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FOREWORD

Approximately 160 gamma irradiators and 1200 electron accelerator based processing units are in operation worldwide. In recent years the IAEA has prepared a directory of industrial gamma irradiators and held several meetings on developments in radiation technology applications. Developments involving the engineering of new sources (both isotope and electrical), high power accelerator applications, etc. have been reported recently, making a review and evaluation of this progress timely. Therefore the IAEA organized a technical meeting in Vienna, Austria, from 28 to 30 April 2003 to review the present situation and the potential contribution of radiation technology to sustainable development. Engineering developments and other features of radiation sources, both isotope and accelerator, were discussed.

Recent research has concentrated on three fields: medical and food products, polymers, and environmental pollution control. The stability of radiation sterilized medical implants, as well as the uses of radiation processing for sterilization or decontamination of pharmaceuticals and pharmaceutical raw materials, radiation synthesis and modification of polymers for biomedical applications have been studied. Since separation and enrichment technologies play an important role in product recovery and pollution control, the possibility of radiation synthesis of stimuli-responsive membranes, hydrogels and adsorbents is being investigated. Finally, aside from the technologies for flue gas and wastewater treatment already in use, further research is ongoing on the treatment of organic contaminants in both gaseous and liquid phases. Environmental applications, which also offer new opportunities, should be carefully reviewed to reflect existing regulations and current knowledge. The increasingly serious problem of polyaromatic hydrocarbons (PAH) emissions may be solved in part by the application of radiation technology. This is being studied on a pilot scale for the removal of dioxins and furans from flue gases emitted by municipal waste incinerators.

This report will be of value to research groups working in the field of radiation technology development and is meant to focus research on the promising fields of radiation processing. Developing Member States with radiation technology programmes will benefit from research in this area.

The IAEA wishes to thank all the participants in the technical meeting for their valuable contributions. The IAEA officer responsible for the organization of the meeting and for this publication was A.G. Chmielewski of the Division of Physical and Chemical Sciences.

EDITORIAL NOTE

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SUMMARY

Background

Radiation processing has been widly accepted for use in many areas of the global economy. Sterilization, polymer cross-linking (tapes, tubes, cables), tire component curing, the conservation of art objects, the irradiation of selected food items are well established technologies. Both types of irradiators, gamma sources and electron accelerators, are used in these processes.

The IAEA through its technical co-operation programmes, co-ordinated research projects, consultants and technical meetings, conferences promotes the peaceful use of radiation technologies. Because of the IAEA's support, some new technologies (such as hydrogel wound dressings, sulphur-free radiation vulcanized latex) were developed and transferred to Member States over the previous ten years. Other promising applications the IAEA has supported are flue gas and wastewater treatment. At an industrial fossil fuel power plant, electron accelerators exceeding 1 MW have been installed to remove SO_x and NO_x from flue gases — a further breakthrough in the large scale use of electron beam technology.

The Advisory Group Meeting on New Developments in Radiation Technology Applications was held in 1998 in Budapest, Hungary. The developments of the *fin de siècle* were reviewed there and a follow-up meeting has been held.

At the beginning of the 21st century, new science and technology development programmes are being defined and implemented, including UN resolutions concerning sustainable development, the Johannesburg protocol, the 6th EU Thematic Framework and others. Thus, the IAEA in response to organized a technical meeting (TM) in Vienna, Austria, from 28 to 30 April 2003, to review the present situation and possible developments of radiation technology that can contribute to a sustainable global environment. Scientists from noted laboratories in Austria, Canada, Egypt, France, Germany, Hungary, India, Rep. of Korea, Japan, Malaysia, the Russian Federation and the United States of America participated in the technical meeting. Reports describing some specific research work in these countries are included herein.

Trends in research and development

Since 1998, the IAEA has been organizing many meetings and co-ordinated research projects in which trends and new developments have been discussed and technical work conducted. Recent developments in the field were discussed by representatives from industry, universities and research institutes during the International Meeting on Radiation Processing, held in Avignon, France, in 2001; the last such meeting was held in September 2003 in Chicago, USA. New developments were also reported during the symposium on "Radiation Technologies in Emerging Industrial Applications" organized by the IAEA in Beijing, China, in 2000. More than 200 participants, representing over 30 countries attended that symposium.

The main research and implementation of developments presented at these meetings concentrated on three fields: (1) medical and health related applications, (2) environment pollution control, and (3) polymers. Post-irradiation stability of radiation sterilized medical implants, the use of radiation processing for the sterilization or decontamination of pharmaceuticals and pharmaceutical raw materials, radiation synthesis and modification of polymers for biomedical applications have been studied. Since separation and enrichment

technologies play a very important role in product recovery and pollution control, the possibilities of radiation synthesis of stimuli-responsive membranes, hydrogels and adsorbents are being investigated. A new horizon opens up with the possible application of radiation processing of polysaccharides. Equipment and study concerning post-irradiation effects on polymers can lead to the basis for new end-use applications. Finally, although radiation processing is already an applied technology for flue gas and wastewater treatment, additional research is going on using radiation treatment of organic contaminants in both gaseous and liquid phases. Reports on the development of radiation sources mostly involve high power electron beam accelerators.

Purpose and discussed topics

Developments concerning engineering and other features of radiation sources, both isotopes and electron accelerators, were discussed. Such developments are very important for the emergence and implementation of new applications. Reductions in costs and improvements in technical reliability are expected, especially where high power is needed for environmental applications. Low energy, self-shielded accelerators for surface curing is another equipment area which may find growing application.

Accreditation procedures, rising demands concerning product and service quality are very important nowadays and quality assurance (QA), dosimetry and operation safety were addressed as well.

Environmental applications, another field which opens new opportunities, should be carefully reviewed in accordance to the existing regulations and state-of-the-art knowledge. The rising problem of the emission of polyaromatic hydrocarbons (PAHs) may be partly solved by application of radiation technology. This is being studied on a pilot scale for the removal of dioxins and furans from flue gases emitted by municipal waste incinerators.

Reports on new technologies related to natural polymer processing, composites, biocompatible and nano-materials and others were presented. The technique of radiation-initiated polymerisation and cross-linking of monomer solutions offers rapid and simple in-situ preparation of tailor-made polymer matrixes and nanostructures. Since ion and electron beams were amongst the first tools used in nano technology (semiconductors, atom doping, surface treatment, micro gels), the role of these techniques in many foreseen developments could be immense. Besides synthesis and cross-linking, degradation mechanisms ought to be given greater consideration. Further attention should be paid to research and development involving inorganic systems, in which gamma irradiation techniques could have some advantages.

In the case of well established radiation technologies, still new developments are possible, such as pharmaceutical sterilization, natural herb decontamination and tissue graft banking. Progress in this field can be achieved through closer collaboration with microbiologists, medical doctors, pharmacists, and agro- and food specialists.

Public awareness and technology acceptance are other factors to be considered to enhance greater understanding of the societal merits of radiation processing.

Conclusions and recommendations

Industry developments: X ray processing for the sterilization of medical devices and treatment of foodstuffs is now a reality. X rays generated from 5.0 and 7.5 MeV electron accelerators have shown remarkably uniform dose distribution. The use of X ray treatment of food and for medical device sterilization warrants global cooperation as industry itself is now addressing some existing limitations in certain countries on the beam voltage permissible for use with X ray conversion. This technique can also open up new product areas, permitting the crosslinking of polymers and composites of significantly greater thickness. High current, high voltage beams also improve the economics of radiation processing in high volume, high through-put environmental applications such as the treatment of liquid effluents and waste streams.

Materials developments: The use of radiation processing to enhance the commercial value of materials is well practiced. Some are under exploited and innovative areas may require global cooperation. Polymer reinforcement with nanometer-scale particles was shown to provide unique high strength, light weight materials. While known to some, the radiation grafting of polymers has been little developed. Such technology can, for example, be used to alter the filtration properties of non-woven or fabric materials. In some specific areas, such as the radiation treatment of cellulose and polypropylene, greater understanding of degradation mechanisms and effects on resultant material properties should be explored. In developing countries, reliance upon regional resources that can be enhanced with radiation processing should be encouraged. To complement this, further investigation is needed into the effects of irradiation on natural resources that can be fashioned into marketable products, such as biodegradable materials.

Compact equipment: Industry has responded to the need for more accessible facilities in a variety of ways. The development of smaller scale gamma irradiators can permit this proven technology to be used on a more regional basis or in more remote areas. A compact, low voltage modular electron beam renders the use of electron beam processing to dry/cure inks, coatings and adhesives more affordable and now useful to a broader range of global interests. Transportable accelerators on vans are being explored. These are intended for environmental remediation and take electron beam processing to site specific problem areas.

Food safety. The acceptance of the use of radiation processing for the treatment of foodstuffs varies throughout the world. However, some regions, such as the European Union, seem reluctant to adopt some well accepted practices of the radiation treatment of certain foodstuffs, such as spices, even though elsewhere, as in North America, such irradiated products are in common use. Harmonization of diverse regulatory requirements in the area of food irradiation and, in particular, when dealing with spices should be fostered. In developing countries, food irradiation is one of the most viable means for preventing post-harvest waste of foods getting to market. In such areas, the means of transport, such as the use of refrigerated trucks on well paved roads, may not be that common. The successful radiation degradation of polysaccharides, such as alginates, that are being used to enhance the growth rate of rice that is now being sold in Vietnam, looks promising with other food plants. Such radiation treatments of natural materials that can benefit crop growth warrant greater investigation. Improving the growth rates of crops would help alleviate food shortages in many parts of the world. However, as with genetically modified foods, such resultant crops will be subject to considerable public scrutiny, even if no scientifically based differences with traditional foods may be found.

Computational methods and dosimetry: A number of codes and computational methods have been developed that facilitate the prediction of dose distribution, even in containers filled with complex products of varying densities. Improved training techniques and international collaboration is needed so that the most reliable codes can be readily used by all. Some simplification of access and minimization of computer demands is needed. Also, dosimetry systems that are widely used to indicate the extent of product exposure to radiation need to be linked into total measurement quality assurance programs to be used by the practitioners of radiation processing.

Safety concerns: The radiation processing industry needs to review existing protocols for facility safety and explore mechanisms to assure that these and any modifications are fully implemented. Such risk assessment or 'total safety analysis' protocols enable those who construct radiation processing facilities and then the subsequent users of such to employ the best possible means of worker protection. Not only must facilities be designed with safety given paramount concern, but all other operational aspects of a radiation processing facility must be taken into account. Any uniform code of practice in this area requires follow-through in terms of implementation and assurances of practice.

Technology transfer: Recognition should be given to the role that well-managed radiation processing facilities play in promoting and expanding the use of this process technology. Such facilities minimize the capital costs and risk for potential end-users while introducing such users to the benefits of radiation processing. Sustained support for the development of full scale facilities for environmental projects, such the treatment of potable water and wastewater, is crucial to bringing forth the societal benefits of this technology.

Education: The co-ordination of educational programs is needed so that some minimum knowledge base is available to all those engaging in radiation processing. This should be done in conjunction with the academic community and undertaken on a global basis. Given the complexity of source technology, materials use and product capabilities, this requires a high level of inter-disciplinary guidance and interaction. Illustrative of this approach has been national activities in some countries. Other institutions that have irradiation processing capabilities need to be networked together to bring forth a more coordinated global effort.

Public awareness: There remains a crucial need for greater public awareness of the environmental and societal benefits of radiation processing. Successful projects involving the use of radiation processing to clean up flue gases by removing undesirable emissions, to eliminate volatile organic or solvent emissions from coatings by use of low voltage electron beam processing, and to enhance the safety of the food supply and its wholesomeness remain relatively little known outside of our community. In addition, the use of radiation processing for water and wastewater treatment warrants greater public support. Such wastewater treatment facility designs must take into account that in many developing countries heavy metals must also be eliminated, perhaps by chemical or filtration means, to render potable water or water suitable for irrigation. The radiation processing industry is well aware of the continued developments in alternative technologies, such as improvements in the use of ETO for medical device sterilization and in diverse chemical processes. Also a listing of alternatives now being used, such as the toxic substances used to fumigate and disinfect foodstuffs, would make the public more aware of the benefits of radiation processing.

ROLE OF RADIATION PROCESSING FOR SUSTAINABLE DEVELOPMENT

S. Machi

Japan Atomic Industrial Forum, Inc., Japan

Abstract. Application of radiation and isotopes is now recognized to be more environmentally friendly in comparison with chemically initiated or assisted process. There are several areas which are of importance for future development of radiation processing applications: removing pollutants from gaseous and liquid wastes, new products and new processes, radiation processing replacing or modifying chemical process to be more environmentally friendly, transfer of radiation processing application to developing countries. Safe handling of radiation facilities and physical protection of radioactive materials are prerequisite of the application of radiation technology. Electron accelerator design and production have well developed in terms of reliability, larger capacity, wider energy range and cost reduction, which are enhancing the applications. This paper covers progress on radiation processing applications with regard to the above mentioning points.

1. INTRODUCTION

1.1. Economic Scale of Radiation Technology in Japan

Total economic scale of nuclear power and non-power (radiation and isotope) application in Japan in 1997 is about US\$99 billion, out of which 52% is from the radiation and isotope application and 48% from nuclear power industry. In the United States the economic scale of radiation and isotope technology is US\$119 billion and that of energy is US\$ 39 billion. The scale of nuclear energy in the US is smaller than Japan because of zero construction of nuclear power plant after the TMI accident.

The economic scale of radiation and isotopes is estimated based on applications as follows:

industry: sterilization of medical products, automobile radial tires, NDT (radiography), semiconductors, medical equipment

agriculture: mutation breeding, food irradiation

medicine: diagnostic imaging, PET, prostate cancer treatment, radio-pharmacy, radiation medicine equipment

The components and economic scale in the application of radiation and isotopes (US\$52 billion), which accounts for 1% of total GDP of Japan are: industry US\$39 billion, medicine US\$12 billion and agriculture US\$1 billion.

TABLE I.ECONOMIC SCALE OF RADIATION APPLICATION IN INDUSTRY IN JAPANAND USA

	Economic scale (bS)		Presto
rtem	U.S.A.	Japon	Isano
(1) Sterilized medical supplies	4.8	2.3	2.0
(2) Semiconductors	37.2	28.4	1.3
(3) Accelerators installed during 29 years	648	308	2.1
(4) Radiographic testing (NDT)	0.65	0.26	2.5
(5) RI application & radiation protection	Not found	0.54	-
(6) Radiation cured radial tires	13.5	8.4	1.6
Partial sum: (1)+(2)+(4)+(6)	56.2	39.4	1.4

Note: Comparison for (3) and (6) were based on North America, 15=1215

1.2. Economic Scale of Radiation Application for Industry

The Table 1 shows economic scale of radiation application in industry in Japan and USA. It is noted that the application of EB for lithography and ion implantation for semiconductor production is the largest component (72% in Japan and 66% in the US).

Production of automobile radial tires using EB cross-linking of rubber sheet is the 2nd largest components in both U.S. and Japan. Five major tire companies are using 23 EB machines for the production of tires in Japan. The economic scale of radiation cross-linked wire and cable and heat shrinkable material which are not included in this survey might be additional components.

2. DEVELOPMENT OF NEW PRODUCTS

Major applications of radiation processing have been the production of polymeric products as shown in Table 2 by using radiation cross-linking and grafting reactions.

TABLE II. COMMERCIALLY PRODUCED CROSS-LINKED OR GRAFTED POLYMERS BY RADIATION Commercially produced cross-linked or grafted polymers by

Products	Applications
Cross-linked polyethylene and PVC	Wire insulation teristant to heat and chemicals, paper for heating systems
Formed polyethylenc	Institution, packing, Booting, materials
Hear shrinkable robuiks and sheets	Food packaging, insulation, protection against corrorion
Cross-linked robber riseets	Automobile tires (high quality), coof protection sheets (weather resistant)
Cross-lucked polymeritane (Japan)	Cable mendation for anniock brake sensor
Cress-lanked nylon (Japan)	Automobile parts needed to heat and chemicals
Super heat ceremont SEC fibre (Jupan)	Metal and ceramic composites, semi- commercial plant
Vulcanized metural reflect later. (Malaysia)	Surgical glovna, condoma
AA grafied PE film (Japan)	Bahtery saparator
Cross-Inded hydrogel	Worland dates sturg
Curing of paints and take	Surface conting and printing
Grafted filter	Deadacant

Radiation cross-linking is an excellent method to improve thermal, chemical and mechanical properties of polymeric products in solid state without using chemical catalysts at room temperature. The processing is clean, simple, fast and energy saving. These merits have large potential for new products in horizon.

2.1. Radiation cross-linked PTFE

PTFE has high temperature resistance and low surface tension. At ambient temperature and atmosphere PTFE degrades by radiation. However, Seguchi found that PTFE cross-links by EB at the temperature just above its melting point and with high dose in inert atmosphere.

The cross-linked PTFE has much better mechanical properties in particular higher abrasion resistance by 1000 times than a non-cross-linked one. Therefore, it can be better used as moving parts without lubricant oil. A semi-commercial plant to produce this new PTFE product is in operation in Japan.

2.2. Polyethylene fiber radiation grafted by acrylamide

Non-woven polyethylene cloth is radiation-grafted with acrylamide to be converted into amidoxime group. The product is very efficient adsorbent for uranium and other rare metals in seawater. As shown in Fig. 1. large scale experiment has been performed in the sea near Aomori using 120 kg of absorbent to collect 1000g of yellow cake of Uranium.



FIG. 1. Pilot plant of rare metal recovery from sea water using adsorbent produced by radiation processing.

2.3. Radiation-crosslinked hydrogel for medical use

Hydrogel wound dressing prepared by radiation crosslinking was first commercialized in Poland and Israel. Radiation has been proven to be an excellent method because of non- chemical crosslinking agent and catalyst of which residues cause skin irritation.

In Japan recently the hydrogel sheet preparation by using EB of 800 keV has been demonstrated as schematically shown in Fig.2. Polyvinyl alcohol and other water soluble polymers are coated on plastic film to be irradiated with 10-20 kGy for crosslinking. PVA has been selected as the most appropriate polymer because of high mechanical strength and water swelling ratio.



FIG. 2. EB Process for Hydrogel Wound Dressing.

Healing evaluation animal tests were shown in Fig. 3 indicating much better healing rate in hydrogel than gauge.



FIG. 3. Evaluation test of radiation cross-linked hydrogel.

The clinical tests have been successfully finished and government approval for clinical use will soon be given.

The IAEA-TEC DOC 1324 published in 2002 compiles reports on radiation processing application for biomedical material including hydrogels by Rosiak (Poland), Sen (Turkey), Hegazy (Egypt), Nurkeeva (Kazakhstan) and Razzak (Indonesia). Hydrogel has large potentials as biomedical material and will be extensively used in the future.



FIG. 4. Production of high heat resistant SiC fibre

2.4. Radiation-cross-linked SiC fiber

Silicon carbide fibre cross-linked by thermal oxidation followed by pyrolysis can stand at 1,200°C. The JAERI and Nippon Carbon Co. have developed new cross-linking process using EB irradiation of 10MGy by continuous demonstration plant of 1 ton per month capacity. The product has much higher heat resistance (1,700°C) than thermally cross-linked one. It will be used for ceramic matrix composites suitable for space plane materials, turbine blades, and engine materials. These applications are under development. Currently, annual production of this new SiC fibre is still 2.4 ton.

2.5. Radiation modification of natural polymers

The IAEA/RCA has been implementing the CRP on radiation processing of natural polymers. In October 2001 the JAERI hold "Takasaki Symposium on Radiation Processing of Natural Polymers". A few interesting application are highlighted in this paper.

2.6. Plant growth promotion by radiation degraded carbohydrates

Alginate degraded by dose of 100kGy to 500kGy has remarkable effect on growth promotion of rice. Field test in Vietnam for tea, carrot, and cabbage also showed the field increase of 15-40% by this promoter. Similar growth promotion effects were observed by degraded chitosan, carrageenan and lignocellulose's extracts. The plant growth promoter produced by radiation processing of chitosan is already in commercial market in Vietnam.

2.7. Anti-microbidal activity of radiation degraded chitosan

The chitosan irradiated at 0.8kGy in N₂ inhibited the growth of *E. Coli* effectively. Antifungal effect was also induced. Fig. 5 shows mango coated by irradiated chitosan can be stored 12 days while control is spoiled. The mango coated by unirradiated chitosan could not ripe. This technique should be useful in tropical countries.



FIG. 5. Effect of radiation-degraded chitosan on mango fruit.

2.8. Radiation croos-linking of natural rubber latex

The IAEA/RCA project developed the technology of radiation cross-linking of natural rubber latex with good cooperation of the member states.

At the Malaysian Institute of Nuclear Technology Research (MINT) about 50 tons of natural rubber latex irradiated annually by Co-60 source with continuous flow system for commercial purpose. The JAERI has studied the application of low energy EB for cross linking of natural rubber latex in 18 litre reaction vessel with stirring at 210 rpm.

The products have several advantages over chemically cross-linked products because of the absence of chemical cross-linking agents.

3. ENVIRONMENTAL PROTECTION

Applications of radiation processing for environmental protection has been extensively studied for cleaning, waste water, water, flue gases, and sewage sludge to meet the need of sustainable development. Environment is still degrading worldwide.

3.1. Cleaning flue gases by electron beams

The amount of emission of SO_2 is still increasing and will be 80 million tons in 2020 damaging environment. In China, for example, coal is the major primary energy source and 26 million tons of SO_2 is emitted in a year 37% of which is from power station.

The EB technology to remove SO_2 and NO_x simultaneously was first developed by Japanese group headed by Mr.Machi, being followed by research groups in Poland, U.S.A. Germany, China, Bulgaria, Brazil and Malaysia. It should be noted that the IAEA has also played an important role for the development of this technology.

At this meeting more detailed report on this technology application will be reported by Mr. Chmielewski, who is an important pioneer of it. Distinct advantages of this EB process are: simultaneous removals of SO_2 and NO_x , valuable by-products of fertilizer, applicability for high SO_2 content flue gases.

In Poland the INCT and Dolna Odra Power Co. have finally achieved this industrial operation of the plant to clean 270,000 Nm^3/h from power station with the strong support of IAEA and Japanese Government. Four accelerators of 800keV, 300kW of the Nisshin High Voltage are used with 2 power supplies.

In Changdu, China a flue gas cleaning plant shown in Fig.6 constructed by Ebara Co. and State Power Cooperation have successfully been operated for almost 3 years. The plant has a capacity of $300,000 \text{ Nm}^3$ /h and is installed with 2 accelerators of $800\text{kV}\times400\text{mA}$. Removal rate is more than 80% by irradiation of 3.2kGy for flue gases contain SO₂ of 500 to 2,400 ppm.

Another industrial plant has been completed in China for Hangzhou Power Company by Ebara Co. and is now under test operation. The plant has treatment capacity of $300,000 \text{Nm}^3/\text{h}$ using 2 units of EB machine of $800 \text{kV} \times 400 \text{mA}$ to remove more than 75% of SO₂ and 40% of NO_x when initial concentration of SO₂ and NO_x is 969 ppm and 200 ppm, respectively.



FIG. 6. A flue gas cleaning plant in Changdu, China.

Haigfeng Wu of Tsinghua University reported at the meeting in JAERI in December 2002 that 4 power plants have submitted the plant to the Government to install EB plant for cleaning flue gases. Many other power plant managements are actively evaluating this technology because in a few years the regulation for SO_2 emission will be stricter. In December 2000 INET (Institute of Nuclear Energy Technology) of Tsinghua University completed a newly designed pilot plant to clean flue gas using EB. The plant has the capacity of 10,000m³/h and demonstrated to remove 92-99% of SO_2 and 30-33% of NO_x at the lower dose of 1kGy. The process is semi-dry system as shown in Fig. 7 using wet type precipitator for the by-product of ammonium nitrate and sulphate.



