LÉON VANDEVELDE

NUCLEAR CHEMISTRY AND SERVICES

Background

The main mission of the department is to support internal R&D programs by providing on request the necessary chemical and radiochemical information to fulfil their objectives. We also perform analyses for some external clients, particularly in the field of nuclear materials.

Own R&D projects are generally restricted to the testing and the improvement of sample preparation techniques. Whenever possible, these developments are realised in the frame of interlaboratory comparisons.

A fundamental approach of a more sophisticated technique like Inductively Coupled Plasma Mass Spectrometry (ICP/MS) is carried out in a PhD work.

Objectives

The primary objectives of the department are to support R&D programs by delivering in time the most appropriate information about the chemical or radiochemical characteristics of the investigated materials or processes. This supposes the availability of efficient analytical instruments managed by experienced operators. We therefore aim to a continuous upgrading of the instruments performances and a corresponding training of the operators.

In order to control and improve our competence, we participate to various proficiency testing schemes and other interlaboratory comparisons.

The activities of the service are governed following the QA-principles prescribed in the ISO 17025 norm. The extension of the accreditation by BELTEST remains one of our main objectives.

Programme

ICP/MS (Inductively Coupled Plasma Mass Spectrometry) is a powerful instrument for the elemental and isotopic analysis of samples. The low detection limits that can be reached in solutions, make it a possible alternative technique to SSMS (Spark Source Mass Spectrometry) for the determination of impurities in nuclear fuel.

One of the major drawbacks of a nuclearised version of the ICP/MS instrument is the modification of the geometry of the sampling interface to pass through the wall of the glove box. As a consequence the sensitivity of the system is poorer than previously expected. The manufacturer has developed a new design for this interface but its replacement implied to work temporarily with an open glove box and thus required a careful preparation to avoid accidental contaminations and to respect the ALARA principles.

We planned to organise, together with the Institute for Reference Materials and Measurements (IRMM Geel), a workshop on the "Thematic network on the analysis of thorium in workplace materials". The goal of the European network is to determine, through the participation of different European laboratories, the most appropriate methods for the determination of thorium in materials used in the workplaces and possibly to find an explanation to some incoherences experienced in a previous interlaboratory exercise.

Although the last results have already been presented at the end of last year, the final report for the ARIANE project (Actinide Research In A Nuclear Element) must be delivered during the first quarter of the year. This project co-ordinated by BELGONUCLEAIRE aimed to improve the source term prediction for high burnup fuels.

The main concern for a safe handling and storage of radioactive wastes implies a better knowledge of their characteristics. This involves the control of the proficiency of the analytical methods by means of intercomparisons.

The interlaboratory exercise for the analysis of several radionuclides in a reactor water concentrate and a resin sample was set-up last year but started actually this year with the delivery of the samples. The full title of the project co-ordinated by CIEMAT is "Interlaboratory Radiochemical Analysis Comparison on a Primary Waste Flux". Eleven different laboratories from eight European countries participate to the exercise.

In the frame of the characterisation of special wastes, we also have to carry out a chemical and radiochemical characterisation of an irradiated beryllium matrix originating from our BR2 reactor.

In order to extend our analytical possibilities to the characterisation of glass embedded high level wastes, we started experiments with mock-up samples.We prepare an analytical scheme that can later be applied for the analysis of COGEMA glass.

The continuation of the Institute of Research and Innovation (IRI; Japan) project (tests on an advanced ion-exchange resin for the reprocessing of nuclear fuel) implied also numerous analyses of actinides and fission products in feed solution and separated

Scientific staff

MIREILLE GYSEMANS,
EDUARD HELSEN,
PETER THOMAS,
MARLEEN VAN BOCXSTAELE,
LUDO VANDENWEYER,
FRANK VANDERLINDEN,
LÉON VANDEVELDE,
KAREL VAN GILS,
STEFAAN VAN WINCKEL,
FRANS VEN

Supporting staff

ERIK AEGTEN,
LUC GELENS,
GUSTAAF GENEN,
PATRICK LYCKE,
WENDY MENTENS,
MAGDA OOMS,
ANDRÉ STUBBÉ,
KAROLIEN VAN ROMPAEY,
KURT VERHEYDEN,
ELS VERHEYEN,
HENNIE WARMOES

fractions. Most of the analyses must be performed by ICP/MS.

For the RETROSPEC project (retrospective dosimetry based on niobium impurities), we had to investigate several chemical techniques to separate niobium from structural reactor materials and activation products.

Since the introduction of the QA-system and our accreditation by BELTEST, most of the techniques which are more or less routinely used, have been validated and accepted within the scope of accreditation. This responds to a demand from internal as well as from external clients.

