	BFS	8	1016
UO <sub>2</sub> (recycled)	BOR-60	235	8695

Total (by the end of 1998):

FSAs - 714, FEs - 29402,

HB - High Background, reactor - grade Pu,

LB - Low background, weapons - grade Pu,

recycled, product of irradiated fuel reprocessing at the "Mayak" Plant

When FEs were fabricated manually in the process line located in the shielded glove-box ( $\approx 1300$  FEs for BN-350,  $\sim 300$  FEs for BN-600,  $\approx 9000$  FEs for BOR-60), the proper FE output was equal to 100% actually in terms of all quality parameters under control.

### 3. Waste management

Management of waste and circulating products of the granulated fuel and FE and FSA fabrication technology can be divided into several processes:

- reprocessing of circulating products generated in the course of preparation of granulated fuel and FE production;
- processes of off-gas purification from chlorine, sublimates and radioactive aerosols;
- purification of spent electrolyte from impurities;
- incineration and purification of flammable waste.

Besides, certain methods of U and Pu extraction from the sludge after salt purification and U and Pu extraction from solutions after decontamination of very dirty parts of equipment have been developed.

All the waste and circulating products which contain NM and which are formed in the course of fuel and FE production are collected in separate containers; if possible, samples are taken with the aim to analyze NM content. When the U and Pu concentration is determined the waste and circulating products are processed in a chlorator-electrolyzer followed by the conditioned fuel production. Before reprocessing the waste products are accumulated and stored in the shielded box. The estimated data on the amount of waste are given in tables 2-3.

**TABLE 2. Solid radioactive waste in the ERC facility generated in the course of MOX-fuel production (estimation at the annual output of 1000 kg fuel)**

Type of waste	Waste output kg/kg fuel	Amount of waste kg per year	Fission material concentration, g/kg waste	
			Pu	U
High-level products directed to the storage (when the real output increases up to 1000 kg they will be processed with Pu extraction)				
Concentrate after salt purification	0.013	13	0.02%	0.08%
Cleaning cloth and packing materials	0.02	20	0.033%	0.040%
High-level waste subject to disposal				
Equipment made of pyrographite	0.2	200	0.4	1.6
Filters	0.13	130	0.7	1.5
Low-level solid waste subject to disposal				
Components of metal equipment, monitoring devices and remote control (after decontamination)	0.7	700	-	-

TABLE 3. Liquid radioactive waste in the ERC facility (estimation at the annual output of 1000 kg fuel)

Type of waste	Specific activity Cu/L (Bq/L)	Amount, L/year	Product concentration, g/L	
			Pu	U
Solutions after passing through the recycling and chlorine absorption installation	$3.1 \times 10^{-6}$ $(1.15 \times 10^5)$	$2 \times 10^5$	$5 \times 10^{-5}$	$1.5 \times 10^{-4}$
Desorbing solutions	$\times 10^{-9} \div 1 \times 10^{-7}$ $(3.2 \div 37) \times 10^2$	$12 \times 10^5$	$1.4 \times 10^{-7} \div$ $1.6 \times 10^{-6}$	$4.2 \times 10^{-7} \div$ $4.8 \times 10^{-6}$

#### 4. Accounting system

The objective of material accountancy is to check NM availability or to detect NM losses in the course of inspections or physical inventory taking (PIT). In the NIIAR the system of NM double accountancy (double registration system) has been accepted. It is characterized by the following principles:

- any loss must result in the corresponding equal gain in some other place;
- each NM accounting operation is registered at least in two registration documents.

The facility for granulated fuel production and FE and FSA fabrication (ERC) is part of the Chemical and Technological Division of SSC RF NIIAR. Its director and chief engineer bear personal responsibility for the entire NM accounting system operation in this division. In the division there exists a team responsible for NM accountancy, they provide the current activity on NM control and accountancy. The administrative management of this team is under the responsibility of the chief engineer of the division. As to the methodological supervision, it is the responsibility of another division of the Institute, i.e. Storage, Transportation, Control and Accountancy Division.

The three - level system of NM accountancy has been accepted in the Institute.

Level one - NM accountancy in the whole MBA;

Level two - NM accountancy in the facilities, installations and storages;

Level three - NM accountancy in the process operations in facilities.