FIG. 7. Semi-dry EB process for cleaning flue gas in China.

The IAEA Project in Bulgaria of the pilot plant construction and operation for cleaning flue gas of coal burning power plant containing as high as 5,000 ppm of SO₂ is being implemented by the support of the JAERI. The plant will be in operation in a few months after the delay. The plant capacity is $10,000 \text{ m}^3/\text{h}$ using 2 accelerators of 800kV donated by the JAERI.

The JAERI Takasaki has achieved the efficient removal of dioxins from the flue gas of municipal waste incineration plant by using a pilot plant of 1,000 m³/h capacity. Accelerator used is $300kV \times 40mA$ and dose given to the flue gas is up to 14 kGy. The plant was installed at waste incineration plant near Takasaki. More than 90% of dioxin (poly-chlorinated-di-benzo-paradioxins and poly-chlorirated-di-benzo- furans) was removed at 14 kGy.



FIG. 8. Pilot plant of cleaning flue gas of municipal waste incineration plant.

3.2. Radiation of Waste Water

A pilot plant to clean waste water from dyeing complex has been in successful operation in Taegu, Korea by EB-TECH Co. since 1998 in cooperation with Prof. Pikaev of Russia. The irradiation plant has the capacity of 1,000m3/day using an accelerator of $1 \text{ MeV} \times 40mA$. Waste water is irradiated under the beams as a thin layer curtain. Dose given to water is from 1 to 2kGy. Irradiated waste water is then treated biologically to decrease BOD and COD. By combining radiation and biological treatment dye molecules are more efficiently decomposed because radiolytic products of water such as OH radicals convert the molecules into bio-degradable structure.

The EB-TECH Co. has proposed an industrial scale plant based on the pilot experiments of which capacity is $20,000m^3$ /day using 2 units of 400kW (1MeV) accelerator. Total construction cost is estimated as US\$ 4-4.5 million. Treatment cost including construction and operation is calculated lower than that of UV and ozone.

4. RADIATION FACILITIES

4.1. Gamma and electron beam irradiators

Co-60 facilities are extensively used for sterilization of medical products and food irradiation. Some of the sterilization plants are using high energy (5-10 MeV) electron beams, which is more economical for large throughput volume. For most of industrial applications electron beam (EB) accelerators are more extensively used. In Japan about 300 EB accelerators are used out of which about 180 units are low energy accelerators less than 300kV. In North America about 650 EB units have been installed.

It should be noted that the very low energy EB accelerator in the range of 30 kV to 70 kV have been recently commercially available at lower price, which is enough for printing and coating of 10 to 20 μ m thick. The EB curing is clean and saving energy because no organic solvent evaporation is needed. Further more the quality of coating is better than conventional process. The production of such a low energy electron accelerator is now possible because of availability of extremely thin foil for beam window made of Si and Ti. For the high energy EB reliable and large capacity 10 MeV machines are available being used for sterilization of medical supplies and food irradiation. In general, accelerator has better public acceptance than Co-60 because of easy and safe switching off beam.

4.2. Ion Beam Applications

Ion beam is extensively used for the ion implantation of semiconductor. For example in Japan about 1,000 ion implanters are in operation. Radiation processing by ion beam is in development in some countries including Japan. Membranes with micro-pores are produced by the polymer chain scissor with ion beam irradiation followed by the chemical etching. The membrane is used for separation process. Crosslinking of polymer film by ion beams followed by chemical etching of non cross-linked portion can produce nanometre wires. This technology is being studied by the groups of JAERI and the Osaka University in Japan.

5. TECHNOLOGY TRANSFER TO DEVELOPING COUNTRIES

The IAEA is implementing some 50 TC projects to promote radiation processing technology and application in developing countries achieving remarkable outcome. For example, in Vietnam, industrial scale Co-60 irradiation plant was constructed 3 years ago to treat foods and medical products under the IAEA-TC project. The plant is now in full operation for 24 hours a day to irradiate frozen shrimps. Vietnam has a plan to install EB machine in the near future. The MINT of Malaysia is an excellent model to have achieved commercial application of radiation processing in the past several years by the support of IAEA and Japanese Government.

Since radiation processing has a potential for the expansion in the field of industry, environmental protection, health care, and foods which have large socio-economic impact in

developing countries, the IAEA should further promote R&D and technical cooperation for the application of radiation processing.

In view of technology transfer, the government of Japan has been implementing Scientist Exchange Programme countries for 16 years to invite about 1300 scientists and engineers from East Asia, China and Korea to stay in nuclear research institutes for 3-12 months for research and training. This programme is highly appreciated. The Forum for Nuclear Cooperation in Asia (FNCA) is another framework for multilateral cooperation funded by Japan and partly by Australia. The FNCA is implementing 11 projects in the field of agriculture, medicine, electron beam application, research reactor, nuclear safety culture, public information, human resources development and waste management. Every year workshop of each projects, the Coordinators Meeting and the Ministerial Level Meeting are held to exchange scientific information and to discuss policy issues on cooperation and nuclear policy. The projects are fruiting tangible results.

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PROSPECTS AND DEVELOPMENT OF RADIATION TECHNOLOGIES IN DEVELOPING COUNTRIES

K.S.S. Sarma

Radiation Technology Development Section, Bhabha Atomic Research Centre, Trombay, Mumbai, India

Abstract. Radiation processing has established itself as a commercially viable technology that can be beneficially used to enhance the characteristics of many materials. More than 200 gamma irradiators and over 1000 industrial electron beam accelerators are operating world wide in the established applications like medical product sterilization, polymer modifications and a few in number for food irradiation applications. Studies on economic feasibility have been carried out on industrial and semi-industrial scale, for emerging applications like flue gas treatment, industrial, domestic water and sewage sludge treatment. Many developing countries have realized the immense potential and benefits, with some of them installing gamma irradiators/ electron accelerators mainly for medical products sterilization and polymer modifications meeting specific needs. Efforts are also being continued by governmental / national bodies through setting up technological/pilot scale demonstration plants to encourage the private entrepreneurs to use /set up such facilities. However, two factors - the cost and non-availability of the associated equipment are hindering the effective utilization of the technology to make a big way in developing countries. This paper presents the details on the developments, prospects and the ongoing efforts to enhance the utilization of radiation technology for various applications in developing countries, with a specific mention about India.

1. INTRODUCTION

Radiation technology comprising both gamma irradiators and high energy accelerators are yielding tremendous industrial and societal benefits in the fields of polymer, healthcare, food and environment. Radiation induced polymerization and polymer modifications, namely surface curing, crosslinking & degradation brought out value addition to the products through environment-friendly, economically beneficial ways and has emerged as a multi million dollar industry [1]. Presently, processing of materials using high energy electron accelerators (200 keV to 10 MeV) constitutes the largest commercial radiation application. World over, there are over 1000 accelerators operating in the wire/cable, heat shrinkable, surface curing and other related industries. Radiation processed polymers possess superior mechanical, electrical and thermal stability compared to conventionally crosslinked ones. The process is simple and can be controlled by only one single parameter, i.e. absorbed dose, the quantity that varies with the application as mentioned in Table 1.

TABLE I.SOME OF RADIATION PROCESSING APPLICATIONS AND DOSE REQUIRED

Application	Dose required (kGy)
Disinfection	0.25-1.0
Food preservation	1.0-25.0
Medical Sterilization	20-30
Curing of coatings	20-50
Polymerisation	50-100
Crosslinking of polymers	100-300
Depolymerisation of PTFE	800-2000
Coloration of Diamonds	>>>2000

The success of radiation processing in industry is largely due to the advantages viz. a) desired chemical changes are induced at room temperatures, b) much purer product is obtained as chemical catalysts, initiators etc. are not required - hence environment-friendly, c) larger throughputs at high dose d) ability to hook up to the industry for existing on-line processing, e) energy efficient, f) switch on & off type radiation (machine sources) - hence more safe and increased public acceptance.

Radiation sterilization of medical products is another commercially realized industry, with no left over residues present unlike chemical treatments (ethylene oxide gas, heat treatments), presently carried out more with gamma irradiators mainly due to the ability of larger penetration thickness of gamma radiation than electrons. However, with the availability of new generation high energy electron accelerators having high throughputs, easy operation and increased utilization efficiency, EB sterilization is tend to grow further. Food irradiation to preserve food materials is another promising field using radiation which is currently being successfully applied by both gamma irradiators and accelerators. Rapid development of industry world wide resulted in polluting air as well as water due to the emissions of toxic gases and effluent disposals. Radiation technology is able to solve the problems in a more promising way. Efforts are on in the developing countries to beneficially utilize radiation technology in the fields specific to the country's needs simultaneously competent to the current conventional methods.

2. APPLICATIONS

2.1. Polymer Modifications

2.1.1. Radiation Crosslinking

The reactive species generated in the polymers like polyolefin when exposed to high energy electrons are responsible for the formation of cross links or net works resulting in increased molecular weight, improved physical characteristics like viscosity, tensile strength, elongation, high temperature operating/dimensional stability. Due to less or no additives, sensitizers, crosslinking agents need to be added in this process, the insulation thicknesses are generally less than the one developed using conventional technique for the same rating in case of cable insulations. The aging characteristics, solvent resistance are found to be far superior compared to the conventional chemical methods. Curing of thin insulations or products at the finished stage (on the extruded cable) is an advantage using electron accelerators. By employing suitable product handling mechanism, high speed curing can be carried out on line in the cable factories. However, such radiation crosslinkable insulation compounds are to be separately developed for specific use depending upon the requirement. Many such compounds have been developed, patented and are rather expensive to buy.

Electron accelerators are most favoured sources for these applications because of their ability to deliver high doses at large throughputs and thin products to be processed. Proper choice of electron energy has to be made for effective cable insulation irradiation by evaluating optimum thickness of the insulation - the thickness or depth where the dose equals the surface dose of the material when irradiated perpendicular to the beam of electrons. Thick cables require high energy. In case of thicker products, the irradiation should be carried out from both the sides. In case of cable irradiation, due to the presence of metal core, the opposite side of insulation does not get irradiated. Hence, the cables are generally turned to impart uniform dose, i.e. by rewinding the cable under beam several times.

In China, there are 45 industrial electron accelerators and 123 gamma radiation distributed in 28 provinces, municipalities or autonomous regions for various radiation processing applications. Malaysia has six EB accelerators for commercial use in this area with a 3.0 MeV EB / 90 kW and a 200keV/ 4kW at MINT for R&D, cross-linking and curing applications. A 300 KeV and a 3 MeV electron accelerator have been in operation for surface curing, wire& cable cross-linking in Indonesia. Korea has got nine electron accelerators installed in five cable factories and two tire companies have dedicated accelerators for curing of tire rubber sheet. Two more accelerators are producing polyolefin foams. A Rhodotron type accelerator has been operating in Iran for the past few years for radiation crosslinking and sterilization. In India, eight EB accelerators and six major gamma facilities are available for radiation processing applications as mentioned in the following Table 2.

S.N.O.	Application	Plants	Scale of Operation
1.	Radiation Sterilization	Co ⁶⁰ gamma irradiators –	Commercial
		3 nos.	
2.	Sprout Inhibition and other	"KRUSHAK" - Co ⁶⁰ gamma	Technology Demonstration
	low dose applications	plant	
3.	Disinfection of Spices	Spice Plant- Co ⁶⁰ gamma plant	Technology Demonstration &
			Commercial service
4.	Sewage sludge treatment	SHRI, Co ⁶⁰ gamma plant	Technology Demonstration
5.	Polymer modifications,	6 EB accelerators in operation	Commercial (3 nos.) and others
	coloration of diamonds	1 under construction	for Technology demonstration,
	and other applications		commercial service and R&D
6.	Food Irradiation and	10 MeV, 10kW EB accelerator	Technology Demonstration and
	Radiation Sterilization	under construction	commercial service

TABLE II. RADIATION PROCESSING APPLICATIONS IN INDIA

The following processing applications [2, 3] have been developed in India, using a 2MeV, 20 kW industrial EB accelerators in close collaboration with Indian industry and other R&D institutionssome of which have been exploited commercially.

<u>LDPE'O' rings</u>: The effect of crosslinking enhances the normal working temperature of simple polymers like LDPE, thus can be used as a cheap substitutes for the costly speciality polymers. Process optimization has been carried out to irradiate LDPE 'O' rings to withstand the temperature up to 200°C. These rings are used as gaskets for drum closures. Using a 'multi-spindle conveyor system' to impart uniform dose, the rings are being processed at a throughput of 100, 000 rings per day on commercial basis, meeting the industrial requirement.

<u>Wire & cables</u>: Electron beam crosslinked wire & cable insulations based on PE, EPDM, EVA and PVC materials have been indigenously developed in collaboration with Indian cable manufacturers. Product and accelerator parameters have been optimized and a cable irradiation conveyor has been used to irradiate larger length cables for process optimization. Around 100 km cables of varying thicknesses were processed and evaluated at the industry to suit the specifications required by Indian Railways [4]. Based on the successful results, the private industry had set up a 3 MeV, 150kW (Dynamitron type) accelerator in 2002 to produce EB crosslinked cables.

<u>Heat shrinkable products</u>: The 'memory effect' where an already stretched crosslinked polymer on heating shrinks back to its original dimensions led to the production of heat shrinkable products like films for food package wrapping, tapes, sheets, cable end caps for various applications. This is the second largest commercial industry in crosslinking. EB crosslinked heat shrinkable cable joints based on PE materials have been developed in collaboration with the Indian industry and process optimization studies have been completed. Longer length sheets (~ 100 m) were processed at the accelerator facility and were expanded at the factory line. The trials were successful and a full fledged set up to irradiate the sheets on industrial scale as a technology demonstration / service is planned.

<u>Radiation Degradation</u>: Radiation induced molecular weight reduction in PTFE scrap to obtain fine powder for use as dry lubricants is another promising application using electron accelerators. Micro fine powder obtained from the EB irradiated PTFE scrap having molecular weight of $10^4 - 10^5$ Dalton, which is other wise difficult to be pulverized using conventional techniques, is regularly supplied to the local industry in India. Reclamation of vulcanized butyl rubber scrap that can be reused in other rubber blends is commercially being exploited in China.

<u>EB Processing of viscose rayon</u>: Viscose industry with an annual production of 600,000 tons world over is a cause of major concern for polluting the atmosphere with the liberation of H₂S and other organic sulphides. India, one among the largest producers of viscose rayon, along with other developing countries, has taken up studies on the role of radiation technology, the effect of preirradiated cellulose in place of raw cellulose sheets in order to reduce the chemical consumption during the viscose process. From the laboratory scale experiments an optimum dose has been arrived at to bring down the DP to the desired levels leading to the requirement of lower quantities of CS_2 during dissolution process. Final fibre has been produced during the pilot scale studies, using high quality viscose with 25% reduction of CS_2 consumption. Studies are continued on viscose pulp drawn from various origins of the country. A regional training course and workshop has been conducted by India through IAEA for the RCA countries involving personnel from rayon industry.

<u>Radiation Processing of Natural polymers</u>: (Chitin/Chitosan): It is realized that radiation processing can also be utilized beneficially to improve the existing methodologies used for processing natural polymers or to impart value addition to such products by converting them to more useful form. Chitin is the most abundant natural polymer and is found in the exoskeleton of the marine life such as shrimps, crabs, shellfish etc. Since the biodegradation of chitin is very slow, accumulation of large quantities of discards from processing of crustaceans has become a major concern in the seafood processing industry worldwide. Work has been initiated to study the possibility of utilizing radiation technology for extracting chitin/chitosan under mild conditions and in higher yields. The results showed that while the yield of chitin remains unchanged after irradiation, the heating time for deproteinization at 110° C could be significantly reduced from 8 hr to 2hr. Therefore, pre-irradiation of shrimp waste can result in tremendous amount of energy and cost saving of the process. Besides this, irradiation of shrimp waste also increases the storage time of the waste by reducing the microbial rotting of the waste

<u>Super absorbent hydrogels</u>: The water soluble cellulose derivative carboxymethylcellulose (CMC) on exposure to radiation in the presence of monomers like acrylic acid (70% neutralized with NaOH) leads to the three dimensional crosslinked network structure that can absorb large quantities of water (about 460g/g of dry gel), thus acting as super absorbing hydrogels. The presence of ionic groups within the gel due to grafted acrylic acid are chiefly responsible for high affinity towards water. Such hydrogels have found many applications in the areas such as personal health care products and soil conditioner in rain deficient regions. Seeds have been successfully germinated in the swollen gels and studies on soil amended with gel did not show any adverse] change in soil microflora, indicating that such gels can be gainfully utilized in agriculture as soil conditioner. Studies to assess its use in agricultural application are planned in future.

Grafted membranes for selective adsorption of Metal ions: Radiation induced grafting is a potentially very important application of the ionizing radiation since it offers a unique method to modify the bulk or surface properties of a substrate, although the industrial success has been small so far. Many of the desired characteristics can easily be incorporated in a much inexpensive material to meet the demands of a particular application. Radiation grafting of anionic and cationic monomers to impart ion exchange characteristics is a very promising area and is being actively investigated. Amidoxime group containing fibrous absorbents for the recovery of uranium from sea water or useful metal ions from wastewater has been synthesized by radiation grafting of acrylonitrile followed by amidoximation of cyano groups with hydroxylamine. Experimental conditions have been optimized for achieving over 110% grafting of acrylonitrile on non-woven PP using post-irradiation grafting method using EB and subsequently converting acrylonitrile into amidoxime. The membranes have been tested under actual test conditions in sea water and have been found to selectively take up uranium from sea water. Further studies are in progress.

2.2. Radiation Sterilization

Radiation sterilization is an established application of radiation processing, a well-known and high volume industry. The process is easy to be controlled unlike in conventional techniques like temperature, pressure moisture, and gas compositions. Due to high process throughputs, radiation sterilization has become competitive to conventional steam treatments also. Presently a number of gamma irradiators are being employed for the sterilization. The number is more compared to the electron accelerator facilities. However, with the increasing availability of high energy accelerators in the range 5 to 10 MeV, coupled with the bremsstrahlung photon converters, radiation sterilization using electrons is also becoming competitive technique. By double side irradiation, packages of bulk density 0.15 g/cc can be irradiated uniformly up to a thickness of 35 cm using a typical 8 MeV accelerator.

There are about 21 irradiators (excluding China) that include India, Indonesia, Malaysia and Thailand. Radiation sterilization has been in use in India for medical products for three decades. ISOMED Plant, with ISO-9002 accreditation, at Trombay sterilizes about 13,000 m³ of medical products annually. Over two million DAI (midwifery) kits and delivery packs were radiation sterilized at ISOMED plant of BRIT, and distributed for use in rural areas for preventing infection of mothers and helping to minimize infant mortality rate, through rural health programmes funded by WHO. In a study, the International Institute of Population Studies (IIPS) has found that the infant mortality rate has fallen by 25-30 % in Rajasthan, Madhya Pradesh, Maharashtra and Uttar Pradesh as a result of distribution of the kits in these areas. Three more indigenously made irradiators are operating in the country. The expertise in design and development of commercial gamma irradiation plants are made available to other developing countries as well as local entrepreneurs through government body, BRIT (Board of Radiation and Isotope Technology), an enterprise of Department of Atomic Energy, India.

<u>Wound dressing hydrogels</u>: A PVA based hydrogel wound dressing possessing excellent healing properties using gamma irradiation has been developed. The product has been extensively tested in the local hospitals, patented and commercially launched recently in India by a local industry.

2.3. Food Irradiation

The process of irradiating the food has been established as safe for general application up to an overall average level of absorbed dose of 10 kGy. Many countries approved various products for irradiation. Co-60, Cs-137 radioisotope sources, x- rays generated by machine sources up to 5 MeV and electrons up to 10 MeV can be employed for the food irradiation purposes. The upper limit has been chosen so that no significant radioactivity is induced in the foodstuff. Many countries including developing countries like India, Indonesia, Bangladesh, and Thailand have set up food processing fields using radiation as shown in Table 3.

S.N.O	Country	No of irradiators	Food Commodities
1	Algeria	1	Potato
2	Argentina	1	Spices, spinach, cocoa powder
3	Bangladesh	1	Potato, onion, dried fish
4	Brazil	3	Spices, dehydrated vegetables, fruits, vegetables, grain
5	China	11	Spices and vegetable, Chinese sausage, garlic, apple, potato, onion, dehydrated vegetables, sauces, rice, tomatoes
6	Croatia	1	Spices, food ingredients, dried beef noodles
7	Czech. Rep	1	Spices, dry food ingredients
8	Cuba	1	Potato, onion, beans
9	India	2	Spices, onion, potato
10	Indonesia	2	Spices, rice
11	Iran	1	Spices
12	Korea Republic	1	Garlic powder, spices, condiments
13	South	4	Spices, shelf-stable food, fruits
	Africa		
14	Thailand	1	Spices, fermented pork sausages, enzymes
15	Ukraine	1	Grain
16	Vietnam	1	Onion

TABLE III.RADIATION PROCESSING FACILITIES FOR FOOD IRRADIATION IN SOMEDEVELOPING COUNTRIES

In India, a Spice Plant to process spices and other products using gamma radiation, with an initial throughput of 20 ton/day, has been operating since January 2000 at Navi Mumbai, A technology demonstration gamma irradiation plant KRUSHAK with 10 ton/h capacity for sprout inhibition of potatoes & onions and to treat other products requiring low doses, has also been commissioned in Lasalgaon, Maharashtra state. The indigenous technology has been offered to private entrepreneurs and site clearance has been obtained to set up two plants for food processing. The 2 MeV accelerator is being utilized to generate preliminary data to enhance the shelf-life of various meat products. A 10 MeV electron accelerator is coming up in Mumbai, India which is expected to be commissioned by the end of 2003, catering to the needs of health care and food industry.

2.4. Waste Management

The main areas of interest in waste management are radiation treatment of natural and polluted water, decontamination of industrial liquid wastes, treatment of sewage sludge, purification of flue gases, conversion of agro waste into useful animal feed etc.

The Coordinated Research Project (CRP) on 'Irradiation treatment of water, waste water and sludge'[5] was established to study the effects of ionizing radiation on refractory organic pollutants and pathogenic microorganisms and parasites in the treatment of water, waste water and sludge involving research institutes from eleven countries including Argentina, India, Indonesia Egypt, Ghana. The CRP demonstrated the capability of utilizing electron beam and gamma radiation treatment as a means of removing both the natural and synthetic contaminants from municipal and industrial liquid effluents. Sewage sludge being a good source of plant nutrients like organic carbon, nitrogen and various trace elements can be hygienized by radiation which can be used as safe organic manure and soil conditioner in agriculture.

Sufficient experience has gained and useful data has been generated using a Co-60 gamma irradiator (SHRI) in India to hygienize liquid sewage sludge, which is under operation for the last ten years and the usefulness of the technology has been proved in this field. Proposals for setting up such facilities in near future are under consideration. Studies are initiated using electron beam accelerators to treat at large throughputs for big cities soon.

<u>Upgradation of cellulose waste into Animal feed [6]</u>: The agricultural waste containing cellulose has high level of lignin which animals can not digest. Using radiation processing of such waste the lignin content can be reduced significantly so that the animal digestability can be improved. Inoculating such decontaminated agro wastes with useful organism like mushrooms and subsequently improving their nutrition values by fermentation further improves their quality of animal feed. The program of upgradation of cellulose waste into animal feed has been initiated by IAEA through RCA region. Malaysia conducted preliminary economic evaluation of processing oil palm empty fruit bunch to animal feed by radiation processing. Such radiation treated cellulose waste can be used for mushroom cultivation. The resultant waste after harvesting mushroom act as good nutritious animal feed rich in protein supplements.

2.5. Color enhancement of gem stones

Gems and jewellery constitute an important segment of the Indian exports. Colored diamonds and precious stones command a better saleability in the market. The use of radiation treatment of gem stones for colour enhancement has emerged as a novel way to produce colored gem stones. India is exploiting this application commercially.