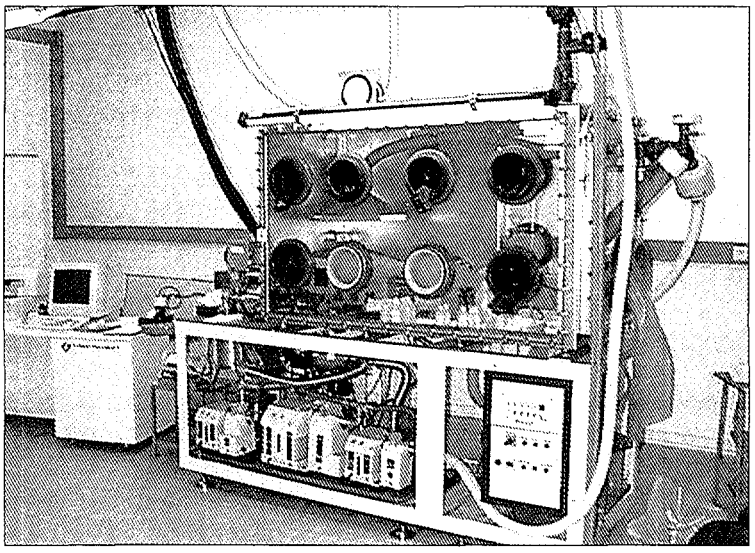
We intended to broaden the scope of accreditation with two new methods: Total Organic Carbon (TOC) analysis and Ion-Selective Electrode measurement (ISE) for fluoride determinations. Both methods are extensively used for waste characterisation and migration studies.

Beside these more specific objectives, we have to provide the usual analytical support to internal and external clients.

Achievements

We successfully replaced the interface of the ICP/MS in glove box respecting the ALARA principles.

We also carried out additional minor interventions on a number of electronic components of the system in order to decrease the detection limits. The first preliminary measurements on a dissolved uranium oxide reference material certified for its impurity content delivered promising results.



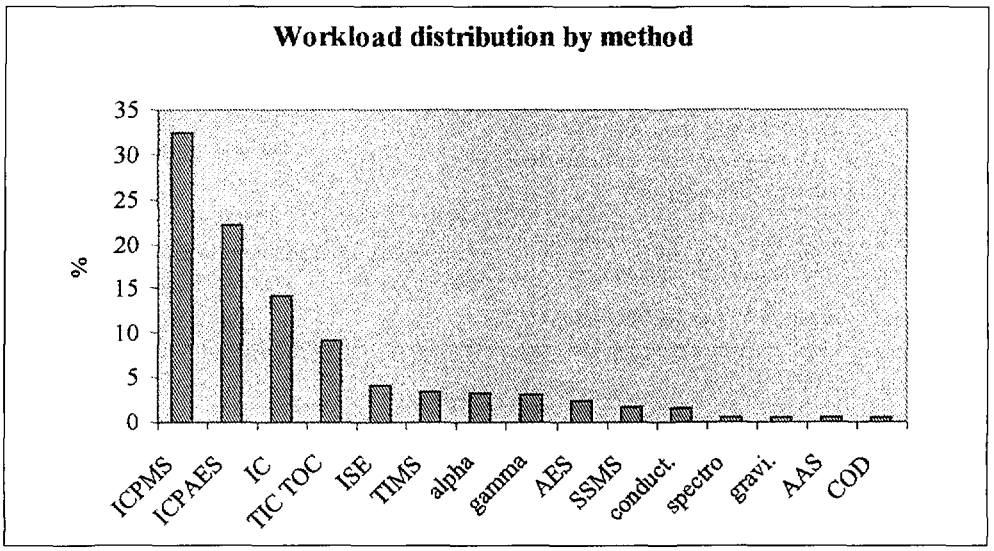
The nulearised version of the ICP/MS after the latest improvements

The "1st European Workshop on the Analysis of Thorium in Workplace Materials" was held on March 27 and 28. About seventy participants took part to the discussions around the results obtained by the network laboratories and the subjects presented at the poster session. New approaches for sampling techniques and for reference materials aiming to a better understanding – and solution – of the problem were suggested during the meeting. The publication of the proceedings is foreseen in 2002.

The final version of the report about the ARIANE project was approved. The recognition of the quality of the work performed by the laboratory has led to the preparation of new similar projects. We performed preliminary dissolution tests for the planned burnup analyses of uranium silicide and aluminium-uranium fuel in 2002.

The resin and reactor water concentrate samples originating from ENRESA were delivered during the second quarter of the year. We immediately started the dissolution and separation processes to determine the content of ^{3}H, ^{14}C, ^{55}Fe, ^{63}Ni, $^{89/90}Sr$, ^{238}Pu, $^{239/240}Pu$, ^{241}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm, ^{234}U, ^{238}U. At present we have measured all the gamma-emitters. The second progress meeting of the project was held at SCK•CEN on 5-6 July 2001.

We chemically and radiochemically characterised beryllium material originating from the second matrix of the BR2 material testing reactor. We dissolved the samples in sulphuric acid and analysed the solution for the presence of impurities by ICP/MS. Gamma emitting radionuclides were determined by using a n-type HPGe detector previously calibrated with a mixed radionuclide source. Due to the very low abundance of ^{94}Nb, a separation was necessary to lower the background level. The measurement of ^{244}Cm could be performed by alpha-spectrometry without any separation from the dissolved matrix. The determination of plutonium necessitated a prior purification on a strong anion exchanger before the alpha-spectrometry measurement. Pure beta-emitters like ^{90}Sr and ^{63}Ni were measured by liquid scintillation counting (LSC) after separation by extraction or ion exchange chromatography respectively.