**Level one** is the top level and is being carried out by the specialists from the NM accounting team of the division. They are responsible for records in the NM accounting logs, for the reports about NM movement which should be ready every quarter. Each type of NM is being registered in a separate log.

**Level two** is under the responsibility of NM accounting team (for NM in the storages) and the persons in charge for NM accountancy in the facilities (one person from each facility). It means that the members of NM accounting team combine the functions of NM accountancy and storage, because they are responsible for receiving NMs for the division, their storing in the storage facility, their shipment with the aim to be involved into the work and their transfer to different parts of the Institute.

**Level three** is carried out by the responsible persons who directly perform the work in the facility after receiving NMs from the person in charge in this facility. They keep account of NMs in process logs, covering each process operation in the course of reprocessing or when transferring NM from one responsible person to another (for instance between shifts), when giving NM to the

person in charge for accountancy after its complete or partial reprocessing in the form of final products, waste, arbitrary samples, etc. They fill in record cards (passports) which accompany each final product and submit total or intermediate balance to the person in charge for accountancy in order to keep account and records on reprocessing for the first two levels.

The forms of process logs are developed in view of requirements to reprocessing technology and NM accounting system at the level of this technology. Formally the process logs and other documents of the third level of accounting are not considered as NM accounting documents, they are not under requirements of the double registration system. But the process data serve as the input data for NMC&A and the reliability of accounted data depends to a large extent on the process measurement.

The main principle of the three - level system of accountancy is the principle of being under control from the top down and reporting from the bottom upwards, i.e. the specialists who keep account of a certain level of accountancy can control all the levels below both in terms of records and NM availability.

In the existing control and accounting system the facilities for granulated fuel production and FE and FSA fabrication cover two independent accounting areas which have the features typical of MBA, i.e.:

- all the NM movements in these areas are measured and registered in the corresponding key measurement points;
- they have certain physical boundary lines and do not overlap other MBAs;
- personal responsibilities have been determined in these areas in terms of NM accountancy.

Thus, the material balance in the facilities is closed at independently as necessary, for instance, after reprocessing a fuel batch or before PIT.

For the time of physical inventory taking the facility operation is stopped for a while when fuel batch reprocessing under program is over. During this time each facility is involved in working out certain documents on all the batches that have been reprocessed, on material balance for each batch and on the product passportization. As all the NMs used in the processes are compactly concentrated into the solid phase and are reprocessed portion by portion with obligatory accounting activity, the PIT process is significantly simplified and can be restricted by checking accounting documents with a number of confirmatory measurements for the particular items.

## **Conclusion**

A group of "dry" technologies and processes of vibro-packing granulated fuel in combination with unique properties of vibro-packed FEs make it possible to implement a new comprehensive approach to the fuel cycle with plutonium fuel. Testing of a big number of FEs with vibro-packed U-Pu oxide fuel in the BOR-60 reactor, successful testing of experimental FSAs in the BN-600 reactor, reliable operation of the ERC facilities allow us to make the conclusion about a real possibility to develop a safe, economically beneficial U-Pu fuel cycle based on the technologies enumerated above and to use both reactor-grade and weapons-grade plutonium in nuclear reactors with a reliable control and accounting system.



## UNIFIED INSTRUMENTATION FOR DETERMINING FISSILE AND RADIOACTIVE MATERIALS

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The paper is concentrated on the approach to developing a unified instrumentation aimed at detection of fissile and radioactive materials. Specifications of monitors which are involved in this set are given below.

Currently in the VNIIAR the unified instrumentation for detecting fissile materials (FM) (mainly uranium and plutonium), and radioactive materials (RM) has been developed almost completely. This instrumentation is aimed to equip various facilities: nuclear facilities (including radioactive plants and nuclear material storages), border check stations at the customs, transport junctions, administrative buildings and other facilities.

The instrumentation of identical purpose (portal pedestrian and vehicle monitors, hand-held monitors) has a very narrow specific application and cannot be used for other tasks. It makes the process of equipping the above-mentioned facilities more difficult because it requires the list of instrumentation types under design to be extended and this results in increasing the costs. So it will be reasonable to develop a set of instruments unified in terms of their application.