3. CONCLUSIONS

The prospects of radiation technology, though very bright, will depend much upon the availability of the equipment and related technology at affordable cost. The availability of the accelerators and subsequent demonstration of technology will help in encouraging the entrepreneurs to adapt the technology in a commercially successful and environment friendly way. Realizing the

immense potential of the applications in different fields, a program has been chalked out in India to meet the country's needs for sustained utilization of radiation technology. The Department of Atomic Energy, India has set up a centre comprising of three indigenously made electron accelerators - 500 keV, 3 MeV and 10 MeV covering a range of applications like radiation curing, crosslinking, food irradiation, sterilization, colour enhancement of gem stones.

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A STATUS REPORT FROM AN ADVANCED ELECTRON BEAM SERVICE CENTER IN AUSTRIA

J. Mittendorfer^a, F. Gratzl^b

^aHigh Tech Consulting, Wels, Austria ^bMediscan GmbH, Kremsmünster, Austria

Abstract. In this paper the status of advanced electron beam service centers is described using the Austrian company Mediscan GmbH as example. Arguments pro and contra product treatment in a service centre against inline processing are discussed and the electron beam process together with design guidelines and process qualification considerations are presented. Product qualification and validation, which plays an important role in electron beam technology is found to be a key competence of a service centre. Criteria for operating a successful service centre conclude this paper.

1. INTRODUCTION

Electron Beam (EB) Technology may be described as product treatment using a beam of accelerated electrons or X-rays to gain a beneficial effect. Product treatment may be sterilization of medical devices, disinfestations of foodstuff, crosslinking of polymers, doping of semiconductors or degradation of polymers or pollutants in stack gas or water.

An electron source delivers radiation energy which induces the desired effect. The result of the irradiation may be a dose to the product, as it is the case for medical device sterilization or just an effect on the treated material (e.g. crosslinking or coloring of gemstones). The location where the electron or X-ray treatment is performed may be either in-house or a dedicated service centre.

The decision whether to perform the irradiation process in a service centre against building a system on site is guided by several key questions and arguments which are summarized in the Table 1.

PRO SERVICE CENTER	CONTRA SERVICE CENTER
No investment needed	Transport Cost
No operation/maintenance crew needed	Non disclosure of products and volume
Outsourcing of a critical process step	Logistics
Flexibility in volume	Time delay and lack of flexibility

TABLE I. ARGUMENTS FOR TREATMENT IN A SERVICE CENTER

The relatively high investment cost of an EB-facility is generally a hurdle when it comes to the decision between an inline-process and outsourcing to a service centre. High volume or high value of the product will favour an inline solution, but often a contract service is preferred in the beginning. The necessity for a qualified operation and maintenance crew, outsourcing of a critical, audited process step and the flexibility in volume are additional arguments for contracting to an EB-service centre.

Transport cost is a strong argument for an inline process and for a distance of several hundred kilometres the transport cost may exceed the sterilization cost. In an inline process no information about product and volume is given to the outside. Service centres, however maintain generally a very high standard of confidentiality. Flexible and efficient logistics can be an argument for an inline process, but the EB process is very fast and advanced service centres do "just-in-time" processing whenever possible.

For sterilization applications the task of a service centre is to deposit the required dose in the context of a quality system (ISO 9002, ISO 46002, and ISO 11137). The dose is determined by the bio burden and the required SAL (Sterility Assurance Level) following international standards [1]. For medical device a SAL of 10^{-6} is defining "sterility", whereas for the pasteurisation of food packaging a

SAL of 10^{-3} may be sufficient. To boost the service spectrum and acceptance, additional tasks may be performed by a service centre, e.g. distribution of products to the final destination.

For material treatment the task is to set up a reproducible process so the desired effects are established (e.g. gel content, mass flow index, and colouring of gemstones). Application where the desired dose has to be applied within small boundaries, setting up a statistical process control system with a feedback loop to the customer may be necessary [2].

2. MEDISCAN GMBH

A typical example of an advanced electron beam service is Mediscan GmbH, located in Kremsmünster, Austria. Mediscan, a member of Greiner was founded in 1995 and is operating an electron beam facility with two electron 10 MeV electron accelerators: a 15 kW Linac from Titan Scan and a 35 kW Rhodotron from IBA. Both systems are equipped with a sophisticated product handling system utilizing the latest developments in engineering and control. The product handling, process control, security system and all other subsystems have been developed by Mediscan engineers. Fig. 1 shows one of the two treatment vaults housing the Linac and part of the conveyor system. In addition Mediscan owns a Co-60 facility (1.5 MCi) in Seibersdorf, near Vienna and a 14 kW 500 keV ICT accelerator for research in water treatment and polymer modification.

The Mediscan service spectrum ranges from sterilization of medical devices, decontamination of food packaging and doping of semiconductors to process development and design of turn-key systems and modules for electron beam systems.



FIG. 1. The Linac treatment vault.

3. THE EB PROCESS

The absorbed dose of radiation energy, measured in Gray, is basically determined by three parameters:

- Average beam current
- Conveying speed through the radiation zone
- Beam geometry and scan width

S-Band linear accelerators deliver a pulsed beam, with a pulse length in the order of 10 to 20 μ s. The average beam is calculated as the time integral over the pulse current. Figs. 2 shows a typical example of a beam pulse and explains the concept of pulse and average beam current [3].



FIG. 2. Pulse and average beam current.



FIG. 3. Plot of absorbed dose vs. inverse of process conveyor speed.

The dependency of the dose from the conveyor speed is shown in Fig. 3 as dose-inverse speed distribution. The linear relationship between these two quantities is used for system validation purposes.

The beam energy determines the electron range and hence the treatable product size. Its choice is guide by several criteria:

- The products should be treated in their final shipping containers
- Penetration has to match the box size and produce an acceptable dose uniformity ratio (DUR) within the product.
- Energy should be kept minimal because of the shielding demand

For medical device sterilization electron energy of 10 MeV is typically chosen. Even then most products are treated from opposite sides to reach the necessary penetration and dose uniformity ratio. A useful quantity to roughly determine the necessary electron energy is the product of the density and penetration length, usually called surface weight or standardized depth $z=\rho.x$. If ρ is measured in g/cm³ and x in cm then z has the unity g/cm². Fig. 4 shows the Monte Carlo calculated depth dose curves in Polystyrene (PS) for different energies. The dependency of the atomic composition of the material has to be evaluated and will change the depth dose curve [3].



FIG. 4. Depth dose distribution for 5 to 12 MeV electrons in PS [4].



FIG. 5. Horizontal and vertical beam lines for product treatment

A critical decision when designing an electron beam treatment facility is the choice of the irradiation topology, namely horizontal or vertical irradiation.

Using a horizontal beam line has the benefit of a simple turning mechanism for double sided irradiation which is easy to validate. A scan magnet with an offset is important to match the product and conveyor layout. A vertical beam line is appropriate for large, flat boxes or when bulk material like granulate is treated. A turning mechanism for boxes is usually more complex and it has to be taken into account that some product must not be turned over. To guarantee a controlled irradiation layout, boxes are usually guided along a reference plane. In this case an asymmetric scan is needed as well. Fig. 5 explains the concept of horizontal and vertical irradiation topology.

An important step during process qualification is the validation for process interrupt. This feature enables a restart of the process with a box in front of the beam after a stop and can be handled with a ramp down- ramp up algorithm of the process conveyor servo-drive. An acceptable horizontal surface dose distribution during a process interrupt is shown in Fig. 6.



FIG. 6. Acceptable surface dose distribution for a process interrupt.

4. PRODUCT VALIDATION

Products have to be validated for electron beam treatment before undergoing routine irradiation [1]. This task is divided into two steps:

- Product and packaging material qualification Set up a program to demonstrate the quality, safety and performance of the product throughout its shelf life.
- Product Validation Validate a treatment scheme that the required dose in reached anywhere in the product and the maximum allowed dose is not exceeded.

Part of product validation is a detailed dose map, during which the minimum dose D_{min} , the maximum dose D_{max} and the reference dose D_{ref} outside of the box, together with their statistical variations are established. The correlation between the dose on the reference location and the minimum/maximum dose is used in routine processing for product release.

Absorbed doses are measured at Mediscan with calibrated radiachromic films. The calibration is done in-situ using Alanin transfer dosimeters. In addition PS-calorimeters are used for cross-check and backup. Mathematical models may help to ease and validate the dose mapping process [5].

Product validation requires a clear understanding of the EB-process, training and extensive hands-on experience. EB-service centres like Mediscan treat thousands of different products and hence possess a huge knowledgebase of products and procedures. Product validation is therefore a key competence of an advanced service centres which may further exploited by consulting for companies which think of an inline process.

5. CONCLUSIONS

The success of an electron beam service centre depends on many factors which are summarized as following:

- Reliable and well maintained equipment operated by a highly trained staff
- A well specified maintenance contract with the accelerator vendor (e.g. response time for interventions, spare parts availability)
- Optimized, yet versatile product handling system capable of running a wide range of products
- High quality and controlled process with optimized, lean procedures and paperwork
- Efficient warehouse, product flow and logistics capable of "just-in.-time" processing.

Electron beam treatment has proven its role in medical device sterilization and other applications and well managed, high quality service centres play an important role to demonstrate the usefulness of this technology. The success is even increased if besides product treatment additional services like process consulting and logistics support are provided and facilities act as problem solver for the industry.

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RADIATION PROCESSING FACILITIES: MALAYSIAN EXPERIENCE

Khairul Zaman Hj. Mohd Dahlan

Radiation Processing Technology Division, Malaysian Institute for Nuclear Technology Research (MINT), Kajang, Malaysia

Abstract. Radiation processing technology has been proven to enhance industrial efficiency and productivity, improve product quality and competitiveness. For many years, variety of radiation crosslinkable materials based on synthetic polymers have been produced either in the form of thermoplastic resins, polymer blends or composites. Today, effort is focused towards producing environmentally friendly and biodegradable materials using natural polymer. Radiation Processing Technology Division of MINT has carried out research program to utilize indigenous materials such as natural rubber and rubber based products, palm oil and palm oil based products and polysaccharide. Now the new challenges for the Division is to move from the laboratory to industrial scale and to seek opportunities in the competitive market place.

1. INTRODUCTION

As a government research institute, research activity of MINT is designed to meet the country development plans and aspiration of developing knowledge driven economy (k-economy). It is recognized that knowledge is the main driving force for economic growth of a country. In this connection, R & D program in Malaysia is designed to generate knowledge that can meet market demands and needs as to enable country to sustain the economic growth in a long term.

The government supports R & D and technologies that promote growth (increase export & reduce import); enhanced industrial efficiency, productivity and competitiveness; generate homegrown technology with own brands of goods and services; reduce labor with increasing automation and improve quality of life.

The Second Malaysian Industrial Master Plan (IMP 2) for 1996 - 2005, focuses on the manufacturing strategy and cluster-based development emphasis on the full integration of operations along the value chain R & D and product designs to marketing and distribution. Within the manufacturing industry, advanced materials such as composites, either polymer-based, metal-based or ceramic-based are given priority. It is much so, if resource based material can be integrated into the development of advanced materials. In this case, natural rubber and oil palm are the main sources. The by-products of the two resources such as rubber wood, rubber wood fibers, oil palm fronds and empty fruit bunches are the primary materials for further utilization - value added and meeting the zero waste concept.

In addition to composite materials, the modification of resource base materials that have commercial value is also strategically important. Epoxidised natural rubber and thermoplastic natural rubber elastomer are amongst the products that have been developed and commercialized in Malaysia. Modified palm oils such as polyol and epoxidised palm oil are developed and used as starting materials for polyurethane and polyester based resins for various applications.

2. RADIATION PROCESSING PROGRAM

In the light of the above strategy, the current industrial application of nuclear technology such as radiation processing fits in well into the country's development program. Radiation processing is one of the industrial processes that can be used for cross-linking, grafting, elimination of microorganisms, modification of organic compounds. The radiation processing technology can be an integral part of the manufacturing line for the production of flame/fire resistant wire and cable, heat shrink tube, hot water tube, heat shrink film for packaging, sleeve, composite materials, viscose rayon and many other profile and molded products. It has been proven as unique and commercially viable process. On the other hand, the materials used for radiation processing are specifically compounded and are not easily available. Therefore, the introduction of radiation processing technology in local industry requires different approaches in the following categories:

a. Established technology/material/product – acquisition of technology and adopt it in a local environment. This type of project still requires research for familiarization and sharpening of skill and technical know how. However, more developmental work is required to ensure the technology can be applied locally. This kind of project must have end user who is ready to utilize the technology without which the project will not take off.

b. New technology/material/product – require intensive research and followed by developmental work using pilot plant. This type of project will have longer R & D cycle and will face tougher task to bring the technology to the market either for OEM (Original Equipment Manufacturer) or REM (Replacement Equipment Manufacturer). It will require a good promotional and marketing strategy.

c. Radiation processing projects are formulated by taking into account the national priorities such as manufacturing strategy, resource based clusters, expected output of the project to meet the market needs. The following are some of the possible materials, processes and outputs of the research projects undertaken by MINT (Table I).

Materials	Process	Output
		Product/Application
Natural rubber:		
- SMR (Std.Malaysian Rubber)	- Blending/Composite	- Composite profiles such as panels,
- Epoxidized natural rubber	- Modification/ synthesis	frame, flooring for construction
- Latex	- Compounding	industry.
- Rubber wood fibers	- Extrusion	- Continues extrusion tube/hoses,
	- Injection molding	sheet/film, pipe, foam, membrane as
Oil palm:	- Coating/lamination	industrial products
- Crude and refined oil	- Casting	- Continues composite sheet, foam for
- Expoxidized palm oil	- Gamma vulcanization,	automotive parts such as window trim,
- Oil Palm fibers	- Electron beam crosslinking,	rear shaft and front panel.
	grafting and curing	- Injection molding of modified and
Polysaccharide:	- Radiation degradation	composite materials for automotive
- Starch from Sago & Tapioca	- Cleaning the gases and	parts
- Chitin and Chitosan	water.	- Hydrogel for bio-medical applications
- Carrageenan		such as wound dressing, slow release,
The sum of the start		micro encapsulation and coating
Inermoplastic:		- Hydroger for nearth care products
Polyothylopo (LLDPE)		such as temperature reducer –rever, ice
Lipper Density Polyethylone		Modified natural polymor for
(LDPE)		- Mounted natural polymer for
- Ethylene Vinyl Acetate (EVA)		promoter fruit coating water
- Polypropylene (PP)		absorbance for plant
- Polyvinyl chloride (PVC)		- Biodegradable foam and films for
- Polystyrene (PS)		nackaging and nacking
		packaging and packing.
Thermoset Composite:		- Clean air release to the atmosphere
- Polvester		- Fertilizers as by product
- Epoxy		- Clean drinking water and treatment of
		industrial wastewater before discharge
Environmental pollution:		to the environment or to be reutilized
- SOx and NOx from power		for industrial used.
stations and incinerator.		
- Industrial waste water and		
drinking water		

TABLE I.SOME POSSIBLE MATERIALS PROCESS AND OUTPUT EXPECTED FROMRESEARCH ACTIVITIES AT MINT.

3. CHALLENGES AND OPPORTUNITY

There are many established applications of radiation processing technology in industry such as follows:

- Radiation sterilization of medical products/pharmaceuticals/herbs/cosmetics,
- Radiation preservation of fruits/species/agricultural products,
- Radiation crosslinking of heat and fire resistant wire and cable insulation, heat shrinkable, tube, hot water tubes, 'O' rings, foam sheet and some molded plastic products,
- Radiation curing of coatings of temperate woods, metal, plastic, aluminum, etc.,
- Radiation curing of printing inks based on vegetable oil resin,
- Radiation curing of adhesive based on vegetable oil resin.

Most of these technologies are straight forwards, require research for developmental work which is essential to acquire know how in the process and operation of the plant. The examples of the technologies are gamma sterilization/preservation, electron beam crosslinking/sterilization and radiation curing technology. The technology can be adopted very fast as long as the facilities are available. More often to set up such facility would require intensive capital investments that need to be justified with the availability of market.

If one has to make a business decision, ability to sell the products with a reasonable profit margin is off prime importance. However, for a research organization, one must always have to look beyond a product. Continuous improvement, development of new products and innovation are the important elements, however will not guarantee a sustainability of economic growth of a country. The knowledge to use the technology, the knowledge to expand and diversify the use of technology and the knowledge to modify and upgrade the technology have more assurance for a long survival of the economic of the country. In other words, one has to invest on the technology in order to move forward. This is one of the challenges faced by many organizations.

MINT has two radiation sources, gamma and electron beam. Both plants are well equipped with the handling facilities that enable them to provide irradiation services to industry and more so to enable researcher to acquire knowledge and develop know how in radiation curable material and product development. With a request from company, a research on radiation curable heat shrinkable materials was conducted. Now, this material has been successful developed and transferred to a local company through research contract agreement for production of heat shrinkable tube. At present, electron beam irradiation center of MINT is providing irradiation service to this company.

MINT has set another mark of her technology advancement by embarking on the development of electron beam machine with the objectives to upgrade the knowledge and know how of local engineers, to have the ability to maintain, repair and fabricate Malaysian made machine. Local capability is this aspect is critical in order to push the application of electron beam technology into the industry.

In order to move ahead and to compete in the international market place, Malaysia needs her own brand, materials and products. This can be done either by ODM (own design manufacturer) and/or REM (replacement equipment manufacturer). To embark on this type of projects, one must have a good picture of market trends and needs. To protect the research finding and invention of MINT, the Intellectual Property Committee (IPC) has been established and Intellectual Property Right (IPR) Manual has been developed. IPC is given a task to coordinate and manage the request for patent, technology transfer and commercialization.

Malaysia is rich of natural polymer such as natural rubber and rubber based materials, palm oil and palm oil based materials, starch from sago and cassava, chitin and chitosan from shrimp shelves. These materials are currently being used in the country and the technology to support the production and utilization of the materials are well established. Being natural polymer, they are environmental friendly and biodegradable and they are well known as 'green polymer'. With the current low market price of these materials, it is more prudent to diversify its usage and to give more value added to the materials. This is the challenge that MINT is taking to add high value to the materials in order to find and develop applications for current and new market.

The typical R & D cycle (Figure 1) is as follows i.e.:

- Research work at the laboratory scale
- Developmental research at pilot scale
- Industrial scale.

Each product has to go through the above stages of development in order to determine its technical viability. At the laboratory research, the Radiation Processing Technology Division is well equipped with polymer processing facility such as melt blend mixers, compounder/extruder, cold and hot roll pressed, hot and cold press, table top injection molding machine, rheometer, etc. Subsequently, the materials will be subjected to physical, mechanical, analytical and thermal analysis in order to establish the material specifications. Figures 2 and 3 shows polymer processing and polymer testing laboratories respectively.



FIG. 1. R & *D Cycle.*

For the physical and mechanical testing, the Division has sufficient equipment such as several tensile machines, impact tester, and hardness tester; melt flow indexer, scratch and abrasion testers, adhesion tester, etc. For analytical and thermal analysis several equipment are available such as FTIR, SEM, STEM (under procurement), GPC, HPLC, DSC, DMA, TGA (under procurement). Where do we go after this level of research?



FIG. 2. Polymer processing laboratory.



FIG. 3. Polymer testing laboratory.

The government research institutes carry out most of the research activities in relation to new material development. Meanwhile, R & D by the industrial sector is more focus on product improvement and problem solving in nature. Therefore, the industrial sector depends very much on the government research institute to prove to them that their research finding is workable at the industrial level processing and the new materials can be transformed into the intended products. This is a lab-industry interphase stage. Usually at this stage, it involves the development of process and products at the minimum level of industrial processing, i.e. pilot scale. Currently, MINT has five pilot scale facilities as shown in Figures 4 to 8:

a. Gamma sterilization plant with the maximum Co-60 strength of 2.0 MCi for sterilization of medical disposable items, irradiation of food items, herbal and other products (SINAGAMA)

b. Electron beam processing facility for crosslinking of tubes and wire & cables (ALURTRON)

c. Continuous gamma irradiation of latex using gamma pilot plant (RAYMINTEX) – Gamma facility is designed specifically for irradiating natural rubber latex (liquid) to produce pre-vulcanized latex for dipped products such as surgical gloves, balloon, condom.

d. Pilot plant to process animal feeds from palm oil empty fruit bunches wastes using gamma irradiation (STERIFED).

e. Flue gas treatment using electron beam accelerator: a semi-pilot scale of the out put of 400 cubic meter/hr gas from diesel generator.


FIG. 4. SINAGAMA: Gamma Sterilization plant for sterilization of medical disposable items, food items and other products.



FIG. 5. ALURTRON: Electron beam accelerator, 3.0 MeV, 30mA, with handling facility for research and continuous irradiation of wire and tubes.



FIG. 6. RAYMINTEX – Radiation plant for prevulcanization of natural rubber latex.

This gamma facility is designed specifically for irradiating natural rubber latex (liquid) to produce pre-vulcanized latex for dipped products such as surgical gloves, balloon, condom.



FIG. 7. STERIFEED. Animal feed plant for processing of oil palm fibers waste into animal feeds.

A semi-pilot scale facility of electron beam treatment of flue gases of 400 cubic meter/hr generated from diesel generator is presented in fig. 8.



FIG. 8. Irradiation vessel for flue gas treatment using electron beam.

For the polymer materials processing, several machines have been installed such as a twinscrew compounder of 15 - 50 kg/hr out put (Figure 9), extrusion machine for profile and for continuous products such as for insulation of wire (Figure 10). Injection molding machine of 60 - 80ton clamping force is under procurement. These machines are crucial for inter-phasing the laboratory scale research with the industrial scale requirements. Several work for the processing of new compounds using the above machines have been established and ready to be transferred to industry such as:

- Flame retardant compounds for wire insulation
- Heat shrinkable compounds for tube commercialization stage
- PVN-ENR compounds for under-hood automotive parts
- Agro-fibers polymer compounds for automotive parts

All the above compounds require electron beam processing. In addition, electron beam processing of the following materials are under development and some are in the process of commercialization:

- Hydrogel from sago-starch commercialization stage
- Modified sago-starch commercialization stage
- Starch modified compounds for biodegradable foam products
- Starch modified compounds for biodegradable packaging film



FIG. 9. A twin-screw compounder for polymer blend/composites processing.



FIG. 10. Extrusion machine for profile and wire insulation.

On the other hand, the acrylate based oligomers as one of the important ingredients for radiation crosslinkable compounds synthesized from palm oil products have also been developed and a semipilot scale synthesis facility is under procurement. Several applications of acrylate-palm oil oligomers are:

- Pressure sensitive adhesive
- Printing ink
- Coatings

For research organization to develop the materials and products until stage 3 is costly and high risk. It is the role of private sector to commercialize the technology. The cooperation of private sector can be in various formed such as:

- making available the existing plant at their factory for the industrial pilot trial study
- providing fund to buy pilot plant facility
- making payment for the trial run conducted oversea at the machine manufacturer facility.
- requesting for a government support through the commercialization research fund scheme and from industrial grant scheme.

In most cases, the industry chooses the last option whereby they will request government funds for the commercialization of the research finding.

After the successful pilot scale trial, it is important for the project group to follow through the development of the project up to the initial commercial production. The role of inventor in every stage of R & D till commercialization is essential. Scientists today should equip themselves with the knowledge on market needs and trends in their own fields. They should be sensitive to the political, economy and social changes around them. The implementation of AFTA (ASEAN Free Trade Agreement) within these few years will have strong bearing on the economy of Malaysia and ASEAN countries. AFTA will result less trade barrier, high competition with less profit margin and high volume for bigger market size. The target market is regional and global rather than local. This can be a threat and challenge for Malaysia in the near future and the Malaysian success is depend on how to turn this threat into opportunity.