In the frame of the characterisation of COGEMA vitrified waste, we undertook exploratory tests with inactive glass of the same type. For the analysis of most of the elements, the sample must be destroyed by fusion and further dissolved in acid. In order to evaluate the possibility to recover some specific radionuclides like ^{79}Se, ^{93}Zr, ^{107}Pd, ^{126}Sn we applied this treatment to inactive glass powder mixed with oxides of selenium, zirconium and tin and with a chloride solution of palladium. We obtained rather good recoveries ranging from 82 to 95 percent with a borax smelt, except for palladium. Since none of the materials used was radioactive, all the measurements were performed by ICP/MS.

For the IRI separation experiments, we analysed about 150 elution fractions for their content of uranium, actinides and fission products in order to obtain the separation profile.

We carried out preliminary experiments on a few samples in the frame of the RETROSPEC project. We tested three methods to separate Nb by using an anion exchange resin. They all gave valuable results.

The renewal audit of BELTEST took place on June 12 and 13 and on September 19.

Both the TOC and the ISE methods could be added to the scope of accreditation. The auditors only formulated minor recommendations, which were immediately met. The conclusions of the audit were all in all very positive and the renewal of the accreditation has been granted.

Besides these specific actions, we also responded to the daily demands of internal and external clients.

The total number of samples increased by more that 40 percent as compared to 2000.

Most of the internal demands arose from projects dealing with radioactive wastes: waste characterisation and migration studies represent respectively 10% and 31% of the requests.

Demands from the radioecology increased significantly from 5 percent in 1999 to 21 percent of the total.

The number of demands arising from external clients remained at the same level as past year.

We also refined some sample preparation methods like the UV-digestion for the treatment of clay water and automated instruments like the ion chromatograph with a sample changer.

A new method was validated for the routine analysis of anions by ion chromatography. It increases the range of measurement and allows bypassing the previous level controls. We also developed methods for specific anions like formate, acetate, tartrate, oxalate and thiosulfate that are important for the analyses of clay water.

The hot-cell for chemical applications that serves principally for the preparation of samples for radiochemical burnup determinations, is more than 25 years old and will be replaced in order to meet the new analytical needs. We removed all the sample rests and carried out the necessary maintenance to get it in standby.

Perspectives

Many burnup determinations are planned in the near future. The LHMA laboratories will co-ordinate these projects, execute the post-irradiation examination and prepare subsamples for further destructive radiochemical analysis in our laboratory.

The FUTURE programme concerns the irradiation in the BR2 reactor of a uranium-aluminium fuel.

Other projects like GERONIMO, REBUS or the burnup analysis of a U_3Si_2 fuel are planned in 2002 or at the beginning of 2003.

We received a new call for bid for an "ARIANE-like" characterisation programme on 4 UO_2 and 4 MOX samples to be started in 2002 and achieved in 2003.

Another new call for bid for the determination of critical nuclides in nuclear reactor resins, concentrates and filter was also received. The analyses should start in 2002.

In the frame of the characterisation of COGEMA vitrified waste, we will perform scale analysis on an inactive sample. This will include the pre-treatment in a hot-cell of LHMA.

We expect many analyses requests arising from the dismantling of the PRACLAY maquette.

A new staff member joined the department for the daily exploitation of the ICP/MS instruments. After his qualification, he will be in charge of the validation of some routine analyses to allow their accreditation by BELTEST. We will validate the nuclearised version for the determination of impurities in MOX fuel.

-	University of Innsbrück
-	Ith & Safety Laboratory Sheffield (Sheffield, UK)
CEA	Commissariat à l'Energie Atomique (Sanclay, France)
FZ	Forschungszentrum Jülich (Jülich, Germany)
ENEA	Ente Per Le Nuove Tecnologie, l'Energia E l'Ambiente Saluggia (Brasimone and Bologna, Italy)
IRMM	Institute for Reference Materials and Measurements (Geel, Belgium)
NPL	Universiteit Gent –National Physical Laboratory Teddington (Teddington, UK)

none

-	BELGONUCLEAIRE (Dessel, Belgium)
-	FBFC International s.a. (Dessel, Belgium)
-	Electrabel Doel (Doel, Belgium)
-	Electrabel Tihange (Tihange, Belgium)
-	MDS Nordion s.a.
IRE	Institute Radio-éléments (Fleurus, Belgium)

Presentations

M. Van Bocxstaele, "Interactions of Metrology in Chemistry with accreditation", training course: "Principles and Applications of Metrology in Chemistry" (TrainMiC), organised by IRMM, Maribor, Slovenia, October 11-13, 2001.

M. Gysemans, F. Ven, E. Aegten, "Werken in alfakast", training course: SCK•CEN, September 11-12, 2001.

Reports

S. Van Winckel, M. Gysemans, R. Boden, D. Huys, P. De Regge, L. Vandevelde, "ARIANE International Programme: SCK•CEN Results", R-3504, February 2001.