The experience of designing and operation of domestic and foreign radiation monitors, the analysis of the conditions of their application showed that it is expedient to develop a set of instruments with a common metrological support and methodological control and, if possible, the development should be based on the common technical solutions. The set of instruments must comprise:

- a radiation monitoring system (RMS) meant to automatically detect ionization sources in moving objects (pedestrians, vehicle, luggage, mailing, etc.)
- two types of hand-held monitors meant to localize radiation sources in a suspicious object:  
type I - a hand-held monitor of a small size;  
type II - a high-sensitive hand-held monitor designed to detect radiation sources behind shielding and to carry out careful inspection in arguable cases.

The monitors are designed in accordance with the branch standard OST 95 10539-97, called "Radiation monitoring equipment. General technical requirements and methods of testing".

The monitors under design are based on the gamma-spectrometric method of radiation monitoring which consists in recording and analyzing characteristics of X-ray and gamma-sources power spectra within the range of 40-3000 keV at the background level whose value is measured and taken into account during the signal analysis.

Gamma-radiation is recorded by means of a scintillation detector. The electronic processing part of monitors is actually a device for amplitude selection with one or two tunable windows. The radiation monitor determines the gamma-quanta intensity in the optimal energy range and in case of brief exceeding the radiation background level it generates a sound signal and reflects the corresponding excess of intensity on the digital display.

The main monitor parameter is a detection threshold, i.e. FM mass which is detectable in 50% and even higher number of cases at the preset external conditions (background, geometry, method of inspection, etc.). This parameter is especially important for detecting low-active FMs (for instance, low-enriched uranium pellets), as well as in case of attenuation of radioactive and fissile material radiation in the result of the use of various vehicle types, containers and other protective shields. In the given set of instrumentation their compatibility in terms of the detection threshold has been provided, i.e. the radiation source determined by the radiation monitoring system is also detectable by the hand-held monitor of the first and second types.

A high detection sensitivity of monitors is mainly provided by the scintillator, its material and geometrical sizes. Several types of scintillation detectors have been investigated: NaI (Tl), CsI (Tl), BGO and plastic ones with various sizes. The best characteristics were demonstrated by the scintillators based on cesium iodine.

The instrumentation metrology is based on the single set of reference FM sources, common techniques of metrological certification, instruments testing after production and their calibration during operation.

The proposed common solutions have been implemented in the unified set of instrumentation for radiation monitoring which consists of a radiation monitoring system RMS-1 and hand-held radiation monitors GNOM-1 and STRAZH-2.

The radiation monitoring system RMS-1 comprises small-sized detectors TMGI69, a control unit and a tuning panel.

The number of detector units (1-8) and the distance between them can be chosen proceeding from the preset configuration of the range under control and the detection threshold. A small dimension of the detecting unit and its structural flexibility make it fast and easy to install RMS-1 in the access control points (check points) of any type which are already in operation or to equip doorways, conveyors for checking luggage, belongings, mailing, etc.

Availability of interface RS485 (RS432) allows RMS-1 to be used together with (as a part of) other systems.

A tuning panel with a sound and visual indication of FM detection (supplied separately) provides easy tuning and independent application.

A hand-held monitor GNOM-1 consists of two units: a portable small-sized detection unit and a control unit. These two can be either joined directly and form a compact mono-unit or joined with a rod of 0.5...3.0 meters long. Due to that the monitor is very convenient for examination of hardly assessable places, vehicle, railway carriages, and rooms with the ceiling height up to 5 m for detecting the places of unauthorized FM storage, radioactive contamination, radioactive leaks from pipelines, etc.

The monitor has an audible and visual digital indication. When the ionizing radiation exceeds the preset threshold, the monitor actuates a continuous audible warning and the sound pitch varies with the change in the gamma-quanta count rate. It is possible to connect it with the computer through a standard interface. The monitor has an automatic control of a storage battery cell discharge.

A wide temperature range of its efficiency is provided by introduction of an automatic compensation of environment temperature impact.

Specifications of a universal set of monitors

Characteristics	Type of Monitor		
	RMS-1	GNOV-1	STRAZH-2
Detection Threshold, g			
uranium	10	2	0.3
plutonium	0.3	0.15	0.03
False alarm frequency	1/1000 inspections	0.1 - 1/min	0.1-10/min
Time span of continuous operation	Round-the-clock	8 h	20 h
Operating temperature range, °C	-40 - +50	-40 - +50	-30 - +50
Mass, kg	Detector - 4,5 Control unit - 1,5	0.47	2.2
Overall dimensions (volume)	Detector - 495x67x69 mm <sup>3</sup> Control unit - 200x200x200 mm <sup>3</sup>	0.5dm <sup>3</sup>	252x96x12 mm <sup>3</sup>

A high-sensitive hand-held monitor "STRAZH-2" is made in the form of a single unit.