For future projects, radiation processing of nanocomposites based on indigenous and natural polymer will be given more attention. On the other hand, specialty materials/products in biomedical, electronic and ICT, automobile, aerospace and construction will be the major expected output of research in future.

4. CONCLUSION

In the past several years, there is a significant progress and development on the application of radiation processing in Malaysia. Government continues to support R & D in this field by providing the necessary infrastructure, facility, training and research funds. Various mechanisms for commercialization are also in placed to facilitate the transfer of technology from laboratory to industry.

In the private sector, several units of electron bream machines are in operation such as 3 units for heat shrink films and 2 units for crosslinking of wire. For gamma sterilization facility, four industrial plants are in operation including one at MINT. It is envisaged that radiation processing will continue to contribute to the progress and development of industry in Malaysia.

RADIATION PROCESSING IN EGYPT

R.M. Yousri

National Centre for Radiation Research and Technology, Cairo, Egypt

Abstract. The National Centre for Radiation Research and Technology (NCRRT) was established in 1972 aiming at promoting research and development using ionizing radiation. Radiation processing of medical and food products in addition to industrial applications is the main concern and consideration at NCCRT. The centre is divided into three divisions, including twelve Departments, in addition to central laboratories, industrial radiation processing and research units, different workshops, and technical and agricultural service sectors. The main objectives of NCRRT are to facilitate the concrete application of radiation technology in environmental studies as well as the processing of selected materials for use in various industrial applications. In this respect, the following activities and efforts are being considered.

1. INTRODUCTION

The National Center for Radiation Research and Technology (NCRRT) was established in 1972 aiming at promoting research and development using ionizing radiation. Radiation processing of medical and food products in addition to industrial applications is the main concern and consideration at NCCRT. The center is divided into three divisions, including twelve Departments, in addition to central laboratories, industrial radiation processing and research units, different workshops, and technical and agricultural service sectors.

The main objectives of NCRRT are to facilitate the concrete application of radiation technology in environmental studies as well as the processing of selected materials for use in various industrial applications. In this respect, the following activities and efforts are being considered:

- 1- Extension of the market capacity for radiation-processed products in order to generate a substantial income is necessary. Therefore, a new gamma irradiation facility will be installed at Alexandria City (near the seaport) for the export of sterilized medical products as well as irradiated fresh and dried foods. The first stage of the project is completed, while the second and the final stages will be completed within the coming two years.
- 2- Intensify the collaboration with the industrial companies, such as CID Pharmaceutical Co., to finalize the proper implementation of EB accelerator for the commercial production of wound-dressing hydrogels. A contract was issued and signed with the Ministry of Health for that purpose.

Discussion is in progress with VIVIRAD Co. (formerly, High Voltage Corporation) to upgrade the existing electron accelerator, specifically to increase the beam energy from 1.5 MeV to 3 MeV. Such an upgrade requires a funding of about 0.5 Million US \$. We are hoping that this can be supported through a bilateral agreement between Egypt and the IAEA. Through this modification, the following can be accomplished:

- Production of wound-dressing hydrogels of different thickness.
- Production of heat-shrinkable materials by radiation crosslinking technology.
- Crosslinking of larger wire and cable insulators.
- Recycling of agriculture waste products.

- Improvement of the physical and mechanical properties of wires and cables of different sizes. Several meeting have been held with a large commercial company (El-Seweedy Industries) in this regard. They have visited European facilities to investigate feasibility and now are quite keen on proceeding with NCRRT. However, higher electron beam energy would be necessary to proceed with such industrial applications.
- 3- Production facility for radiation-sensitive indicators is being expanded to meet the demand. The R&D for these indicators was carried out at the NCRRT. They are used for process control for radiation applications. We are aiming to fulfil the needs of the region.
- 4- The enhancement of the utilization of the peaceful applications of the nuclear energy to the welfare and sustainable development of Egypt.
- 5- The adoption of the principle of full transparency regarding all nuclear activities in Egypt.
- 6- The commitment to internationally accepted nuclear treaties promoting the peaceful applications of nuclear energy (for example the NPT) and compliance to the international safeguard system for unclear materials as long as these commitments don't endanger the national security of Egypt.
- 7- Securing the radiological environmental safety of Egypt, from both internal and external radiation hazards.
- 8- Enhancing the public awareness of the benefits of the peaceful uses of nuclear energy and addressing the public concern on nuclear issues.

2. TECHNICAL STATUS AND ACHIEVEMENTS AT NCRRT

2.1. Gamma Facility and Laboratory Sources

Mega Gamma I type J-6600 irradiator is a self-automated industrial Co-60 facility and is applied for radiation sterilization of medical products and devices and foodstuffs irradiation. It was constructed in 1979 with strength of 500 kCi. The current activity is close to 275 kCi. The recently upgraded transport and control system from analogue to digital using PLC system was done in year 2000 and it allows perform the automatic operation at certain irradiation dose level and screening all the information related to the facility. Safety conditions and ventilation equipment are provided according to IAEA instructions. Suitable loading and unloading zones are present to fulfil necessary storage surfaces in the facility. The gamma plant has a tote box conveyor system of 0.25 cum per tote box.

Five laboratory size gamma sources are used for basic and apply research. They are as follows:

- Cobalt Gamma Cell, model GC 220, MDS Nordion, Canada. Activity Co-60 sources at March 2001 : 91 Ci, dose rate 0.019 Gy/s,
- Caesium Gamma Cell, model GC 40, MDS Nordion, Canada. Activity Cs-137 source at March 2001: 91 Ci, dose rate 0.009 Gy/s.
- Cobalt Gamma Cell, model 4000A, BARC, India. Activity Co-60 sources at March 2001: 1897 Ci, dose rate 1.4 kGy/h.
- Cobalt Gamma Cell, model Issledovatel, Russian Atomic Energy Authority, Russia. Activity Co-60 source at March 2001: 10642 Ci, dose rate 7.1 kGy/s.
- Cobalt Gamma Cell, model 220, MDS Nordion, Canada. Activity Co-60 sources at March 2002: 10 kCi, dose rate 9 kGy/h.

The gamma laboratory sources installed at NCRRT are sufficient for wide research program implementation and high –dose reference dosimetry laboratory use.

2.2. Electron Beam Accelerator Facility

The electron accelerator was installed in the Centre under IAEA technical and financial support. Accelerator is installed vertically in concrete shelter placed in technological hall. The nominal technical specification of the accelerator is as follow:

- Electron energy 1.5 MeV,
- Beam current up to 25 mA,
- Beam power up to 37.5 kW,
- The length of the scanned beam up to 90 cm.
- The other instruments provided by IAEA under technical support program for accelerator facility are as follow:
- Conveyor system,
- Transport system for continuous irradiation of wires and tubes,
- Chart recorder,
- Set of test and maintenance tools and accelerator spare parts.

2.3. Radiation Safety Systems and Procedures

56 persons in NCRRT were considered as radiation workers due to their activity related to exploitation of sources of ionizing radiation (gamma industrial facility, 5 gamma cells, electron accelerator) in 2002. The basic radiation protection measures at the NCRRT are mainly related to International Basic Standard, IAEA, No 115, 1994 and International Commission of Radiological Protection, 1991. The practical application of these measures is dealt with three principles:

- no occupational radiation exposure should be adopted unless it produces sufficient benefit,
- personnel doses should be kept as low as reasonably achievable (ALARA) taking into account economic and social factors,
- the individual doses should be subjected to specified limit: whole body dose equivalent of 20 mSv/y for occupational exposure and 1 mSv/y for public, considering work load as 2000 h/y.

The following radiation monitoring instruments are used to control external radiation exposure around gamma sources installed at NCRRT:

- 1. Eberline X-gamma monitor up to 30 mSv/h,
- 2. ND-3000 X-gamma monitor up to 10 mSv/h,
- 3. Ludlum model 44-88 detector for alpha and gamma radiation.

The maximum gamma radiation levels around NCRRT irradiation facilities were carefully investigated at different positions (top, around shield, 1 m distance, and control panel). It should be noticed that dose rates at some positions around the irradiation facilities are higher than radiation protection limits. These positions are very occasionally occupancy areas during maintenance of this unit under the radiation protection supervision.

2.4. High-Dose Reference Dosimetry Laboratory

A reference laboratory for high-dose dosimetry has been established at NCRRT to provide traceability for the calibration of dosimetry systems required for radiation processing facilities with

electron accelerators and gamma sources. The main activities of High-dose Reference Dosimetry Laboratory are related to:

- traceability to industrial and research facilities,
- dose mapping procedures,
- training programs on dosimetry and quality assurance,
- inter-comparison dose programs,
- immediate assistance in case of problems arise at an industrial gamma and electron facility,
- provides seminars for personnel at their facilities on information related to dosimetry and process control.

Reference Dosimetry Systems are based on:

- Dichromate aqueous system for gamma rays,
- Polystyrene (PS) calorimeters for electron beam,
- Alanine-EPR measurements both for gamma and electron beam,
- Ethanol chloro benzene (ECB) dosimetry system.

The principal equipment located in High-Dose Reference Dosimetry Laboratory is as follow:

- Unicam UV/VIS spectrometer model 8625,
- Dedicated ESR spectrometer for alanine dosimeters measurements,
- Environmental control chamber model 518,
- Millipore system for water purification,
- Co-60 source Gamma Cell 220-2.

3. CENTRAL LABORATORIES

The NCRRT laboratories are equipped with high number unique analytical instruments. The instruments listed bellow are used in different scientific applications and investigations on selected processed materials:

- X-rays Diffraction (XRD) MODEL DP-DI, Shimadzu, Japan,
- X-rays Fluorescence Analyzer model EDX4, Philips,
- X-rays Forces model DX-95,
- Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AEA) model Ultima, Joban Yvon, France,
- Nuclear Magnetic Resonance (NMR) model NMR-300 MHz, Bruker, Germany,
- Electron Spin Resonance (ESR) model EMX, Bruker, Germany,
- Ultramicrotome model III, Co. LK13, for sample preparation for transmission electron microscope,
- Transmission Electron Microscope (TEM) model JEM-100 CX, JOEL, Japan,
- Scanning Electron Microscope (SEM) model JEOL-JSM 5400, JEOL Japan,
- High Performance Liquid Chromatography (HPLC) model WIS 201 + FL 2000 Detectors, Thermo Separation Products, USA,
- Amino Acid Analyzer (AAA) model Biochrom 20 Swede, Pharmacia Biotech,
- Atomic Absorption Spectrophotometer (AAS) model Unicam 939, Unicam, England,

- High Performance Microwave Digestion System model MLS 1200 ega.
- Differential Thermal Analysis (DTA) model DTA-50, Shimadzu, Japan,
- Differential Scanning Calorimeter (DSC) model DSC-50, Shimadzu, Japan,
- Thermal Gravitational Analysis (TGA) model TGA-50, Shimadzu, Japan,
- Thermal Mechanical Analysis (TMA) model TMA-50, Shimadzu, Japan,
- Gas Chromatography / Mass Spectrometer (GC-MS) model Finnigan SSQ 710, Finnigan Mat, USA,
- Gas Chromatography / Mass Spectrometer (GC-MS) model hp 890, Hewlett Packard, USA,
- Super Critical Fluid Extraction (SFE) model hp 7680 T, Hewlett Packard, USA.

4. MAJOR FIELDS OF INTEREST IN RADIATION TECHNOLOGY AT NCRRT:

4.1. Radiation Processing of Food

Radiation processing of food is one of the main activities of NCRRT and it was started in 1973. The Department of food irradiation has several laboratories namely food technology, biochemistry, microbiology, nutrition, wholesomeness and radiation dosimetry. There are more than 200 Ph.D. and M.Sc. thesis produced by NCRRT and the universities in relation to food irradiation and 6 theses in relation to the economic aspects of the technology.

In June 1996, the Egypt Health Authority has granted an unconditional clearance for food irradiation, up to the level of 10 kGy for herbs, spices and dried anion and dried garlic. This is a commitment of the Government and NCRRT in particular to the application of gamma radiation in reducing losses and contamination due to microbial pathogens, bacteria, yeast, molds as well as insects and parasites. Other food items such as fresh fruits are still under consideration of the Ministry of Health in collaboration with NCRRT and Ministry of Agriculture. Irradiation services for foods are provided by Mega Gamma irradiation facility at NCRRT, Cairo. It was estimated that ~ 50 tons of food items per annum are irradiated at Mega Gamma irradiation facility. Several food products are treated by Gamma rays such as medicinal herbs (various types), spices, specific dry food items (Onion, Garlic, Potatoes and Vegetables).

A second new license was issued for treatment of fresh onion / garlic / potatoes / sweet potatoes. Currently, NCRRT is constructed an industrial food irradiation plant together with a medical product sterilization plant at the port of Alexandria. The buildings of the plants are under construction.

The following are some of the projects on food irradiation that are currently being carried out at NCRRT:

- i. Inhibition of sprouting of potatoes, onions and garlic
- ii. Shelf-life extension of vegetables and fruits such as strawberries, tomatoes, eggplants, plum, apricots figs, olives, etc ...
- iii. Radiation preservation of meat and meat products, and chicken and chicken products for increasing shelf life.
- iv. Radiation preservation of fish, seafood and fish products.
- v. Wholesomeness of irradiated food.
- vi. Elimination of pathogenic microorganisms from animal feed.
- vii. Disinfection of store grains and their products.
- viii. Identification of irradiated foods in cooperation with institution in Berlin, Germany,
- ix. Packaging system for irradiated food.
- x. Conversion of agriculture waste into animal feeds.
- xi. Study of insects in relation to food irradiation.

Microbiology Department under the Biotechnology Division is also conducting some basic studies on microbiology in relation to food.

4.2. Radiation Sterilization of Healthcare Products

Research activities on radiation sterilization of pharmaceuticals, medical and healthcare products are carried out by several Departments such as Radiation Biology Department, Health Research Department, Microbiology Department, and Natural Product Department and supported other laboratories such as quality control laboratory for bio-burden, sterility test and Dosimetry.

Up to date there were more than the above Departments, which were supervised by more than 60 Professors and Associate Professors, produced 100 Ph.D. and M.Sc. thesis. The main facilities used for research work are gamma cells/chambers.

The commercial radiation sterilization activities started in 1980 as 500 boxes were sterilized using the Mega Gamma 1 type J-6600 irradiator with initial activity was 500 kCi. In 1997, the quantity of gamma sterilization products has increased to 19,000 boxes. The type and number of irradiated products also increased ranging from blood lines, droppers, kidney filters, petri bottles, aluminium foil, plaster dressing, dressing, valves, surgical gloves, cat gut-chromic, masks dressing, medical packages, catheters, medical preparations (antibiotics), syringes needles, intravenous sets, and pharmaceuticals products to medicinal herbs, spices and dry food items. Up to now, NCRRT has provided radiation sterilization services to more than 70 companies for more than 200 types of products.

Currently an industrial sterilization gamma plant for medical products is being constructed at the port of Alexandria, Egypt.

4.3. Electron Beam processing of Polymers, Biomaterials and Surface Curing of Materials

It is another field of research that has many applications. NCRRT is equipped with 1.5 MeV, 25 mA, electron accelerator that is in operation and in good condition. This electron accelerator is used mainly for radiation processing of polymers and biomaterials such as wound dressing hydrogels, stimuli-response membranes, and drug-delivery systems for control release as well as for surface curing of materials.

Polymer Chemistry Department and Radiation Chemistry Department are the two main users of the electron beam accelerators. Other Departments that have interest on electron beam accelerator are Radiation Engineering Department, Solids and Accelerator Departments and Radiation Physics Department. Several testing laboratories such as Central Laboratory (mechanical, thermal and analytical equipments), Dosimetry laboratory and Radiation Protection Laboratory support research activities on radiation processing of polymer, biomaterials and surface curing.

There are more than 30 Ph.D. and M.Sc. thesis generated from radiation processing research at NCRRT. At the Department of Polymer Chemistry and Radiation Chemistry, there are more than 50 scientific staffs. The strength of the radiation processing research at NCRRT is on the following:

- Radiation grafting of membrane for various applications in industry, medical and agriculture such as for removal of detergents/pesticides, removal of toxic elements and separation of radio-nuclides.

- Radiation processing of hydrogels for wound dressing, drug delivery, magnetic/electric sensitive, hydrogels containing functional group for recovery metal ions, hydrogels for agricultural use control release of pesticides.
- Recycling of waste polymer such as rubber and agricultural waste such as rice husk, cotton husk for making composites as shielding and container for transport.
- Polymer blend and composites such as rubber/polymer/glass fibres composite.
- Electron beam processing of polymeric industrial products such heat shrinkable products.
- Wood polymer composites and surface curing of coatings of wood products

Currently, some of the products generated from research activities are at the stage of commercialization. NCRRT is in negotiation with El-Sweedy Co. to irradiate heat shrinkable tubes at commercial scale. Meanwhile, NCRRT has entered into agreement with the pharmaceutical company for commercialization of the hydrogels for wound dressing. About 2,500 pieces of hydrogels has been distributed to several hospitals for clinical tests. In addition, 100 pieces of electron beam curing of laminated and surface coatings of wood has been distributed to several companies as promotional and demonstration of the electron beam curing technology.

In conclusion, electron beam processing of polymers, biomaterials and curing of surface coatings are very promising for industrial applications. NCRRT has started to produce products and to convince the industry to take up the research findings for commercialization. This achievement can be shared among the country in the region as well as neighbouring countries.

5. INTERNATIONAL COOPERATION WITH NCRRT

NCRRT has active international cooperation with the International Atomic Energy Agency (IAEA), Arab Atomic Energy Authority, IOM (Institute of Surface Modification), Leipzig, Germany; Julich-Germany; Takasaki Radiation Chemistry Research Establishment-JAERI, Japan; Polymer Institute, Budapest, Hungary; Hungarian Food Institutes; Canadian Atomic Energy Limited; Syrian Atomic Energy Commission, Saudi Arabian Universities etc. The cooperation consists of training/attachment of scientists and research projects.

Currently, NCRRT has two TC projects and three CRP with the IAEA in relation to radiation processing such as follows:

Technical Cooperation Projects (TC project):

- Upgrading the EB accelerator for industrial application (EGY/8/015).
- Establishment of High Dose Reference Laboratory (EGY/1/023).

Coordinated Research Projects (CRP):

- Radiation synthesis of stimuli responsive hydrogels and membranes for separation processes (11511/RO).
- The use of radiation processing sterilization or decontamination of pharmaceuticals and pharmaceutical raw materials (10353/RO).
- Use of irradiation to insure hygienic quality of fresh pre-cut fruits and vegetable and other minimally processed food of plant origin (302-D6-EGY-11680).

NCRRT has also received several IAEA trainees from Africa and Arab countries such as from Saudi Arabia, Syria, Jordan and Iran in the fields of polymer modification and improvements using radiation induced grafting and co-polymerization. In the past NCRRT has also actively involved in hosting many AFRA and AAEA training courses, meeting and workshop in relation to food irradiation, radiation processing of polymer and radiation sterilization.

With regards to commercialization, NCRRT annual income generated from the irradiation services in 2001-2002 is 0.7 million LE from medical products sterilization and 0.3 million from food irradiation. The irradiation services were provided to 57 medical companies and 20 food companies. This income is 60% higher than the previous year 2000-2001.

In 2003, negotiation with private companies is underway to commercialize hydrogels for wound dressing and to provide electron beam services for crosslinking of heat shrinkable tubes. Two new gamma irradiation plants will be constructed at the Alexandria City for medical product sterilization and food irradiation. The budget for installation of the cobalt sources for both plants has been approved and waiting for implementation.

ADVANCES IN STERILIZATION WITH X RAYS, USING A VERY HIGH POWER RHODOTRON AND A VERY LOW DUR PALLET IRRADIATOR

Y. Jongen, M. Abs, J.-L. Bol, B. Mullier, E. Poncelet, G. Rose, F. Stichelbaut

Ion Beam Applications S.A., Louvain-la-Neuve, Belgium

Abstract. A new concept of X-ray irradiator for high-density products on pallets is proposed. Monte Carlo simulations are applied to predict the performance of this system and compare it to alternative pallet irradiators. The Monte Carlo predictions are in good agreement with experimental data obtained using pallets of different densities. To take full advantage of this innovative pallet irradiator, IBA has also developed a very high-power electron accelerator able to produce high-intensity X-ray fields.

1. INTRODUCTION

IBA is actively involved in the development of X-ray irradiation systems for medical devices and food products. The superior penetrating quality of high-energy X-rays versus electron beams or 60Co gamma rays enables the treatment of products in large containers [1].

However, the double-sided method that is generally used can lead to an unacceptably large dose uniformity ratio (DUR), defined as the ratio of the maximal to the minimal dose absorbed in the product, for high-density products such as foodstuff on standard industrial pallets.

Recently, a new concept was proposed by MDS Nordion to improve the treatment of highdensity palletized products with X-rays [2]. This concept makes use of a turntable in front of the X-ray beam to irradiate the product from all sides and a collimator to shape the beam. IBA and MDS Nordion have decided to jointly develop this concept under the name Palletron and IBA is pursuing the engineering and development using Monte Carlo simulation tools.

Due to the relatively low conversion efficiency of electrons into X-rays, the use of X-rays on an industrial basis requires high-power high-energy electron accelerators. Based on its experience acquired the development of the Rhodotron [3, 4], IBA has launched a few years ago a vigorous R&D program to develop such high-power electron accelerator devoted to X-ray industrial applications. This research program resulted in the TT1000 Rhodotron aimed at delivering 5 MeV and 7 MeV electron beams with a current intensity of 100 mA.

2. THE PALLETRON SYSTEM

To obtain a rather uniform dose in a high-density pallet, it is crucial to be able to deposit a sufficient amount of dose at the centre of the product as compared to its surface. In the Palletron, the pallet is rotating around its vertical axis. The centre of the pallet is then constantly facing the X-ray target while a surface section only spends a limited amount of time in front of the target. To further amplify this effect, a collimator absorbs the X-rays emitted at large angle with respect to the initial beam direction, these radiations contributing only to the surface dose.

The main elements of a Palletron system are schematically represented in Fig. 1 (from [2]). An electron accelerator (20) is used to produce a high-energy electron beam (15) that is sent on an X-ray target (30) usually made of a high-Z material such as tungsten or tantalum.

High-energy X-rays (45) are produced by Bremsstrahlung and are emitted in all directions from the target. An adjustable collimator (110) is used to shape the initial X-ray beam into a collimated radiation beam (50) passing through a well-defined aperture (170). The product (60) rotates on a turntable (70) in the path of the collimated X-ray beam. The electron accelerator, the collimator, and the turntable are connected to the control system (120) so that the beam intensity, the size of the collimator aperture and the turntable rotation speed can be determined and adjusted either before or during the product irradiation.



FIG. 1. The Palletron: main elements (schematic view).

The design of the Palletron is created by IBA thanks to CERN's Monte Carlo (MC) simulation tool kit GEANT 3.21 [5]. This MC package allows a detailed simulation of the interactions between electrons/photons and materials disposed in complex geometries. GEANT is able to simulate the dominant processes that can occur in the energy range from 10keV to 10 TeV for electromagnetic interactions.

The basic elements of the Palletron have been introduced in the MC. The X-ray target consists of a 1.2 mm tantalum sheet in front of a 2 mm cooling channel filled with water and backed by 2 mm stainless steel. The collimator is represented by a stainless steel plate with a 14 cm thickness. Rotating the product by 2° steps approximates the pallet movement. The product dimensions correspond to standard US pallets: 100 cm (width) x 120 cm (length) x 180 cm (height).

The dose distribution obtained inside a rotating pallet depends on two main factors: the collimator's aperture and the rotation speed profile. These two parameters must be chosen as a function of the pallet bulk density in order to obtain a rather uniform dose. As an example, the speed profile optimized for a pallet of 0.8 g/cm3 density is shown in Fig. 2. The choice of this speed profile is motivated by two features of the system:

- Due to the rectangular shape of the pallet footprint, the product depth seen by the collimated X-ray beam is strongly dependent on the orientation of the pallet with respect to the beam direction. The rotation speed must be minimal when the product depth is maximal, i.e. when one of the pallet corners is facing the target.

- Points located at different positions on the surface of the product will have a different rotation velocity. The rotation speed must then be modulated to ensure an equal irradiation time for each point at the surface.

Another fundamental irradiation parameter is the collimator aperture. The influence of the collimator aperture on the dose distribution obtained in a pallet is described in Fig. 3. The left plot shows the evolution of the DUR and the minimal dose rate obtained in a 15 cm thick horizontal layer in the middle of the pallet. The right plot shows the 2D relative dose distributions obtained in this central layer for three different values of the aperture A. For a small aperture (case A), the maximal dose is located in the centre of the product as this centre is always irradiated by the beam while any portion of the surface is irradiated for a very short period of time. The DUR is large and the minimal dose rate is small. When increasing the collimator aperture, the dose uniformity improves while the minimal dose rate increases, until one reaches an optimal situation represented by case B, corresponding to an aperture of 16 cm. In this optimal situation, the DUR reaches a value of 1.3. If the aperture continues to increase, one observes a degradation of the dose uniformity related to an increase of the surface dose compared to the central dose (case C). At the same time, the minimal dose remains constant as it is given by the dose at the centre of the product.



FIG. 2. Rotation speed profile used for high-density pallets irradiated with the Palletron.

To obtain a uniform dose distribution along the vertical axis z, the electron beam is scanned along the X-ray target. Due to the large height of the pallets, the use of a homogeneous electron density on the target is not appropriate, a scanning width of about 260 cm being needed to reduce the dose variations along z below 10%. A better solution consists in using a non-homogeneous electron density as the one presented in Fig. 4(a). With a scanning width of 210 cm, it is then possible to limit the dose variations to about 5% along the pallet vertical axis as demonstrated in Fig. 4(b). Such an electron density can be obtained by applying a sinusoidal current to the sweeping magnet.



FIG. 3. Left: Evolution of the DUR and minimal dose rate as a function of collimator aperture in a pallet of 0.8 g/cm3 density. Right: Relative dose distributions obtained in the central plane of a pallet for different values of the aperture A.

Thanks to the good dose uniformity maintained along the z-axis, the global DUR obtained inside a whole = 0.8 g/cm^3 pallet is only slightly affected, becoming 1.4 for a pallet instead of 1.3.



FIG. 4. Choice of the scanning function along the X-ray target: (a) electron beam density along the scanning axis; (b) relative dose distribution along the pallet vertical axis for a product density of 0.5 g/cm3.

3. PALLETRON PERFORMANCE

The evolution of the DUR as is presented in Fig. 5(a) as a function of the product density for electron beam energies T0 of 5 MeV and 7.5 MeV. For densities below or equal to 0.4 g/cm3, no collimators are needed and a constant rotation speed is applied to the turntable. For densities larger than 0.4, collimators are introduced to limit the X-ray beam lateral expansion and a non-uniform rotation speed profile is required.

Figure 5(a) shows that very similar results are obtained at 5 and 7.5 MeV for densities above 0.4 g/cm3. For light products, the DUR has a tendency to increase due to the larger penetration of the X-rays leading to an over exposition of the pallet centre compared to its surface. This effect increases with the beam energy but can be controlled by the insertion of an absorption bar between the target and the product in order to subdue the intensity of the beam in the forward direction.

The treatment capacity obtained with the Palletron is shown in Fig. 5(b). These calculations are based on a beam intensity of 100 mA, a minimal dose requirement of 2 kGy and a pallet transfer time of 20 seconds. The Palletron being a single load system, this transfer time corresponds to the time needed to unload the treated pallet from the rotating table and load a new one.



FIG. 5. Palletron performance figures at 5 and 7.5 MeV: evolution of (a) the DUR and (b) the treatment capacity as a function of product density.

4. EXPERIMENTAL VERIFICATION

In order to validate the MC predictions, experimental tests have been conducted at the IBA facility located in Bridgeport, New Jersey. Pallets with footprint $100 \times 120 \text{ cm}^2$ corresponding to different bulk densities ranging from 0.18 g/cm3 up to 0.76 g/cm3 have been irradiated at 5 and 7 MeV with a beam intensity of 25 mA. Due to the scanning width of 200 cm and the homogeneous electron distribution along the target used at Bridgeport, the pallet height was limited to 150 cm. The dose distributions inside the pallets were measured by horizontal grids of CTA and Far West dosimeters located at various heights. The collimator consisted of 9 cm thick steel plates located on both sides of the X-ray target. The collimator was used together with a variable pallet rotation speed for the tests corresponding to densities of 0.57 g/cm3 and 0.76 g/cm3. The collimator was removed for the 0.18 g/cm3 test and a constant rotation speed was applied to the pallet.

Density (g/cm ³)	$T_0(MeV)$	D _{min} (Gy/min)	DUR _{Data}	DUR _{MC}
0.18	5	300±15	1.32 ± 0.09	1.33 ± 0.02
0.57	5	157±8	1.26 ± 0.09	1.27 ± 0.02
0.76	5	98±5	1.50 ± 0.11	1.45 ± 0.03
0.76	7	259±13	1.50 ± 0.11	1.47 ± 0.02

TABLE I. RESULTS FROM PALLETRON EXPERIMENTAL TESTS



Central plane - $\rho = 0.76 \text{ g/cm}^3 - T_0 = 5 \text{ MeV}$

FIG. 6. Relative dose distribution in the central plane of the high-density pallet irradiated at 5 MeV: (a) experimental data; (b) MC prediction.

The relative dose distribution measured in the middle of the high-density pallet irradiated at 5 MeV is presented in Fig. 6. The measured data is in good agreement with the MC prediction. Results from the Palletron tests are presented in Table 1. The minimal dose rate measured in the pallet middle plane is shown in column 3 while the DUR obtained in the data and the MC is compared in columns 4 and 5.

5. THE TT1000 RHODOTRON

Although the use of X-rays allows obtaining very good dose uniformity in pallets loaded with heavy products, this process suffers from the relatively low conversion efficiency from electrons to X-rays: this efficiency is about 8% for 5 MeV electrons and increases to 14% for 7.5 MeV electrons. In order to compete with the treatment capacities offered by the largest ⁶⁰Co sources, IBA has launched several years ago a vigorous R&D project to develop a high-power, high-energy electron accelerator primarily devoted to the generation of high-power, high-density X-ray fields. This project is now reaching its final phase with the qualification tests of the first prototype of this new machine called the Rhodotron TT1000.

As for the previous Rhodotron's developed by IBA (TT100, TT200 and TT300), the TT1000 is a recirculation accelerator where electrons gain energy by crossing several times a single accelerating cavity as shown in Fig. 7 [3,4]. This feature makes it possible to operate the machine in a continuous mode.



FIG. 7. Accelerator median section and electrons trajectory (D: deflection magnet, C: cavity, G: electron gun, L: focusing lenses).

The electrons are generated in a vacuum environment by the source (also called electron gun), located at the outer wall of the cavity. They are drawn away and accelerated by the radial field, which transmits them its energy. The electrons undergo a first acceleration toward the inner cavity wall. Then, they pass through openings in the centre conductor. Since the electric field is reversed when they emerge in the second part of the cavity, electrons are accelerated a second time, completing a crossing of the diameter. An external magnet then bends the accelerated beam and sends it back into the cavity for a second acceleration cycle.

The electron beam therefore travels along a rose-shaped path, which explains why the name Rhodotron was chosen (« *rhodos* TT1000, each», means rose in Greek). In the Rhodotron time the electrons cross the cavity, their energy increases by 1.2 MeV. Six passes and five magnets are therefore required to obtain a 7 MeV beam.

At the exit of the accelerator, the cylindrical shaped beam of high-energy electrons is transported or guided through beam lines from the accelerator to the radiation vault. There, the beam is expanded in a scan horn by scanning and then hits the X-ray target, which converts these electrons into X-rays.

The Rhodotron consists of the following major components: an electron gun, a RF power source, the accelerating cavity, the external deflection magnets, a cooling system, and the beam delivery system.

5.1. Accelerating Cavity

The Rhodotron accelerating cavity is a half-wavelength coaxial cavity (a tube surrounding a central conductor, both tubes having coincident axes), shorted at both ends and resonating in metric waves at 107.5 MHz (fig.8). It is made out of rolled, shaped and welded plates of steel. Once inside of the cavity, the flanges and the other reference surfaces have been machined; the cavity is electrochemically copper-platted.

Cavity outer wall has a diameter of 2 m. This has been designed in such a way that the time it takes for an electron to leave the centre, make a bend and come back to the centre is exactly one RF period. The cavity design has also been optimized in order to maximize the shunt impedance (ratio between the energy gained by the electrons and power dissipated into the cavity). The conical shape of the central conductor extremities has produced significant improvements.



FIG. 8. TT1000 Cavity: Rhodotron.

5.2. RF System

The RF system of the Rhodotron TT1000 has been designed to deliver 1000 kW. Around 110 kW are needed to build the electric field inside the cavity allowing an energy gain of 1.2 MeV by crossing. An additional 700 kW is then available to accelerate the electron beam at 7MeV.

The RF system consists of a main, voltage-controlled oscillator followed by a chain of amplifiers. The amplifiers up to 100 W are made of solid-state devices. The next three stages use power vacuum tubes.

The final power amplifier tube (fig.9), based on a Diacrode accelerating cavity, and is located partially inside the inner conductor. This configuration makes it possible to directly connect the anode cavity and the accelerating cavity with an inductive loop. Consequently, this eliminates the need for a pressurized wave-guide, contributing to the compactness of the machine.



FIG. 9. TT1000 Final Amplifier: Rhodotron.

5.3. Deflection Magnets

The primary role of the deflection magnets is to bend back the electrons emerging from the cavity and to redirect them toward the cavity centre. A second role is to contribute to the electron focusing.

In the Rhodotron, the electric field distribution provides part of the focusing at the cavity beam holes, the other part being provided by the magnets. All magnets have similar design. Centre to centre travel is equal to one wavelength. In the TT1000, the centre to centre travel for the first magnet is 2.

5.4. Electron Gun

The electron gun is located at the outer wall of the accelerating cavity (fig.10). The gun is grid-modulated synchronously with the RF frequency because electrons have to be injected into the cavity when the field is accelerating. The pulse width is about 60° of the RF period. The injection energy is 60keV, and the peak value of the injected current is 1A (mean value: 100mA).

The electron gun uses a planar cathode-grid assembly where the grid is grounded to the Whenelt. The cathode grid potential is automatically adapted to produce the desired beam current by electronic circuitry located in the power supply room. The design uses a dispenser type indirectly heated cathode and a gold-plated tungsten wire grid. The electron gun is class-C operated. The accelerating gap is optically designed to compensate space charge forces, and to form a converging beam of circular cross-section.



FIG. 10. TT1000 Electron Gun: Rhodotron.

5.5. Beam Transport System

Due to the high beam power to handle, a brand new beam line and scan horn, including the X-ray target, are under development allowing more efficient product sterilization or pasteurization. The proposed layout is presented at figure 11.



FIG. 11. TT1000 with its beam line and scan horn: Rhodotron.

5.6. Cooling System

The cavity is efficiently cooled by a water jacket on the inner conductor and on the end flanges and by discrete water channels along the outer diameter. Heat flow problems were carefully analyzed and optimized during the Rhodotron design phase.

Three cooling loops are provided:

- One normal water loop to cool down the cavity;

- One demineralised water loop to cool down the final power amplifier tube as well as other machine subsystems;

- One demineralised water to cool down the 700 kW power dissipated by the beam in the X-ray conversion target.

They are delivered complete with pumps, stainless steel heat exchangers, and water conditioning, and monitoring and temperature regulation units.

6. TT1000 PERFORMANCE

The TT1000 has been designed to produce 5 MeV and 7 MeV electron beams with a maximal intensity of 100 mA, corresponding to beam power of 500 kW and 700 kW, respectively. As shown in Fig. 12, the TT1000 exhibits for these high powers a remarkable electrical consumption efficiency that is above 50% both at 5 MeV and 7 MeV.



FIG. 12. TT1000 power efficiency at 5 MeV et 7 MeV.

After several years of R&D, the first TT1000 prototype has now entered into the final test phase. The most recent experimental tests have confirmed our expectations, the machine having delivered a continuous 80 mA beam for several hours at energy of 7 MeV. Development work continues to reach the design goal of 100 mA beams.

7. CONCLUSIONS

The Palletron is an innovative X-ray pallet irradiator allowing the treatment of high-density products loaded on industrial pallets with remarkable dose uniformity. Used in conjunction with the new high-power Rhodotron TT1000 developed at IBA, this system offers a very competitive alternative to the largest 60CO facilities without the drawbacks related to the use of large quantities of radioactive materials.

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ACCELERATOR TECHNOLOGY FOR RADIATION PROCESSING: RECENT DEVELOPMENT

Z. Zimek

Department of Radiation Chemistry and Technology, Institute of Nuclear Chemistry and Technology, Warsaw, Poland

Abstract. The progress in accelerator technology development means not only the growing number of unit but also lower cost, compact size suitable to the production line, beam shaped equally to the process, reliability and other parameters which are important in radiation processing application. The computers for automatic control and parts like power switches, thyristors, thyratrons and new generation of microwave sources are the best examples of the technology transfer which allows to perfect accelerators constructions. Demands coming from growing fields of radiation processing technology implementation stimulate R&D process of accelerator technology. Direct, transformer type accelerators, single resonant cavity accelerators and powered by microwave sources linear accelerators have been found the most suitable for radiation processing. Although the present level of accelerator development can satisfy most of commercial requirements the reliability is the most important parameter which should be improved to meet industrial standards. It should be noticed that the progress in accelerator technology is not a quick process but can be easily noticed in longer time scale. New accelerators constructions can frequently offer better economic and technical characteristics but only long time operation can revile weak points of certain accelerator in specific industrial conditions.

1. INTRODUCTION

The first charged particles accelerator has been constructed nearly 80 years ago. The fast grow of accelerator development was connected to rapid grow of nuclear experimental studies at that time. Cascade generator, electrostatic accelerator, linear accelerator and cyclotron were constructed in a short period of time at the beginning of thirties. The main differences between those accelerators were based on difference in electric field generation, related to this accelerating section construction and the accelerated particles trajectory shape. The primary accelerator application was strictly related to the field of nuclear physics. The fast development of accelerator technology created the opportunity to increase the field of application towards chemistry, medicine and industry. New ideas for accelerator construction and progress in technical development of electrical components were the most importance factors in process of accelerator technology perfection. Nearly 14000 accelerators have been applied in different brunches of scientific, medical and industrial activity up to now as it can be found in Table I [1].

TABLE I.	TOTAL WORLD ACCELERATOR POPULATION (AFTER W.SCHARF AND W.
WIESZCZYCKA	, 1999)

Accelerator category	No. in use
High energy accelerators (E>1GeV)	112
Radiotherapy	>4 500
Research accelerators	~1 000
Medical radioisotope production	~200
Radiation processing incl. R&D	$1\ 000 - 1\ 500$
Ion implanters	~6 000
Synchrotron radiation sources	~80
Total in 1998	~13 400

The progress in accelerator technology development means not only the growing number of unit but also lower cost, compact size suitable to the production line, beam shaped equally to the process, reliability and other parameters which are important in radiation processing application. Advances in high power switches technology, core amorphous ferromagnetic materials, modulator macro-pulses technology, CW operation of microwave generators are being transfer continuously to industrial accelerators development. The computers for automatic control and parts like power switches, thyristors, thyratrons, new generation of microwave sources are the best examples of the technology transfer, which allows perfecting accelerators construction.

2. PROGRESS IN ACCELERATOR DEVELOPMENT

Industrial accelerators development is still in progress not only because new kind of application but also because demands of lower cost, more compact size suitable for production line, a beam shaped equally to the process and other parameters which are important for radiation processing implementation [2, 3]. The certain stages of accelerator technology development can be recognized in past fifty years:

- adaptation of the accelerators primary built for scientific experiments,
- electron energy and beam power increase in certain accelerator constructions,
- dedicated accelerators for R&D, pilot plant and industrial facilities,
- computer control system for accelerator start up, full operation and technological process management,
- accelerator technology perfection (reliability, electrical efficiency, cost),
- accelerators for MW beam power level,
- compact and more efficient accelerator construction,
- very low energy accelerators.

Direct, transformer type accelerators, single resonant cavity accelerators and powered by microwave sources linear accelerators have been found the most suitable for radiation processing. Although the present level of accelerator development can satisfy most of commercial requirements the reliability is the most important parameter which should be improved. Further accelerator development may reduce the investment cost and increase the energy efficiency, what is especially important for environmental application where payoff from radiation technology has usually no economic background. Practical any accelerator construction must be compromise between size, efficiency and cost. The final decisions regarding accelerator construction should be taken in relation to the field of accelerator application to optimize accelerator performances. High power electron accelerators have been developed to meet specific demands high throughput processes like flue gas treatment and x-ray conversion for food processing. High power accelerators have substantially increased the capacity of the process with reduced unit cost of operation.

The new accelerators ideas and construction are being developed and tested continuously. Not always the final results are being acceptable for industrial application. The induction linacs in spite of their performances have never been commercialized as industrial accelerators. More frequently modern components are being adopted to improve existing technology. For example the microwave device magicon may provides power up to 5-10 MW in the CW mode and 500-1000 MW in pulse mode with efficiency 60-80% at the wavelength range 0.02-1 m. Relatively narrow frequency band (0,5%) and high amplitude and phase stability of such instruments may promise successful implementation in accelerator technology. Accelerators for transmutation of nuclear waste and other intensive radiation processes which require high beam power and high electron energy would be good field of application this technology in future. Published technical report describes design of high power CW linac energized by two 1,2 MW CW L-band klystrons to produce an electron beam with the energy of 10 MeV and current up to 100 mA [4]. The average beam power is supposed to be 0.2-1 MW for duty factor 20-100 %.

3. DIRECT TRANSFORMER ACCELERATORS

The construction of the accelerating structure depends on principle of accelerator operation and is related to the specific way of electric field formation. The electron gun is installed at one side of accelerating structure. The other side is connected to beam extraction device. Accelerating structures for direct and high frequency accelerators applied different principles of electrical isolation. DC voltage is used to accelerate electrons in direct acceleration method. The necessary DC voltage power supplies are usually based on use high power, oil or gas filled HV transformers with suitable rectifier circuit. They are simple and the most reliable accelerator component. HV cable is usually used to connect HV power supply and accelerating head for voltage level usually not higher than 0.8 MV. The MV voltage level in conventional transformer is impractical because of technical problem with insulation and dimensions of such device. Different type of inductance or capacitance coupling makes possible to increase relatively low primary voltage up to 5 MV by multistage cascade systems.

The power supply systems are used to provide energy for accelerating process. The most important parameters are related to voltage, loading current, time characteristics, size, weight and stability of electrical parameters. The high voltage DC power supplies with different principle of operation and construction were specially developed for direct accelerators where voltages up to 5 MV are being used. Parallel inductance or capacitance coupling systems are frequently used with suitable rectifying sections to increase the voltage level on the output of power supply. Interesting practical solution was proposed by Nissin HV, Japan. Several facilities were building based on Cockcroft-Walton cascade generator which allows obtaining accelerating voltage up to 5 MeV and average beam power 150 kW [5].

Power supply is the crucial part of any transformer accelerator. The specific constructions are made according to technology developed by certain accelerator producers. Several accelerator manufactures have produced nearly thousand accelerators based on such constructions. Among them ESI, USA; RPC Industries, USA; Polymer Physics, Germany; Nissin HV, Japan; NIIEFA, Russia have used power line transformer power supply technology. HVEC, USA and Vivirad, France have used ICT transformer system. Cockcroft-Walton cascade generators have been produced by Nissin HV, Japan. Unique Dynamitron system was developed by Radiation Dynamic, USA. Accelerator ratings and its efficiency are different for different power supply construction as it can be seen in Table II.

Type of supplyy	power	Power transformer	line	ICT transformer	Cockcroft-Walton	Dynamitron
Ratings		150-1000 kV		300-3000 kV	300-5000 kV 30-1000 mA	500-5000 kV
Frequency		50/60 Hz		50/60 Hz	1-3 kHz	50-100 kHz
Insulation Efficiency		Oil/ SF6		SF6	SF6 70-80 %	SF6 30-60 %
Remarks		Low energy,		Medium energy,	High energy,	High energy,
		Hign power,		Hign power,	Hign power, Large dimensions,	Low emciency.

TABLE II.CAPABILITY OF D.C. POWER SUPPLIES COMMONLY USED IN DIFFERENTTRANSFORMER ACCELERATORS

The power supplies with coreless transformers are used in accelerators developed in INP, Russia [6]. The maximum voltage 2.5 MV level was obtained (ELV-8) and loading current up to 800 mA has been used in Torch accelerator. The most recent achievement is the high power accelerator which accelerates electrons up to 1MV with total beam current 400 mA (ELV-12). The same company was engaged in construction of compact electron transformer accelerator operated at frequency 25-50 kHz. Semiconductors are used to build frequency converter from power line frequency up to 50 kHz what allows to build compact power supply with the specific power up to 130 kW per cubic meter of the volume occupied by this unit [7].

New and promising technology based on transformer was proposed by JINR from Russia [8]. The accelerator produces an electron beam pulses with duration 10 μ s. Pulse current can achieve 1 A with repetition rate 18 kHz. The electron pulses are produced by cold pyrolitic carbon mosaic cathode having a threshold volt-ampere characteristic. Accelerator reliability should be carefully tested before industrial implementation.

Interesting technical solution was discovered in France and described in literature twenty years ago [9]. A filament free system was proposed to be used in low energy transformer accelerator. Secondary emission system stimulated by helium ion plasma generator was applied. Unfortunately nearly 15 years were needed to commercialize that accelerator construction. Wire ion plasma accelerators were built by SHI, Japan. Compact accelerators WIPL type with electron energy 130-250 and beam power up to 50 kW were constructed for surface curing processes.

New quickly growing family of low energy accelerators has been developed recently. Very compact low energy pulsed accelerators were developed in Russia. Energy range 100-200 keV has been obtained with compact size and weight limited to 30 kg [10, 11]. Interesting constructions were proposed recently by US based companies AEBeam [12] and AIT [13]. Sealed-off accelerators can generate beams with energies 50-125 kV for wide range of technical applications. Table III contains basic ratings of recently developed direct accelerators.

Type of accelerator	Basic parameters	Manufacturer	Reference
Coreless transformer	1 MeV, 400 kW	INP, Russia	[6]
HF transformer (25-50 kHz)	0.5-1 MeV, 30 kW	INP, Russia	[7]
High repetition pulses	0.2-0.7 MeV; 10 µs, 18 kHz	JINR, Russia	[8]
Wire ion plasma	130-250 kW, up to 50 kW	SHI, Japan	9]
Compact, low energy, pulse	100-200 keV, 1 μs	Istok, Russia	[10]
Compact, low energy, pulse	180-250 keV, 4 ns, 30 kg	IE, Russia	[11]
Low energy	80-125 keV, up to 40 mA	AEBeams, USA	[12]
Low energy	50-70 kV, up to 50 W	AIT, USA	[13]

TABLE III.	RECENT ACHIEVEMENTS IN DIRECT ACCELERATORS CONSTRUCTION
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4. SINGLE RESONANT CAVITY ACCELERATORS

Single resonant cavity accelerators are based on the large resonant cavities working at the frequency, which may vary from one to several hundred MHz. The high power vacuum tubes are applied to provide necessary electromagnetic energy, which is used to accelerate electrons in accelerator this type. Those cheap and reliable components require relatively simply and compact DC or pulse modulators to generate high frequency oscillations. Medium and high electron energy level with appropriate beam power can be obtained. First industrial accelerators this type were developed in Russia more than 30 years ago. It was based on one coaxial resonator operating in pulse regime. The resonator was made of two separate halves mounted inside of stainless steel vacuum part. The central cylindrical part of resonator formed the accelerating gap. The electron injector consists of a grid, made in upper electrode to control beam current by changing the value of positive bias voltage on the cathode with respect to the grid. The self-excited generator made on industrial vacuum triode is used to form HV oscillation inside of coaxial cavity and provide necessary energy for electron acceleration process. Family of ILU type accelerators is offered in energy range 1-5 MeV and beam power 50 kW [14]. Several resonant cavities arrangement is proposed to increase to obtain electron energy 5 MeV and beam power up to 300 kW. In both cases the resonators are fully made of copper due to magnetic insulation, which exists along the accelerating structure and electromagnetic wave application.