Its high sensitivity makes it possible to use the monitor to detect radioactive materials with low activity, including those which are shielded, inside the production cycle. This device is used for checking vehicles, luggage, belongings, mailing and for monitoring radioactive contamination in the environment, food products, etc.

The audible and visual indication of FM detection allows the monitor to be used in the conditions of insufficient light or too loud noise.

The built-in system of efficiency control and battery cell discharge control results in a high reliability of the device operation.

Thus, the designed universal set of instrumentation based on common technical solutions and metrological support plus its small dimensions allows to install it actually in any check point without any significant changes in the room lay-out to facilitate its maintenance.



## EXPERIENCE GAINED WITH CERTIFICATION OF INSTRUMENTS FOR THE SYSTEM FOR NUCLEAR MATERIAL PHYSICAL PROTECTION, ACCOUNTING, AND CONTROL (MPC&A)

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The experience gained is analyzed, data referred to, and problems emerging in the certification of imported equipment for MPC&A purposes, which was a pioneer experience for Russia. Some upgrades are proposed for certification procedures concerning imported equipment.

Federal Law of RF "On the use of nuclear energy" envisages obligatory certification of equipment, items, and technologies for nuclear facilities, sources, and storage facilities (CEST). Certificate of compliance is provided by a certifying body based on the results of sample tests accomplished by the Testing Laboratory.

The certifying and Testing Laboratory must be certified for specific works. The Russian Institute for Automatics (VNIIA) has performed necessary procedures for the organization and accreditation of the Testing Laboratory for Physical instruments subordinate to the Russian Standardizing authority Gosstandard (IFLA) using ionizing radiations (alpha-, beta-, neutron ones). On June 29, 1998, the Licensing Certificate № РОСС. RU0001. 22. ЭР07 was given to the VNIIA by the Gosstandard. The range of items the IFLA is licensed to deal with is given in Table 1.

To date, the IFLA has performed certification tests of a series of equipment specimens for MPC&A-related applications manufactured in the US: portable radiation monitors PRM-470A (manufacturer: TSA Systems, Ltd.) and JHH-21 (manufacturer: Canberra Industries, Inc.), radiation monitors for pedestrians PM-700SP (manufacturer: TSA Systems, Ltd.), and JPM-21A (manufacturer: Canberra Industries, Inc.).

In the course of testing, omissions in the accompanying documents, equipment certified, and problems related to the interactions with the equipment manufacturers were revealed.

Before the beginning of certification tests, the accompanying documents for portal pedestrian radiation monitors and portable ones for special nuclear materials (SNM) shipped were analyzed. Results of the analysis are given in Table 2.

As it can be seen from Table 2, the accompanying documents do not meet the Russian users' requirements to documents for instruments of this class.

Texts of several accompanying documents contain discrepancies. Thus, e.g., in the manual for RM-700 monitor, part 1.2. indicates sensitivity of 10 g high enriched uranium (HEU), whereas part 1.4-3 g HEU are referred to.

As it has been stated above, the accompanying documents to the instruments did not contain several specifications, which necessitated a long coordination of the tests' scope and techniques to be used.

Users' guides being insufficiently clear, additional contacts proved necessary between the VNIIA, manufacturers, and PNNL, in order to make clear the values of the monitor initial characteristics and parameters to be defined by tests (discrimination levels, duration of time intervals when detecting the SNM carried).



**TABLE 1. Range of certified licensing for ILFA**

Items of equipment tested	Index
1. Devices, units, detecting units for information conversion	43 6150
2. Instruments, installations, dosimetry systems	
3. Instruments, installations, radiometry systems	43 6210
4. Instruments, installations, spectrometry systems	43 6220
5. Instruments for measurement and verification of physico-chemical values	43 6230
6. Tools for measurement of physico-chemical values and parameters	43 6340
7. Signaling and actuating devices of security	43 8170
8. Instruments and apparatuses for security signaling systems	43 7230
	437291
9. Engineering tools for security divisions	
10. Detecting devices, active	70 3200
11. Detecting devices, passive	70 3240
12. Detecting devices, active+passive	70 3250
13. Components for tools, auxiliary devices, and spare parts	70 3270
	70 3280
	70 3290
14. Security Controls	
15. Access controls	70 3360
	70 3370
	70 3380

Verification of the monitors performance before testing showed specimen JHH-21 to be not serviceable, and it was returned to the manufacturer for repair.