New concept of single cavity electron accelerator arrangement was invented in France [16]. The coaxial line short-circuited on both ends was proposed to accelerate electrons in standing wave conditions. The electric field is radial with maximum at the median plane whereas the magnetic field is azimuthally and is equal to zero at the median position. That creates opportunity to accelerate electron beam crossing diametrically the cavity without distortion coming from magnetic field presence. Bending devices located outside of cavity are used to successive beam acceleration in the same electric field. The compact construction, high energy and high beam power make this accelerator suitable for industrial application. The Rhodotron concept was commercialized successfully by IBA, Belgium [17, 18]. Using multi pass system across resonant cavity 5-10 MeV electron energy, up to 100 mA beam current and up to 700 kW beam power have been obtained. The powerful compact accelerator constructions are being successfully used in many radiation facilities for high energy high

power radiation processing. The quick progress in Rhodotron accelerators development has been demonstrated by increase of beam power of accelerators offered for industrial applications (Fig. 1). Table IV describes single resonant cavity accelerators ratings.

TABLE IV.SINGLE RESONANT CAVITY	ACCELERATORS MANUFACTURERS
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Type of accelerator	Basic parameters	Manufacturer	Reference
ILU 6 to 10	0.5-5 MeV, 20-50 kW	INP, Russia	[14]
Electronshower	0.3-0.9 MeV, 9 kW	Denki Kogyo Co.	[15]
Rhodotron	5-10 MeV, 35-700 kW	IBA, Belgium	[17, 18]



FIG. 1. The single accelerator beam power as an indicator of 10 MeV Rhodotron accelerators development in last decade period.

5. LINEAR ACCELERATORS

The main feature of this type accelerator is the microwave energy used in electron accelerating process. Power supplies are made on the base of microwave generators with L- S- or X- band frequencies (1.3-9.3 GHz). A large number of small resonant cavities are used to obtain suitable electron energy. Microwaves sources parameters are playing the crucial role in linear accelerators. The klystrons are more stable in frequency and power but they have efficiency of 40-50% in comparison with 70% efficiency of magnetrons but with significantly limited life time. Linacs can be built with traveling or standing wave configuration. The last one technology can obtain higher accelerating gradient in cost of more sophisticated microwave power system and acceleration section technology. Continuous wave (CW) operation may significantly improve electrical efficiency (40%) and afford MW beam power level in near future. The recent progress was related to adaptation higher frequency technology (up to 9.3 GHz). Small and compact accelerators with relatively low electron energy were constructed in recent time (Table V)

TABLE V.LINERAR ELECTRON ACCELERATORS

Type of accelerator	Basic parameters	Manufacturer	Reference
High power	10 MeV, 50 kW	AECL, Canada	[19]
Low energy, compact	3 MeV, 3 kW, 1m	Thomson CSF, France	[20]
Selfshielded	10 MeV, 3-5 kW	Titan Co., USA	[21]

6. OUTPUT AND BEAM SCANNING DEVICES

The sophisticated magnetic systems can be built to shape electron beam according radiation process requirements. A number of different accelerator output devices have been described in literature [22-24]. Electron beam direction may be easily changed and suitable beam spot distribution at the output of electron beam device can be formed. The electron beam in point source accelerators can be easily scanned up to 2-3 m. Two dimensional scanning systems are used to improve efficiency of the window cooling. However, the scanned point source accelerators cannot be operated at the current much greater than 300 mA per one output window because of limited window thermal load due to the foil mechanical strength decay at the higher temperature. To overcome this with the reasonable length of output foil two or even three parallel beam paths (windows) can be applied in one output device. The recent progress in developing new composites for window foils may also increase the permissible beam current density level. New construction of output scanning system has been reported. The distributed electromagnet was used to scan the beam over long distance. The significant reduction of output scanner dimensions has been obtained in such scanner construction (Fig. 2).



FIG. 2. Accelerator output device equipped with distributed scanning magnet.

The different window water and air cooling systems are used to keep the window working temperature in permissible level. Air cooling system is used when electron beam energy is relatively high. Low energy accelerators with high beam power may require water cooling system. The copper supporting grid cooled by water is the common solution in such accelerator construction. Unfortunately up to 30 - 50% of total accelerator power is dissipated by window cooling equipped with supporting grid for low energy of electrons (300 keV). New construction of window system with water cooled grid support presented on Fig. 3 has been recently reported for higher electron energy level (0.75-1 MeV) [25]. Window this type with significantly improved electron transparency and reduced beam power loses may effectively reduce the risk of implosion caused by destruction by powerful electron beam and extend foil window life time.

Handling technique and the construction of the product transport system has a great influence on total facility efficiency and should be well matched to output window structure. Careful engineering of product handling assembles is equally important to successful industrial EB technology implementation as the reliability and design features of the accelerators themselves. This so call under beam equipment has to be designed specifically to the certain radiation process to minimize electron energy losses and increase process efficiency. The accurate control of product speed and positioning including the event of process interruption assure the proper quality of the EB process.



FIG. 3. The construction of window system with water cooled grid support for high electron energy level (0.75-1 MeV).

7. COMPUTER CONTROL SYSTEMS

Analogue control systems were commonly used in early accelerator constructions. Interlock system must fulfill safety requirements in addition to control and operation functions. Protection of the accelerator is provided against mechanical and electrical failure by electrical interlock in every accelerator component and installation. The feedback between the beam current level and the speed of conveyor is usually kept to provide constant dose to irradiated object. The computer or microprocessor driven control systems are only preferable solutions for modern accelerators now. The most favorable features of such system are related to:

- initial data are automatically checked to avoid incorrect data entry and eliminate operator errors,
- automatic start up and shut down procedures,
- automatic monitoring and control functions of every critical parameter,
- simpler and better process control,
- automatic conditioning,
- data logging and graphic display,
- higher reliability and simpler service procedures,
- automatic control allow to reduce the skill level required of machine operators,
- control system based on validated software,
- integrity of the process controlled on real-time base (error detection),
- graphic base operator interface (step-by-step instruction),
- access to the system (password and security).

Control system (Digital Process Controller) computer and I/O coprocessor has been developed to control accelerator operation [26]. The digital systems can be easily adopted for different accelerator construction and parameters. The system not only controls the current electron beam parameters but also provides necessary interlock safety system control and usually can be applied to

control the technological equipment during irradiation process [27]. Parameters measurement (energy, beam current, pulse repetition, scan width, and others), calculation and recording are typically included. Programmable Logic Control processor is usually used to control accelerator equipment. PC computers system is used to provide necessary communication with accelerator operator. LCD touch sensing panel to realize "One Button Control" sometime is used to simplify the accelerator operation [28].

8. X-RAY SOURCES

Electron to x-rays conversion effect was discovered more than 100 years ago. Since discovery x-rays due to theirs unique properties were widely applied in medical and industrial diagnostic instruments. Secondary electrons and x-rays shower are formed during penetration of any material by high energy electrons. The initial electron energy is degradated in scattering, ionization and excitation processes in the matter according to x-ray formation principles. Finally electrons are termalized and initial energy is converted into the heat. The efficiency of electron to x-rays conversion is relatively low and depends on material density and energy of the electron beam. High penetration abilities of x-rays may provide unique opportunity to irradiate a big volume of irradiated object.

The efficiency of the conversion and spatial distribution of x-ray stream are the main parameters of any target for application in radiation processing. The optimization of target construction should be performed to improve is technical and economical features. Under optimal conditions only 7, 6 % of total electron beam power is converted into forward x-ray stream of radiation at the initial electron energy 5 MeV. Up to 76 % of electron beam power has to be removed by cooling system. X-ray applications for radiation processing purpose is found in some circumstances to be economically competitive and offer more flexibility than gamma sources (easy control of radiation safety and intensity of radiation) especially for the irradiation of high density product like food and medical devices sterilization. Recent progress in high power and high energy accelerators development gives an opportunity to construct and apply reliable high power electron beams to convert on x-ray streams suitable for industrial application [29-34].



FIG. 4. The ratings and installation time of high power x-ray converters.

For food irradiation, the maximum photon energy according to FAO and WHO recommendation is 5 MeV now, what means that initial electron beam energy cannot exceed 5 MeV. The international discussion has been initiated to increase electron energy up to 7.5 MeV level [35-37].

To optimize irradiation conditions and calculate x-ray throughput several parameters should be taken into account: density and size of the package, radiation utilization efficiency, dose required, dose uniformity. Certain limits are present due to electron irradiation process which is the basic function of the facility. Two sided two times irradiation (four passes) may be applied to improve dose uniformity and increase x-ray stream utilization. More sophisticated Palletron system was proposed to overcome deep dose distribution [38]. Collimators inserted between the x-ray source and the products are used to shape the x-ray beam. A non-constant scanning of electron beam is applied to obtain a uniform dose distribution along vertical axis. Pallet load is rotating in front of the x-ray beam with a dedicated rotation speed profile. The Palletron system allows obtain max/min dose relation better than 1.5 for all densities between 0.1 and 0.8 g/cm³.

9. FINAL REMARKS

Characteristics steps can be recognized in the past of accelerator development. Present stage of accelerator technology perfection includes: cost effectiveness, reliability, compactness, introduction of MW beam power level and very low energy accelerators. Demands coming from growing fields of radiation processing technology implementation have a strong impact on R&D process of accelerator technology. The R&D in accelerator technology is tightly connected to progress in development of advanced technology in many branches of technical activity (power components, control systems). It should be noticed that the progress in accelerator technology is not a quick process but can be easily noticed in longer time scale. New accelerators constructions can frequently offer better economic and technical characteristics but only long time operation can revile weak points of certain accelerator in specific industrial conditions.

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ADVANCES IN SELF-SHIELDED ACCELERATORS

A.J. Berejka

Huntington, New York, United States of America

Abstract. The use of lead in lieu of concrete for shielding has enabled a significant segment of the electron beam (EB) processing industry to continue to grow. Self-shielded accelerators of 300 kV or less are used in the curing of environmentally-friendly thin-film coatings and in crosslinking extruded polymeric films. New low-voltage accelerator systems have been developed, including very economic modular units, which are expanding market interests. Transportable systems based on placing self-shielded accelerators on vans have found only minor interest for use in environmental remediation, with no commercial success to date. Higher-voltage, around 600 to 800 kV, self-shielded systems have found minimal acceptance in historic markets as in the crosslinking wire insulation and processing of tire components. However, new developments in higher energy, 2.5 MeV, modest current, self-contained systems may find use for in-house sterilization and treatment of products.

1. INTRODUCTION

Reviews of the industrial accelerator market over the years have shown little change in the basic structure of end-use segments [1]. Of the 1200 or more electron beam accelerators in industrial use, low-voltage, self-shielded systems used for surface curing have shown persistent market growth, now being around 30% of the entire market (Figure 1).



FIG. 1. Industrial Electron Beam Accelerator End-Use Markets.

The market use of an industrial accelerator dictates the accelerator voltage of choice and its corresponding electron beam penetration. In the low-voltage, self-shielded area, new accelerators at voltages as low as 80 kV are finding use. These have sufficient penetration to cure the 3 to 8 micron deposition of a printing ink, for example. At the other end of the scale, high-energy 10 MeV systems, that have proven use in the sterilization of medical devices, are finding acceptance in irradiating foodstuffs, where greater beam penetration is demanded (Table I).

Market Segment	Typical Energy	Penetration
Surface Curing	80–300 keV	0.4 mm
Shrink Film	300 – 800 keV	2 mm
Wire & Cable	1.5 MeV	5 mm
Sterilization	10 MeV	38 mm

TABLE I.EB PENETRATION – MARKET END-USES

2. SHIELDING REQUIREMENTS

The majority of industrial electron beam installations rely upon poured concrete (density 2.35) or precast concrete block to provide shielding from the Xrays emitted when uni-directional electron beams impinge upon a material. Guidelines limiting personnel exposure have been established which have lead to the construction of concrete beam vaults of nearly 3.5 meters in wall thickness for 10 MeV accelerators. Experienced accelerator manufacturers and designers of EB processing facilities provide more than adequate worker protection, including adroitly designed labyrinths, safety interlocks and the like. The necessary and required concrete thickness is related both to the electron energy and to the total beam power (kW) of an accelerator (Figure 2).



FIG. 2. Concrete Shielding Thickness (sideward direction, 200 kW beam).

While steel (density 7.8) has been considered for self-shielded EB systems, high-current industrial units rely on the more dense and malleable lead (density 11.3). Whereas with concrete shielding, it is presumed that the worker or beam operator will be at least three meters away from the radiation source, with lead-shielded systems it is presumed that the worker will operate the unit adjacent to it. Manufacturers of lead-shielded units provide more than sufficient thicknesses of lead to assure worker protection. Here too, the thickness of lead required is related to accelerator voltage and power (Figure 3).



FIG. 3. Lead Shielding Thickness (sideward direction, 100 kW beam).

With higher energy or mid-voltage accelerators, the mass of the lead shielding itself can become a concern. On the other hand, a trend for low-voltage accelerator technology to go down in voltage, to as low as 80 kV, also reduce the overall weight and thickness of shielding required (Figure 4).



FIG. 4. Lead Shielding Thickness (sideward direction, 10 kW beam).

3. LOW-VOLTAGE EB DEVELOPMENTS

Historically and more so in recent years, the number of credible suppliers of industrial low-voltage, self-shielded accelerators has been very limited: Energy Sciences, Incorporated (ESI); Nissin-High Voltage (NHV); and RPC Technologies (RPC). These companies all based their accelerator design upon the use of elongated filaments that can extend the electron beam across the width of a moving web or substrate without having to be scanned. Such electron sources are housed in evacuated, lead-lined chambers and powered by conventional coil wound transformers. Such EB equipment, as with the higher-voltage accelerators, is often custom designed for a given end-user and the user's required application. This "one-off" approach has tended to keep the costs, even for these low-voltage units, relatively high.





b. 50 cm Rack Mounted Power Supply



a. AEB Electron Emitter(25 cm diameter x 40 cm height)

c. AEB EB Emission Pattern

FIG. 5. AEB Electron Beam.
Property	AEB Emitter	Historic Low-Voltage EB System
Mode of manufacture:	Modular	Custom designed
	Repetitive	Single unit production
Electron source:	Heated filament	Heated filament
Beam chamber:	Factory evacuated	Evacuated by user
Beam window:	< 10 micron Ti foil	15 micron Ti foil
Beam voltage:	Variable 80-150 kV	Fixed increments
Power Supply:	Solid-state	Large coil wound transformer
Replacement/repair time	e: Minutes	Hours to days

TABLE II. LOW-VOLTAGE EB PROPERTIES

In 2000, a new entrant to the low-voltage accelerator area, Advanced Electron Beams, Incorporated (AEB), has changed the paradigm of EB manufacture and produced much lower cost equipment [3]. AEB produces a modular, factory evacuated electron beam capable of delivering sufficient current for most coating and surface applications. Modular units (Figure 5.a) are produced to standardized designs in volume production. Relying upon a "plug and use" approach, the AEB system removes accelerator maintenance from being a concern of the user. Modules that have exceeded their production life are returned to the factory for refurbishing. A back-up unit can be plugged into use within minutes, thus avoiding costly downtime in production processes. The AEB system also uses solid-state power supplies (Figure 5.b) which enable these low-voltage systems to have excellent energy efficiency, converting around 70% of incoming line power to useful beam. The AEB modular EB comes with a computer control system operated from a touch-screen panel. The electron beam from the standard 25 cm module diffuses so that modules can be placed adjacent to each other to extend the beam width. There is also beam diffusion in the direction of product travel (Figure 5.c). A summary of the differences between the AEB approach and that, which has been historically used in low-voltage EB systems is presented in Table II [4].

Advanced Electron Beams has also produced a compact laboratory unit based on one 25 cm emitter that comes complete with solid-state power supply, computer controls and a touch-screen control panel. This unit has gained considerable acceptance for use in product development and quality assurance programs (Figure 6).



FIG. 6. AEB Laboratory Unit.

ESI, the pioneer in low-voltage, self-shielded electron beam technology, has also developed new, lower but fixed-voltage accelerators. The ESI EZ-CureTM systems have lower accelerator voltages, down to 80 kV. These models are down-sized versions of the traditional ESI units and are also being marketed at more economic prices.

4. TRANSPORTABLE EB SYSTEMS

To deal with site-specific environmental problems, transportable electron beam systems have been developed which consist of placing a self-shielded unit into a large sized trailer, such as those typically used to move goods and products (Figure 7).



FIG. 7. HVEA Transportable System.

Three such transportable electron beam systems have been considered:

In the mid-1980s, Kernforschungszentrum Karlsruhe sought to develop a system using the ESI ElectrocurtainTM self-shielded beam to treat flue gases on site [6]. This system failed when it was found that particulates and corrosion could destroy the 17 μ m beam window and cause beam implosion. (Full-scale stack gas treatment facilities now rely on a double window approach to avoid window puncture and beam implosion.)

In the mid-1990s, the Raychem Corporation, one of the largest end-users of electron beam processing and now part of Tyco Electronics, developed a transportable system to treat and remediate contaminated ground water. A unique feature of this system was the direct spray of contaminated water onto a curved beam window (Figure 8) [7]. Because of lack of commercial interest, this project was abandoned.



FIG. 8. Use of Curved Beam Window.

Also in the 1990s, High Voltage Environmental Applications (HVEA) constructed a selfshielded accelerator in a large van using an under-beam water trough. While not as yet having found a viable commercial use for this equipment, HVEA has conducted over 60 demonstrations and test programs using its transportable system [8]. Table III summarizes the capabilities and status of these transportable systems.

System	Voltage	Power	Status	
Karlsruhe	200 kV	30 kW	Window failure	
Raychem	300 kV	7.5 kW	No commercial interests	
HVEA	500 kV	20 kW	~60 site demonstrations	

TABLE III	TRANSPORTABLE EB	SYSTEMS
		DIDIDIDID

5. MID-VOLTAGE, HIGH-CURRENT SELF-SHIELDED SYSTEMS

Nissin-High Voltage (NHV) offers mid-voltage (800 kV) high current (35 mA) self-shielded systems for use with wire and cable and with sheeting as a standard item [9]. The product being presented to the beam and its handling system are enclosed in a lead-lined vault. However, only a few of these systems have gained market acceptance. Vivirad France is also developing a 550 kV, 35 mA self-shielded unit for use with wire and tubing. The Radiation Dynamics (RDI) subsidiary of Ion Beam Applications (IBA) is developing a 1.0 MeV, 100 mA unit also tailored for more compact use in the wire and cable, tubing and sheeting markets. With these units, the thickness of the shielding required must be considered. At least ~15 cm of lead will be needed for a 1.0 MeV unit (Figure 3). The mass of this unit must consider the footings on which they will stand and the transport to a given site.

6. HIGH-VOLTAGE, SELF-SHIELDED STERILIZATION SYSTEM

IBA has developed a relatively compact 2.5 MeV, 6 kW dual sided, self-shielded electron beam system, the BetalineTM, for use when considering in-house sterilization of medical products (Figure 9. a) [11]. Relying upon a proprietary means of beam splitting, this unit is capable of delivering 2.5 MeV electron beams on both sides of a product transversing between the bent beams (Figure 9.b). As such, it will be capable of sterilizing or treating products with fairly thick cross-sections. At present, the system is designed to move trays of materials through the dual-beam area (Figure 10).



a. Betaline System for in-house sterilization

b. Betaline Beam

FIG. 9. Betaline system.



FIG. 10. Betaline Process Flow.

7. SUMMARY

Self-shielded electron beams have developed along two lines: a) there are now more affordable, lower-voltage units which overcome the price barriers to larger market use of low-voltage technology for the surface curing of coatings and inks; b) higher-energy (1.0 MeV) units have been designed and even fabricated (2.5 MeV). However, industry experience with the use of even mid-voltage (~800 kV) self-shielded systems has shown the difficulty of developing markets for this type of equipment. While transportable beams have been examined and evaluated for environmental applications, they too have not yet found market acceptance.

The fast growing market for surface curing, wherein liquids based on near-zero volatile organic compounds (VOCs) are converted by ionizing radiation to dried, functional inks and coatings, is finding low-voltage electron beam curing to be a point-of-source means of pollution prevention. These self-shielded, low-voltage units will have the most significant impact on environmental quality.

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DEVELOPMENT OF A FAMILY OF LOW, MEDIUM AND HIGH ENERGY ELECTRON BEAM ACCELERATORS

K.S.S. Sarma

Radiation Technology Development Section, Bhabha Atomic Research Centre, Trombay, Mumbai, India

Abstract. Commercial utilization of radiation technology started with sterilization of medical products using isotope gamma sources five decades ago. The pioneering efforts in R&D and applied aspects of radiation chemistry of polymers and the requirements for higher process throughputs matching the industrial production lines, necessitated the use of machine sources like electron accelerators. A range of high energy, high power accelerators suitable to process various materials have been developed for high speed polymerization, crosslinking and degradation applications. Presently, radiation processing of polymers using electrons has become an established technology being exploited commercially using reliable radiation equipment as well a suitable product conveying techniques for wire& cable insulations, heat shrinkable materials, surface curing etc. Advanced, latest generation high energy accelerators coupled with photon conversion targets are being installed and studied for their use in radiation sterilization, food irradiation and environmental applications. This paper gives a brief account of accelerators meant for radiation processing applications.

1. INTRODUCTION

Radiation processed products are currently being used in health care, food preservation, polymer based industries. The value of these goods runs in several billion dollars. In addition, a number of applications are identified in the fields of agriculture and environment. In the initial years major emphasis was on the use of isotope gamma sources because of their availability as by-products from nuclear reactors and the large penetration offered an easy means for irradiation of thicker materials.

However, in recent years, electron beam accelerators (EB) have emerged as preferred alternative for industrial processing as they offer the advantages over isotope radiation sources like a) increased public acceptance since the storage, transport and disposal of radioactive material is not an issue b) ability to hook up with the industry for online processing c) provide higher dose rates resulting in high throughputs. An upper limit in the energy i.e. 10 MeV for electrons and 5 MeV for the x-ray sources has been imposed for the accelerators used for radiation processing to ensure that no radioactivity is induced in the irradiated material.

More than 1000 accelerators are in commercial use world wide, in the fields of polymerization and polymer modifications alone. Efforts are on to employ the accelerators in economically beneficial way for radiation sterilization and food irradiation. Since the penetration of electrons and machine production capacity for a given application is specific to the industrial needs, a single accelerator or one type of accelerator can not meet diverse requirements. Hence a careful evaluation needs to be carried out to select particular type of accelerator for a particular application.

2. ACCELERATOR PARAMETERS

The accelerators used for radiation processing extract the beam in a larger area defined by beam width or scanning width which is typically between 0.5 to 2 m. The product is conveyed to and fro under this zone to get required dose using suitable product conveyors. The electron penetration is proportional to the energy and inversely proportional to the product density. This is the basic formula which describes penetration.

Penetration (cm) = $(0.524\text{E} - 0.1337) / \rho$

E = beam energy in MeV and $\rho =$ density in g/cc.

This equation applies to energies greater than 1 MeV. The most important of these is water as most things which are being irradiated can be regarded as the same density as water.

The main parameters of interest in electron accelerators are energy and current. The energy decides the thickness of the product over which it can be uniformly irradiated and the dose rate at which the process can be irradiated is decided by the current. The process thickness which is defined as the depth at which the dose equals the entrance (surface) dose is a crucial parameter to be evaluated for the material of interest for selection of appropriate beam energy. To increase the process thickness, the product is irradiated from two opposite sides. The following expression gives the relation between the process thickness and the energy [1].

E = 2.63 dp + 0.32 (one-side irradiation) E = 1.19 dp + 0.32 (two-side irradiation)

2.1. Classification of accelerators

Accelerators used for radiation processing are classified in three categories based on the energy.

a) <u>Low energy</u>: The accelerators in the range 400 keV to 700 keV come in this category. Even low energies in the range 150 keV to 350 keV, single gap, non scanned beams with extended electron source and beam currents from a few mA to more than 1000 mA are available for surface curing applications. In the 400 to 700 keV range, the beam currents are available from 25mA to approximately 250mA This type of equipment is available in widths from approximately 0.5 meters up to approximately 1.8 meters in width. All the low energy accelerators are generally self-shielded type and the applications are found in the areas of surface curing of thin films, laminations, production of antistatic, antifogging films, wood surface coatings etc. These units are available in widths from less than one meter to more than two meters. The maximum range of penetration could be up to 60 mg/cm².

b) <u>Medium Energy</u>: Scanned beam systems with energy range 1 MeV to 5 MeV fall in this category. This type of equipment is available in widths from 0.5 to 1.8 meters. These units are characterized by beam powers from 25 kW to 150 kW. These units are used for a range of applications from cross-linking of thicker cross sections of materials to polymer rheology modification, colour enhancement of gem stones, for sterilization of medical products and food irradiation (to a limited use) because of higher penetration ranges. Typical penetration depths in unit density material will be in the range 5 to 25 mm.

c) <u>High energy</u>: The accelerators having energy range 5 to 10 MeV provide the highest penetration thickness and are best suitable for bulk product irradiation. Scanned beams of power levels 25 kW up to 350 kW are available with widths up to 1.8 m. With penetration depths for 10 MeV electrons typically being 50 cm (when irradiated from both sides) for 0.15g/cc product density, this category of accelerators are commonly used for medical product sterilization, cross-linking of thick section products, food disinfestation, waste water treatment, Polymer rheology modification, colour enhancement of gem stone and shelf life extension for food & fruits etc.

2.2. Type of accelerators

Accelerators are made in two type's viz., DC type where a constant beam is extracted; and RF pulsed type where the output beam is of pulsed nature. But both, DC and RF accelerators have become the work horse and are extensively employed. DC accelerators give high average beam power whereas the RF accelerators, generally operated in the pulsed mode, give low average power. On the other

hand RF accelerators have high energy gain per unit length, thus more compact in construction, as compared to the DC accelerators.

Low energy d.c. accelerators from NHV, Japan produce the high voltage through raising the conventional power frequency voltage by a transformer and use cascaded voltage doubler rectifier circuit to increase the output voltage. These equipments are typically supplied with gas filled power supplies as well as with oil-filled power supplies. The accelerator systems are self shielded and the product irradiation geometry forms part of the system. A vacuum chamber with an extended (small) cathode filament on heating gives out thermionic electrons all along the length of the filament which are accelerated and extracted (in case of small cathodes, the beam is made to scan) through a thin titanium foil called window foil. Provision for inert atmosphere during irradiation is made in the system.

In case of medium energy d.c. accelerators, machines are based on transformer type, modified cockcroft-walton or Dynamitron to produce high DC voltage and an acceleration tube in which electron from a small heated cathode are accelerated. These are widely operating for mainly polymer crosslinking applications. The cockcroft-walton accelerator basically consists of a cascade generator and a capacitors assembly to create high DC voltage. NHV, Japan produced accelerators based on modified cockcroft-walton circuits. M/s. Radia Ind. Japan installed 5 MeV, 150 kW accelerator made by NHV, Japan, which can also operate in x-ray mode using tantalum target. Medical products are sterilized with 5 MeV electrons using double sided irradiation up to 4.5 g/cm² thickness and 50 cm. using x-rays with a bulk density of 0.16 g/cc. The Dynamitron d.c. accelerators from RDI, USA, the most wide spread are available in the energy range from 0.55 to 5 MeV with powers up to 200 kW. These machines use charging circuit based on a capacitor similar to that of cockcroft - Walton accelerator. A number of accelerators namely ELV (dc type) and ILU-6 (pulse) have been built by INP, Russia [2]. The ELV d.c. accelerators has cascade generator with a parallel inductive coupling as the high voltage source and employ a coreless step transformer with sectionalized secondary coil. These accelerators are available in the energy range 0.2 to 2.5 MeV with the beam currents up to 200mA. More than 70 accelerators have been supplied in and outside Russia that includes Poland, China, and Korea etc. Pulse linear accelerators ILU series offer electron beams from 0.5 MeV to 5 MeV up to powers 50 kW are based on single resonator cavity with one accelerating gap. In Dynamitron and ELV type accelerators, the current ratings are essentially independent of their voltage ratings. Electrons after accelerating through a number of electrodes in an accelerator column are focused, scanned and extracted through the window foil. Vacuum is maintained in the accelerator column. The equipment is always supplied with gas filled power supplies and takes time to bring back on line in case of failure in power supply or vacuum system. This is due to conditioning time required to re-establish the high voltage and the vacuum system. The cost of SF₆ gas, its availability and the safety precautions during transfer are some of the problems normally faced.

High energy RF accelerators, linacs, deliver powers up to 50 kW are available. The RF based accelerators - Rhodotrons, from Ion Beam Applications, IBA, and Belgium are the latest state of the art machines based on the principle of "re-circulating" a beam through successive diameters of a single coaxial resonating cavity (2m diameter). Such design makes it possible to achieve CW acceleration of electron beams to high energies. With energy gain of 1 MeV per crossing, five or ten successive crossings will obtain energies 5 MeV or 10 MeV respectively delivering powers up to 200 kW.

All the systems today are supplied with a PC controlled /PLC based control system with the specific type or manufacturer of the PLC specified by the buyer. The systems are rugged, designed for round the clock operation with minimum down time, offering efficient troubleshooting through internet communication. Under beam product conveying systems for uniform dose delivery to the products and faster throughputs are equally important that has to go hand in hand with the accelerator systems. These can be interlocked to the operating systems. Various product conveying methods and the gadgets are offered to the users depending upon the application requirements.

The industrial accelerators and the applications being pursued in India are given in the following table.

TABLE I.	TABLE : ELECTRON ACCELERATORS FOR RADIATION PROCESSING
APPLICATIONS	S IN INDIA

S.N.O	Accelerator Details	Under beam	Status	Applications
1.	2 MeV, 20 kW, pulsed ILU-6 type (INP, Russia) with linear and four- window scanning, at BARC, Navi Mumbai	Power roller conveyor system, linear mesh conveyor, 'eight type' cable irradiation conveyor, four sided cable irradiation conveyor, water-cooled irradiation target,	In operation	Multi purpose, Technology demonstration plant R&D and Commercial Service in Crosslinking °radation of polymers, coloration of diamonds etc.
2.	500 keV, 10 kW, self-shielded (indigenous), at BARC, Mumbai	continuous thin film transport conveyor	In operation	R&D, surface curing of thin films, sheets
3.	300-750 keV, 20 kW (max), shielded cell, (indigenous), at CAT, Indore	linear conveyor, tray / sheet transport mechanism	In operation	R&D, Surface curing, viscose pulp degradation
4.	3 MeV, 150 kW, Dynamitron (RDI, USA) at M/s. NICCO Corporation Ltd, Sham Nagar	wire & cable irradiation conveyor, pallet box conveyor	In operation	Multi purpose contract facility, Commercial Service in Crosslinking °radation of polymers, coloration of diamonds etc.
5.	1.5 MeV, 20 kW, ICT type, VHV, M/s. Radiant Cables, Hyderabad	wire & cable irradiation conveyor	In operation	Cable irradiation
6.	750 keV, 10 kW (Russia), self- shielded, at Visakhapatnam	Linear conveyor	In operation	Surface curing
7.	3 MeV , 30 kW cockcroft-walton (indigenous) at BARC, Navi Mumbai	Power roller conveyor system, linear mesh conveyor, 'eight type' cable irradiation conveyor	Under construction	Multi purpose, Technology demonstration, R&D, Commercial Service
8.	10 MeV, 10 kW, RF linac (indigenous) at BARC, Navi Mumbai	'Toway' cart type conveyor	Under construction	Multi purpose, Technology demonstration, R&D, Commercial Service

3. ACCELERATOR PROGRAMME IN INDIA

Electron Beam Centre (EBC) at BARC, Mumbai: DAE, India had realized the potential applications of these beams and chalked out an elaborate programme for developing the accelerators suitable for radiation processing applications. BARC has set up an Electron Beam Centre (EBC) at Kharghar, Navi Mumbai. The centre will house a 3 MeV and a 10 MeV machine. EBC is being established as a novel & unique centre in the country for carrying out R & D and applications in the area of industrial accelerators for radiation processing [3].

A brief account of the accelerator programme for radiation processing is given below.

1) 500 keV DC Accelerator: The accelerator is a Cockcroft-Walton based DC accelerator designed to give 20 mA of electron beam at energy of 500 keV. The accelerator is a self-shielded type and housed at BRIT complex, Vashi, Navi Mumbai. This is the first indigenously developed accelerator in the country, operational at a voltage of 450 kV and power level of 5 kW. Its up

gradation to full power level and complete computerized operation to a single push button machine, are in progress. The regular & routine operation is being carried out at a power level of about 3.5 kW and energy 450 keV. The accelerator is being used for surface modification studies.

2) 750 KeV Accelerator at CAT, Indore [4]: This is a 15 stage cascade multiplier Cockcroft-Walton based DC accelerator that can be operated in the range 300-750 keV. This has been housed in a shielded room so that it can be used for crosslinking of cable insulation, viscose pulp degradation. Two different product conveyor systems have been installed for the applications.

3) 3 MeV, 30 kW DC Accelerator: This accelerator is designed to deliver 30 kW of beam at energy of 3 MeV. The 5 keV beam from the electron gun is accelerated to 3 MeV by passing it through the accelerating tubes. The beam, after traversing through the scan magnet chamber is let out in the air through the scan horn and is used for processing of the materials. The high voltage column comprising of the corona guard rings are connected through the rectifier chains, each of which generates an effective voltage of about 50 kV. In all, there are 70 such chains giving rise to the required voltage of 3 MV. The accelerator tank having a diameter of 2 m and a length of 7m is pressurized with SF6 at a pressure of 6 atmospheres. In addition, the cooling of SF₆ needs special attention because of the severe heat generated in the accelerator tank. The challenge of designing and building a 45 kW, 120 kHz RF oscillator is also being met for the first time.

4)10 MeV, 10 kW RF Electron Linac: It is a coupled cavity linac, capable of giving 10 MeV beam with a power of 10 kW. The 50 keV electron beam from the gun is accelerated to 10 MeV in the cavity having a length of about 1 m. After the energy analysis, the beam is passed to the scan horn through the sweep scanner. The cavity is powered through a wave guide based plumbing line consisting of a circulator, load, directional coupler, power divider, ceramic window etc. The line is fed by a klystron based microwave source which will deliver a peak power of about 6 MW at a frequency of 2856 MHz. This microwave source is being designed and built by SAMEER, Mumbai. It is designed to deliver an average power of 24 kW with a duty factor of 0.4 %. The rest of the systems are similar to the 3 MeV machine.

4. CONCLUSIONS

A wide range of accelerators are available for radiation processing applications with the state of the art technology. These are designed to work round the clock with minimum number of personnel and down time for maintenance. The availability of such accelerators at affordable costs, will definitely help the users to adapt EB technology not only for applications producing value-added products like wire &cables, heat shrinkable, colored diamonds, health care products but also for food preservation and environmental applications in an economically beneficial way.

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RESTRICTIONS AND LIMITS OF ACCELERATOR TECHNOLOGY APPLIED IN INDUSTRY AND ENVIRONMENT PROTECTION

Z. Zimek

Department of Radiation Chemistry and Technology, Institute of Nuclear Chemistry and Technology, Warsaw, Poland

Abstract. Successful radiation technology implementation has been obtained in some specific areas like radiation sterilization, polymer and semiconductors modification. The radiation technology as any other modern technology should overcome certain barriers to achieve successful commercialization. Careful technical and economical evaluation has been performed to obtain information about commercial and national profitability of any radiation technology project before its final approval and implementation. The economical effect of radiation installation is influenced by technical accelerator characteristics. The quality and life time of accelerator main components such as electron gun, RF source, output window foil, pulse power components have the most important influence on accelerator reliability. The accelerator life time can be easily extended up to 20-40 years period if the maintenance service is performed correctly.

1. INTRODUCTION

The accelerators are the principal equipment which defines basic technical and economical parameters of any radiation facility. Over thousand accelerators have been used for radiation chemistry, R&D and radiation processing as the result of continuous progress in accelerators construction and radiation technology development [1, 2]. Beside of radiation sterilization and modification of polymers, which were introduced as an industrial process in mid 50'ties, the wide range of radiation processing technologies have been developed and applied since than. The most common are surface curing, semiconductor modification, teflon degradation, food processing and others. Creation of close circuit processes is one of the significant features of modern technologies where industrial wastes are utilized at the same production cycle. EB process may effectively support such solutions as it was happen in flue gas treatment where SO_2 and NO_x pollutants are being converted into usable fertilizer [3]. The advantages of radiation process are frequently related to its high efficiency and possibility to transfer high amount of energy directly into the object under radiation treatment. Disadvantage is mostly related to high investment cost of the accelerator. It may be effectively overcome in future as the result of accelerator technology development. On the other hand radiation technology effectiveness can be characterized by acceptable price (investment cost) of 1 W beam power provided by accelerator for certain applications (TABLE I).

Acceptable price of 1 W electron beam power	Type of radiation process	Product characteristics
100-250 \$/W	Semiconductors modification	Low dose Small scale High unit price
50-100 \$/W	Radiation sterilization	Medium dose Large scale Medium unit price
<2.5 \$/W	Flue gas treatment	Low dose Very large scale No commercial value

TABLE I.RADIATION PROCESS EFFECTIVENESS

Generally radiation processes are being applied to obtain: new or improved product which can not be done conventionally or traditional technique is more expensive, to conserve energy, reduce pollution, to provide better products (quality, durability, performances), to use less raw material. Finally the success of radiation technology commercialization depends on technology development, equipment capability, market conditions and economical aspects related to the facility exploitation as it was illustrated in Table II. It can be easily noticed that accelerators performances became significant part of responsibility related to successful implementation of any radiation process.

Barriers	Subject	Responsibility
Technology	Scientific basis Technology specification Know-how	Scientists/researchers
Equipment	Accelerator parameters Under beam equipment Auxiliary equipment	Equipment producers
Marketing	Right product Right time Right place	Advertising and promotion
Economy	Investment cost Exploitation cost Profit expectation	Product manufactures

TABLE II. COMMERCIALIZATION OF RADIATION TECHNOLOGY

2. INDUSTRIAL ACCELERATORS AND THEIR PRODUCERS

The electric field acts on electrons as charged particles and gives them energy equal to potential difference across the accelerating gap. That is the basic principle of any accelerator construction. The major difference between different types of accelerators is the method by which the electric field is generated. Industrial electron accelerators can be divided on three groups depends on their construction:

- direct accelerators where high voltage is used to accelerate electrons,
- single resonant cavity accelerators working at a frequency from one to several hundred MHz,
- linear accelerators microwave range where number of resonant cavities is used to accelerate electrons.

The number industrial and scientific organizations have a capability to build accelerators. The main producers are located in USA, Japan, Belgium, France and Russia. In many others countries including China and Poland such capabilities exists as well. The new tendency can be observed in many developing countries where accelerator oriented projects became quite frequent. The most experienced accelerator manufacturers are listed bellow:

- Direct transformer accelerator manufacturers:
- ESI Energy Science, USA,
- RPC Technologies, USA,
- HVEC High Voltage Engineering Co, USA,
- Wasik Associates, USA,
- NHV Nissin High Voltage, Japan,
- SHI Sumitomo Heavy Industries, Japan,
- Institute of Surface Modification, Germany,
- INP Institute of Nuclear Physic, Russia,

- SIEA Research Institute of Electro Physical Apparatus, Russia,
- Vivirad, France,
- Research Institute of Automation for Machine-Building, China,
- Institute of Nuclear Studies, Establishment for Nuclear Equipment, Poland.

Single resonant cavity accelerators manufacturers (100-200 MHz):

- INP Institute of Nuclear Physic, Russia,
- IBA Ion beam Application, Belgium,
- Denki Kogyo Co, Japan.

Linear electron accelerators manufacturers (microwaves 1.3-9 GHz):

- Varian USA,
- Titan, USA,
- RPC Technologies, USA,
- Mitsubishi Heavy Industries, Japan,
- Technical System, United Kingdom,
- Thomson CSF, France,
- IBA Ion beam Application (Scanditronix), Belgium,
- SIEA Research Institute of Electro physical Apparatus, Russia,
- RIA TORYI, Russia,
- Research Institute of Automation for Machine-Building, China,
- Institute of Nuclear Studies, Establishment for Nuclear Equipment, Poland.

The main achievements of industrial accelerators development are presented in Table III.

TABLE III. ACCELERATORS FOR RADIATION PROCESSING

Parameter	Direct DC	Single cavity 100-200 MHz	Linear 1.3-9 GHz	
Electron energy	0.05-5 MeV	0.3-10 MeV	2-10 MeV	
Average beam current	<1.5 A	<100 mA	<100 mA	
Average beam power	500 kW	700 kW	50 kW	
Electrical efficiency	60-80 %	20-50 %	10-20 %	

3. SELECTION CRITERIA

Although there are many different types of accelerators offering a wide range of performance ratings, only few would be suitable for particular application. The general requirements should be formulated in advance before accelerator selection is performed. The most important requirements are listed bellow:

- product to be treated dimensions, densities and throughputs,
- operational schedule and seasonal requirements,
- vertical or horizontal beam direction,
- reliability of the accelerator (availability),
- remote accelerator operation,
- factory assembling test,
- warranty conditions,
- post warranty services,
- staff training,
- facility certification (equipment, safety, personnel).

Table IV describes the most important criteria of accelerator selection. The basic specification for electron energy and beam power should be derived from the process requirements (absorbed dose distribution, the product size, the shape and density, and the throughput rate) to ensure satisfactory results with minimum capital and operating costs. Table V describes accelerator and facility basic parameters which should be correlated with particular process requirements.

TABLE IV. CRITERIA OF ACCELERATOR SELECTION

No	Criterion of selection	Remarks
1.	Fundamental accelerator parameters: - Electron energy - Average beam power	The basic requirements which define technological capabilities and facility productivity
2.	Terms of accelerator purchase: - Price, - Producer, - Terms of delivery and installation, - Warranty conditions, - Exploitation cost.	Economical aspects of accelerator purchase which define investment and exploitation costs; period of time needed for facility completion
3.	Auxiliary accelerator parameters: - Scan performances, - Auxiliary parameters, - Measures and control, - Main components and systems - Auxiliary components and systems - Accelerator external supply service	Auxiliary parameters which may characterize accelerator quality and provide necessary data for facility design

TABLE V. ACCELERATOR AND FACILITY BASIC TECHNICAL SPECIFICATIONS

Electron energy	Beam power
Nominal energy [MeV],	Nominal average beam power [kW],
Electron energy adjustment range [MeV],	Nominal average beam current [mA],
Energy instability [%],	Beam current instability [%],
Energy spread [%],	Type and range of beam current setting,
Energy single selection action,	Beam pulse repetition frequency (min/max),
Energy day to day reproducibility [%], Energy day to day reproducibility [%/24h]	Beam pulse parameters. Continuous electron energy evaluation.

Advantages of certain accelerator construction which should have influence on accelerator selection are connected to:

- application of proven accelerator technology,
- simplicity of accelerator construction,
- long life power components (klystron, tetrode),
- high parameters stability,
- high beam power,
- narrow energy spread,
- wide range of beam power adjustment,
- computer supported control system,
- low accelerator cost,
- low exploitation cost,
- high quality maintenance service.

Disadvantages of certain accelerator construction leads to higher risk and troubles during accelerator exploitation and reduces economic results of radiation process implementation. The most important accelerator disadvantages are listed bellow:

- prototype accelerator construction (limited exploitation experience),
- parameters on the edge of present limits,
- power components with limited life time (magnetron),
- low average beam power,
- high electric energy demands,
- poor accelerator availability,
- high total cost,
- difficulties in spare parts availability.

4. ACCELERATOR EXPLOITATION AND SERVICING

The reliability of the accelerator is one of the most important features of any industrial facility. The ability to work can be characterized by number working hours per break or accelerator availability in percent of total working time. Accelerator reliability depends on many separate accelerator parameters and details of its construction like:

- components quality (life time of main accelerator components),
- circuit design,
- principle of accelerator operation and its construction,
- time of continuous operation (8 h/day; round o'clock).

The quality and life time of accelerator main components like electron gun, RF source, output window foil, pulse power components and other accelerator consumable parts have the most important influence on accelerator reliability. The specific technical problems are connected to output window foil behavior during intense exploitation with high beam current level. The window foil lifetime is related to its temperature and thermally induced stresses. 2000 h is the typical titanium window lifetime value for full power continuous operation. The recent progress in developing new composites for window foils may significantly increase the permissible beam current density level in future. The construction of window system with water cooled grid support for high energy electron accelerators with improved electron transparency and reduced beam power loses due to more efficient electron optic parameters of the system can be best solution to eliminate implosion caused by foil breakdown [4].

Typically industrial accelerator availability is at 95 % level. Better results can be obtained as it was described in literature [5], where 4 h of unscheduled down time per week and 3 times of down time longer than 10h per 6 months was obtain with total accelerator availability 98% up time. Regular

scheduled service is required to reduce risk of unexpected breakdown. Typically regular service should be performed every day; week; ¹/₂ year and year. The detail schedule of such activity depends on accelerator construction and usually is recommended by accelerator manufacturer. Accelerator maintenance can be performed in house or by accelerator manufacturer personnel. The first approach requires highly trained local specialists what is usually recommended. That is related to higher accelerator operation cost what may be compensated by better accelerator availability. The second approach is more expensive but allows overcome the shortages of well educated personnel. The accelerator life time can be easily extended up to 20-40 years period if the maintenance service is performed correctly. The spare parts availability and accelerator parameters become the limiting factors in such case. Accelerator upgrading is sometime suitable solutions when electron energy or beam power increase is needed [6]. Power components replacement is usually required in such case. Accelerator upgrading is connected by accelerator manufactures. The second hand accelerators are offered with moderate price and sufficiently good technical condition. Quite frequent accelerator facility upgrading is connected to computer installation for accelerator parameters measurement and technological equipment control.

5. COST ANALYSIS

Economic and financial evaluation is necessary to estimate commercial and national economic profitability of the project connected to accelerator application before its final approval and implementation. Economic evaluation should include structure of investment and operating cost of radiation facility. The economical effects of radiation installation may be influenced by many factors.

The most important are the following:

- investment cost (accelerator, auxiliary equipment what depends on the accelerator type, monitoring and process control systems, material handling system, building including radiation shielding, project preparation, engineering),
- operating cost (financial cost debt service, maintenance and spare parts, personnel labor, utilities electricity, water, air, etc.),
- utilization of electron beam and dose setting.