The manufacturers overestimated potentials of the equipment they offered when coordinating the requirements (specifications) to devices; as a result, there were faults of the monitors during tests.

Photomultiplier (PHM) of one of the four detector units of RM-700 MHS failed, and it took considerable time to analyze the causes of the PHM failure and replacement with VNIIA, LANL, ORNL, and TSA System Ltd. involved.

Under tests for transport impacts, monitor JPM-21A showed numerous faults and mechanical damages power supply sources, scintillation detector, machine screw). Tests for detecting sensitivity showed the JPM-21A not to meet the II category requirements (10 g HEU and 0.29 g Pu of low burn-up). As a result of long correspondence and analysis of causes of JPM-21A failures with representatives of Canberra, LANL, and VNIIA, it was confirmed that the JPM-21A presented does not meet the above-stated requirements. It turned necessary to spend a considerable time to the instrument repairs, detalizing the test program (scope of the parameters to be certified) with PNNL and with the certifying authority of "Eleron".

TABLE 2. Completeness of the information in the accompanying documents for monitors

Major parameters which must be available in documents	Pedestrian monitors		Portable monitors	
	PM-700SP	JPM-21A	PRM-470A	JHH-21
1. Russian version of the documents	+	+	+	-
2. List of items in the set	+	-	+	-
3. Monitor design parameters:				
- dimensions;	+	-	+	-
- mass;	+	-	+	-
- scheme of internal connections.	+	-	-	-
4. Major specifications:				
- sensitivity;	+	-	-	-
- time of continuous operation;	-	-	-	-
- maximum external background admissible;	-	-	-	-
- frequency of false alarms;	+	-	-	-
- time of operation start after power supply begins	-	-	-	-
- electrical safety;	-	-	-	-
- fire safety;	-	-	-	-
- operation conditions;	-	-	-	-
- conditions of transportation;	-	-	-	-
- storage conditions;	-	-	-	-
- warranty period.	+	-	-	-

Note: "+" - parameter present, "-" - parameter absent.

TSA System Company provided with the monitor the user's guide and maintenance instructions for monitor PM-700SP in English and user's guide and maintenance instructions for model PM-700 in Russian, which do not coincide.

Since completing parts necessary were not available in Russia, the process of monitor restoration took a long time, which resulted in delays with the tests.

In the course of tests, it was proposed to cancel the pedestrian monitor testing for transport-related loads; testing monitor JPM-21A for its accordance with sensitivity category 1 requirement (64 g HEU and 1 g Pu) was suggested.

Based on the results of certification tests, the Certification authority for engineering security support of "Eleron" facility, certificates were provided for monitors JPM-21A (POCC US OSO2.C00408) PM-700SP (POCC US OSO2.C00407), PRM-470A (POCC US OSO2.C00338), JHH-21 (POCC US OSO2.C00337) confirming the compliance with specifications stated, with changes in the test scope accounted for.

## CONCLUSIONS

1. Results of the tests have confirmed the expedience of certification of the equipment, especially imported items.
2. For the use of imported equipment at Russian facilities, it is justified to accommodate the accompanying documents thereto for the Russian standards.
3. Equipment items shipped to Russia should be prepared for the certification tests and/or operation.
4. When taking decisions on the certification of imported equipment, it is expedient to preliminarily estimate the instruments' parameters and its operability in Russia.
5. To solve the question whether the imported equipment is usable for Russia and what engineering support and maintenance is needed for its operation, it would be justified to

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create the Center for engineering support of instruments to be used for MPC&A on the basis of one of institutes dealing with the development of instruments for these applications, with functions and tasks as follows:

- providing the operating manuals in Russian in accordance with the RF standards;
- estimation of parameters and possibility of using the instruments in Russia;
- reception verification of instruments after their transportation to Russia before their certification or installation at the facility it is intended for;
- maintenance (calibration) of the instruments in the course of operation (by facilities' applications);
- repairing equipment using the completing parts shipped from the USA;
- training specialists from nuclear facilities (by their applications) for local maintenance of instruments.

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