The investment costs are always high for any electron beam facility, mainly because of the accelerators price and costs of the building with special biological shield. There is also auxiliary equipment needed like conveyer and/or under beam equipment, cooling and ventilation systems, control and monitoring system. Usually low energy accelerators may offer the lowest capital cost (cost of accelerator, shield) but with low electrons penetration level what causes loss of flexibility.

The debt service is often based on paying off a loan due to investment cost in 5-15 years at certain interest. The administration cost covers management, administration and quality service (dosimetry). Labor cost is related to operator, maintenance and conveyor service personnel.

When the utilization of electron beam is concerned the following factors are to be taken into account: dose distribution (as a function of energy and beam current), type of irradiated items (complex product geometry, material interfaces and nearby surfaces), uniformity of conveyer speed, and the design of the carrier system. The dose distribution in irradiated product with electron beam has nonlinear characteristics. To keep the agreed level of heterogeneity in irradiated object, a part of electron beam energy is not used, which can be a little bit improved by two sided irradiation (up to 20%).

6. FINAL REMARKS

Appropriate accelerator selection should be performed to meet all technical and economical conditions for successful process implementation. Accelerator quality is usually high but certain steps should be taken to obtain industrial standards for accelerator technology and achieve good accelerator reliability. It should be mentioned that accelerator quality is defined by its weakest component and because of that life time of certain accelerator components should be extended.

Commercialization of accelerator technology may be profitable in long term to generate commercial and national profitability. The government policy should promote this technology particularly at the stage of environmental application when risks and benefits are equally important.

Spear parts and major maintenance services are provided usually by accelerator manufacturer. On the other hand computer control system is designed to perform automatic operation of accelerator facility. Due to above the highly trained personnel is not required to run modern accelerator.

High frequency accelerators are more costly due to their more complex construction and much more expensive spare parts like klystrons or magnetrons. This can be compensated by higher affordable electron energy what makes implementation of such accelerators more flexible.

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PRESENT STATUS AND EXPECTED PROGRESS IN RADIATION PROCESSING DOSIMETRY

A. Kovács¹, A. Miller²

¹Institute of Isotope and Surface Chemistry, Chemical Research Center of the Hungarian Academy of Sciences, Budapest, Hungary

²High Dose Reference Laboratory, Risø National Laboratory, Roskilde, Denmark

Abstract. The paper describes the present status of radiation processing dosimetry including the methods used most widely in gamma- and electron processing as well as the new methods under development or introduction. The recent trends with respect to calibration of routine dosimetry systems as well as standardization of dosimetry methods and procedures are also described.

1. INTRODUCTION

Radiation processing has become a worldwide accepted industry both in developed and in developing countries including well established technologies like sterilization of medical products and pharmaceuticals and polymer processing as well as emerging technologies such as food irradiation and environmental and biomedical applications. Some of these technologies do not need exact doses, but in most cases knowledge of the dose applied to achieve the necessary effect is needed. This knowledge is also required in order to avoid at the same time damage to the product. The main application fields of radiation processing dosimetry include installation qualification, operational qualification, process qualification and routine process control.

Beside routine dosimetry applications, recent trends involve the improvement of calibration procedures of dosimetry systems and applicability tests of existing systems. The introduction of new dosimetry systems and methods satisfy dosimetry requirements of new technologies (for example irradiation of products at low temperatures, use of X-ray processing, environmental processes, etc.). Standardization of the most important and reliable systems and procedures is carried out by international organizations.

2. PRESENT STATUS OF RADIATION PROCESSING DOSIMETRY

2.1. The role of dosimetry

Quality assurance in radiation processing technologies relies on well established dosimetry systems and procedures, which guarantee the correct execution of these technologies. Thus traceable measurements of absorbed dose are used to document the successful execution of the irradiation processes. Consequently dosimetry - as part of the total quality system - is an independent, inexpensive and reliable tool to control the irradiation process, and dosimetry plays an important role in the transfer of processes from the laboratory to the industrial stage. Dosimetry provides documentation in these processes if the measurement is traceable to a national standard and if the uncertainty of the measuring system is known. Several factors can affect dosimeter accuracy such as storage conditions or instrumental errors, but suitable calibration procedures (both that of the routine dosimeter and of the analytical instrumentation) are aimed at controlling these potential sources of dosimetry errors.

Various liquid and solid phase dosimetry systems are available to fulfill the dosimetry requirements of the different application fields in radiation processing. These systems can be categorized as primary standard, reference and transfer standard and routine dosimeters. The selection of a suitable dosimetry system depends on the dose range of the irradiation process to be carried out, i.e. on the process to be controlled, as well as on various characteristics of the systems (dose, dose rate and energy dependence, effects of temperature before, during and after irradiation, humidity, light, stability, reproducibility, size, resolution, simplicity of handling, read-out procedure, availability, cost, batch variability, shelf life).

2.2. Application fields and dosimetry systems

Dosimetry plays a key role in the qualification of gamma - and electron irradiation facilities (a), in the qualification of the irradiation process and product (b) and in the routine control of the irradiation technology (c) [1].

a./ Installation Qualification (IQ) is carried out at the new facility to demonstrate that it has been delivered and installed in accordance with specifications. Operational Qualification (OQ) is carried out at regular intervals or after changes that can influence dose or dose distribution to demonstrate that the facility can consistently deliver the radiation process. One of the aims of these procedures is to characterize the irradiation facility relating plant parameters to absorbed dose measured e.g. in a reference product. In the case of a gamma irradiation facility it results in e.g. the determination of nominal dose vs. dwell time using dosimeter systems such as the Perspex "family" [2], radiochromic films (FWT-60 or GEX WinDose)[3], potassium dichromate [4], ethanol-monochlorobenzene (ECB) [5] or ceric-cerous solutions [6], alanine [7], etc. In the case of electron beam machines facility qualification can involve the measurement of electron energy, scanning width and homogeneity and beam spot using mainly film dosimeters (FWT-60, B3 [8], cellulose triacetate (CTA) [9]). The relationship between nominal dose and conveyor speed is determined applying process calorimeters [10], or systems such as alanine, ECB, dichromate, ceric-cerous or film dosimeters.

The other aim of IQ/OQ is to measure dose distribution in a reference product. In electron beam facilities film dosimeters are used for this purpose, while in gamma facilities the use of both liquid and solid phase dosimeters is possible.

b./ The aim of process definition is to determine the minimum dose needed in order to obtain the necessary effect (sterilization, for example) and to determine the maximum dose that can be tolerated by the product and its packaging material. The same type of dosimeter systems can be used for these investigations as mentioned above after suitable selection.

In Performance Qualification (PQ) the product loading pattern has to be determined and the dose mapping of the product has to be carried out using any of the dosimetry systems mentioned above depending on the type of irradiation. Finally the routine monitoring position of the regular irradiation process has to be selected establishing also the relationship between the measured minimum dose in the product and the dose measured at the selected monitoring location.

c./ During routine product irradiation dosimetry is used to demonstrate and document that the irradiation process is under control within specified confidence limits. Routine dosimeters (e.g. Perspex systems, ECB, ceric-cerous, radiochromic films, CTA) are used to measure absorbed dose at regular intervals. In the case of gamma irradiation facilities regularity is often defined as the presence of a specified number of dosimeters at the same time in the irradiation room. In the case of electron beam facilities the routine dose can be measured on the conveyor at specified time intervals, for example every 10 - 15 minutes.

The dosimetry systems used most widely in present radiation processing practice are shown in Table 1.

Dosimeter system	Method of analysis	Useful dose range (Gy)	Nominal precision limits	Reference
Process calorimeters	Temperature	$3 \times 10^3 - 5 \times 10^4$	2 %	[10]
	measurement			
Ceric-cerous sulphate	UV	$10^3 - 10^6$	3 %	[6]
	spectrophotometry,			
	potentiometry			
Potassium	UV-VIS	$5 \times 10^3 - 4 \times 10^4$	1 %	[4]
dichromate	spectrophotometry			
Ethanol-	Titration or HF	$4 \times 10^2 - 3 \times 10^5$	3 %	[5]
monochlorobenzene	oscillometry			
Perspex systems	VIS	$10^3 - 5 \times 10^4$	3 %	[2]
	spectrophotometry			
α-L-alanine	EPR	$1 - 10^5$	0.5 %	[7]
Cellulose triacetate	UV	10^4 - 10^6	3 %	[9]
	spectrophotometry			
FWT-60 film	VIS	$10^3 - 10^5$	3 %	[3]
	spectrophotometry			-
B3 film	VIS	$10^3 - 10^5$	3 %	[8]
	spectrophotometry			_

TABLE I.CHARACTERISTICS OF THE DOSIMETRY SYSTEMS USED MOST COMMONLY INRADIATION PROCESSING

3. RECENT TRENDS IN RADIATION PROCESSING DOSIMETRY

The introduction of new technologies such as electron irradiation at low energies (< 150 keV), X-ray treatment, food irradiation at low temperatures, radiation treatment of pharmaceuticals, environmental applications (waste or drinking water treatment) and stricter requirements on improved quality assurance have resulted in novel approaches with respect to dosimetry methodology and calibration and efforts are on the way to harmonize the existing dosimetry standards for increased international use.

3.1. Novel approaches

3.1.1. New or improved dosimetry methods

a./ Systems based on fluorimetric evaluation

The use of fluorimetric dosimeters is based on the measurement of optically stimulated luminescence (OSL) due to the formation of new fluorescent radiolysis products (e.g. in organic molecules) or radiation induced colour centers (e.g. in inorganic materials) or due to the radiation induced decay of originally fluorescent molecules. The major advantage of this method is its high sensitivity and wide dynamic range. Because of the advantages these rather inexpensive detectors can be used both at medical diagnostics, radiation processing, space studies, etc.

The OSL based Sunna dosimeter film [11] is a new development with potential application in many radiation processing technologies due to its wide dose range (50 Gy - 300 kGy). It is a polyethylene based film containing microcrystalline dispersion of LiF and the formation of the

radiation induced colour centre (M-centre) in the LiF is utilized for dosimetry by measuring one of the two emission bands (530 nm and 650 nm) in the irradiated dosimeter film due to optical excitation at 450 nm (Fig. 1.). Since the red emission (650 nm) was shown to be affected by irradiation temperature as well as dose rate, its use is restricted. By measuring, however, the green emission (530 nm) with a table top fluorimeter, the detector was shown to be suitable as routine dosimeter for food irradiation (200 Gy - 15 kGy) in the temperature range of 0 °C - 40 °C in the cases of gamma-, electron- and X-ray radiation [12]. Dosimetry intercomparison experiments carried out in industrial gamma and electron irradiation facilities have further proven the applicability of this dosimeter for sterilization of medical products (5 - 50 kGy) [13]. Recent investigations revealed the possibility that measurement of the infrared emission of the irradiated dosimeter has very promising dosimetry characteristics in the dose range of 10 Gy - 10 kGy.



FIG. 1. Excitation (450 nm) and emission spectra (530 nm and 650 nm) of Sunna film.

By measuring the optical absorbance of the irradiated films at 240 nm dose determination was shown to be possible with spectrophotometry in the dose range of 5 - 100 kGy. This evaluation method using a specifically designed routine UV reader is advantageous due to the stability of the response of the irradiated dosimeters for days after irradiation [13].

b./ Systems based on the measurement of optical absorption

Tetrazolium solutions were recently found to be useful for dosimetry purposes. Tetrazolium salts are heterocyclic organic compounds, which upon irradiation yield highly coloured water insoluble formazans due to radiolytic reduction. Different tetrazolium salts such as the 2,3,5-triphenyl-tetrazolium chloride (TTC) [14], tetrazolium blue (TB) [15] and the nitro blue tetrazolium (NBT) [16] have been found useful for gamma and electron dosimetry purposes both in solutions and in solid matrices. An aqueous solution of NBT was found suitable to measure low doses (0.1 - 1 kGy) and high doses (1 - 30 kGy) due to the formation of monoformazan first (522 nm) followed by the formation of diformazan (612 nm).

A polyvinyl alcohol (PVA) based TTC film was found suitable for dose measurement in the range of 1 - 100 kGy by measuring the absorbance at 493 nm [14]. PVA based NBT films showed also good performance for dose measurements as routine dosimeters in the dose range of 5 - 50 kGy by measuring the absorbance of the irradiated films either at 540 nm or at 580 nm [17]. Using reflectometric evaluation with a simple hand-held reflectometer, i.e. by measuring reflected light from the irradiated TTC or NBT films (Fig. 2.) these dosimeters can be applied also as label systems together with other films such as the B5 label dosimeter from Risø National Laboratory [17].



FIG. 2. Change of absorbed and reflected light of NBT film as a function of absorbed dose.

c./ Systems based on conductivity evaluation

The conductivity of certain dosimetry solutions (such as the ethanol-monochlorobenzene and the aqueous alanine solution) changes due to irradiation. By applying a high frequency oscillator circuit with a capacitive cell a relative measure of the conductivity change of the dosimeter solution can be obtained, which - after suitable calibration - can be used for dosimetry purposes [5]. Since the measurement is carried out in sealed ampoules the method is a non-destructive one thus making possible the re-evaluation of the dosimeters at any later times due to the stability of the solutions. Applying this method the ECB solution can be used for dose measurement in the dose range of 1 - 300 kGy, while the alanine solution can be used in the 1 - 100 kGy range [18]. The construction of a portable, digital, programmable and robust table-top oscillometric reader has resulted in even faster and simpler evaluation of the irradiated dosimeter ampoules (Fig. 3.).



FIG. 3. Oscillometric reader for evaluation of ECB dosimeters

Recent investigations have shown promise to use polyaniline based polymer composites (as "conducting polymers") for dose measurement using similar high frequency oscillator circuit as mentioned above. The preliminary results indicated the possibility of measuring doses with polyaniline based polyvinylchloride (PVC) and polypropylene chloride (PPCL) in the 5 - 150 kGy and 5 - 200 kGy dose ranges respectively [19].

d./ Development of calorimetric systems

Process calorimeters using water, graphite or polystyrene have been developed and tested for the calibration of routine dosimeters as well as for nominal dose measurements at high and medium energy (1.5 - 10 MeV) electron accelerators used in radiation processing [10, 20]. Due to recent requirements using low energy accelerators (80 - 120 keV) for e.g. curing technologies or sterilization of packaging materials, totally absorbing graphite calorimeters are under development as primary standard systems for dose measurement and for calibration of routine film dosimeters to be used under such irradiation conditions [21].

e./ Semiconductors for routine dosimetry

Silicon diodes and other type of semiconductors have been used for radiation dosimetry for decades for the measurement of dose or dose rate. The use of such devices for routine process control in radiation processing have been studied in detail in the past decade. Recent investigations [22] using bipolar power transistors both in industrial gamma- and electron irradiation facilities together with transfer standard and other routine dosimeters have proved the suitability of their use for routine process control after the necessary calibration. The simple evaluation method (i.e. the measurement of a physical parameter which is directly related to the charge carrier lifetime) of the irradiated transistors with a portable, robust, table-top reader and the stability of the transistors after irradiation makes their potential use for routine dosimetry in the dose range of 50 Gy to 50 kGy promising.

3.1.2. New methodology

Beside the development of new dosimetry systems and the improvement of certain existing ones novel approaches can be observed with respect to methodology used in radiation processing dosimetry. Such an approach is the automation in dosimetry practice in order to reduce human errors. Automated evaluation systems are

under development and already available in optical absorption evaluation [23], in ESR evaluation of the alanine dosimeter system and a new software has been developed at Risø National Laboratory for the automated scanning and evaluation of images used e.g. in dose distribution measurements.

Environmental conditions and their combined effects can influence the response of dosimeter systems so, that the corrections with respect to calibration conditions are difficult to carry out. Current efforts to construct and use real time dosimeters to determine the combined effect of irradiation temperature and dose rate in industrial gamma irradiation facilities [24] are aimed at minimizing these problems.

Mathematical methods have been in use in radiation processing for decades e.g. for the determination of optimum source configuration before replenishment in gamma irradiation facilities or for dose distribution modelling in product packages. Due to recent technical developments more powerful tools are available for these calculations and modelling and new mathematical methods are under development and test to assist product design, dwell time setting, estimation of dose distribution [25].

3.1.3. Calibration of routine dosimeters

Reliable dosimetry requires knowledge of the level of accuracy and precision of the dosimetry systems used. This can be achieved by carrying out suitable calibration procedures of both the dosimeter system and the instrumentation used for the evaluation of the irradiated dosimeter, thereby assuring measurement traceability to appropriate national standards. The main aim of calibration is to establish a relationship between absorbed dose and dosimeter response. Calibration can be carried out

a./ in a calibration laboratory, b./ in an in-house calibration facility or c./ in a processing facility (inplant calibration).

The accuracy of a dosimeter can be significantly affected if the routine irradiation conditions of the dosimeter are different from the calibration conditions. Therefore the recent trend is to calibrate dosimeters under conditions as close as possible to those of regular application (processing conditions) by applying in-plant calibration, whenever it is possible. In this case the use of calibration phantoms is suggested to ensure similar irradiation conditions both for the standard and for the routine dosimeters [26]. Using this calibration method the use of correction factors can be avoided. In the case of calibration in a calibration laboratory correction factors have to be applied, for example due to differences between temperature of calibration and temperature of process irradiation. Calibration in a calibration laboratory should therefore be followed by verification in the irradiation plant [26].

The dosimetry calibration laboratory of the National Institute of Standards and Technology has recently introduced the internet calibration using alanine dosimetry and protected software [27].

3.1.4. Standardization in radiation processing dosimetry

International standards for radiation processing dosimetry methods and procedures are written by organizations such as ISO, EN, ASTM. The standards are important for harmonizing the dosimetry procedures thus assuring the approval of radiation processes by e.g. public health authorities. The most complete activity has been carried out by the E10.01 Subcommittee of the American Society for Testing and Materials called "Dosimetry for Radiation Processing", which has already published 31 standards covering all aspects of dosimetry on this field. In order to increase international use of these standards a decision has been made to convert ASTM standards into ISO (International Standard Organization) standards resulting in one set of standards in radiation processing dosimetry worldwide [28]. Due to this cooperation 25 ASTM standards have been converted by now to ISO standards with a new designation: ISO/ASTM 5xxxx (e.g. 51261 for "The Selection and Calibration of Dosimeters").

The proper use of dosimetry systems can be achieved through standardization (1), using the services of standard laboratories for calibration (2) and by intercomparisons (3). The International Atomic Energy Agency has set up the International Dose Assurance Service (IDAS) [29] to provide irradiation facility operators with assurance concerning their dosimetry measurements, to achieve standardization and to promote reliable dosimetry. During the past 20 years the programme has obtained wide-spread application worldwide fulfilling the original objectives.

4. CONCLUSION

Taking into account the present situation in radiation processing dosimetry we can conclude that standardized and well characterized dosimeter systems and procedures are available for process control. However, due to stricter requirements to improve quality assurance and due to the introduction of new technologies, both improved and new dosimetry systems, procedures and methods are needed for process control under standardized conditions.

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MONTE CARLO METHODS FOR PROCESS DEVELOPMENT AND CONTROL IN ELECTRON BEAM TECHNOLOGY

J. Mittendorfer^a, M. Colon^{b*}, F. Gratzl^b

^aHigh Tech Consulting, Wels, Austria

^bMediscan GmbH, Kremsmünster, Austria

Abstract. In this paper mathematical models for radiation processing applications with special emphasis on Monte Carlo methods are addressed. After a brief remark on the history of Monte Carlo radiation transport codes, typical program packages are introduced. For an in-house research project at Mediscan GmbH, ITS 3.0 was used for one-dimensional calculations and a 3D-interface was designed for GEANT 4. The design guidelines of the interface are briefly discussed together with sample applications in process development and control. Finally RPSMUG, a user group devoted to modeling and simulation in radiation processing is introduced and its mission statements presented.

1. INTRODUCTION

A mathematical model for calculating absorbed dose in radiation processing is an emerging field showing encouraging results and increased acceptance in the radiation processing community.

Mathematical models may be divided into Monte Carlo, deterministic, semi-empirical and empirical models [1]. Monte Carlo methods simulate the tracks of individual particles based on the detailed physics of the interaction of radiation in matter. In contrast to deterministic models which solve the mathematical equations of radiation transport, Monte Carlo samples the interactions as probability functions from cross section data. Energy losses of particles (mainly electrons and photons) in matter from different histories are summed to estimate the absorbed dose.

Historically, a significant impact had the ground-breaking paper of Martin J. Berger, which laid the foundation of Monte Carlo methods for radiation transport [2]. Monte Carlo methods for radiation transport are predominantly used in areas where radiation dosimetry is difficult or impossible, like radiation therapy or space radiation applications.

Today several Monte Carlo programs are available and used for industrial applications, like electron beam or X-ray irradiation. The main field of application is the calculation of dose distribution in complex geometries for process design, product validation or process qualification.

Typical examples of available Monte Carlo codes are EGS, GEANT, ITS or MCNP which are distributed by national and international institutions. ASTM E 2232 "Standard Guide for Selection and Use of Mathematical Methods for Calculating Absorbed dose in Radiation Processing Applications" give an excellent overview over existing computer codes, their availability [1].

2. MOTIVATION FOR USING MONTE CARLO METHODS

The motivation for a company dealing with industrial radiation processing is manifold. First of all Monte Carlo methods have been successfully established in science and have proven their success in mission-critical applications like radiation therapy or space flight and so similar benefits in

^{*} Now at cosine Science & Computing BV, 2333 CA Leiden, The Netherlands.

industrial applications may be expected. Besides this other facts support the use of Monte Carlo programs even more: Monte Carlo Codes is available for Personal Computers, the required hardware is affordable and the execution speed is fast enough for standard problems. In addition, guidance documents are available and with RPSMUG (Radiation Processing Simulation and Modelling User Group) a platform for the promotion of mathematical models has been formed.

3. ONE-DIMENSIONAL PROBLEMS

In a Mediscan GmbH in-house research project the applicability of Monte Carlo programs for an electron beam service centre has been evaluated. A good starting point to get acquainted with Monte Carlo code for radiation processing is the TIGER code of the ITS package [3].

One dimensional problem can be used to calculate depth-dose distributions in various materials and give insight into the relation between electron range and energy. On the basis of these Monte Carlo calculations equations determining e.g. E_p (most probable energy) from R_p (extrapolated range) can be derived, which may serve as acceptance criteria in system qualification [4].

4. THREE-DIMENSIONAL PROBLEMS

The goal of the Mediscan research project was to design a 3D-user input interface to a mainstream Monte Carlo program. Because of the open and programmer friendly interface, GEANT 4 was chosen as Monte Carlo code.

GEANT (Geometry and Tracking) is a well know and wide spread Monte Carlo code developed at CERN [5]. In 1994 a decision was made to rewrite FORTRAN based GEANT 3 in C++, an object-oriented language. GEANT 4 consists of several packages and the user is providing subroutines which define e.g. the materials and geometries involved.

As 3D-frontend the commercial program *3dsmax 4.2* from *discreet* is used. Using the built in macro language additional input windows and task bars were programmed for defining:

- Materials (elements and composites)
- detectors
- electron gun (energy and scan function)



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FIG. 1. Sample user interface of the Monte Carlo input front-end.

With the buttons *Write source files* and *Make executable the* input information is translated into C++ source code, compiled and linked to other GEANT 4 routines (Fig.2).

//--MATERIALS

G4Material* Aluminium = new G4Material("Aluminium", 2.7*g/cm3, 1, kStateSolid, 273.15*kelvin, 1.0*atmosphere); Aluminium->AddElement(elementAl, 1.0);

G4Material* Nitrogen = new G4Material("Nitrogen", 0.1293*mg/cm3, 1, kStateGas, 273.15*kelvin, 1.0*atmosphere); Nitrogen->AddElement(elementN, 1.0);

//--GEOMETRY

G4Box* World = new G4Box("World",50.0*cm,50.0*cm,50.0*cm); G4LogicalVolume* World_log = new G4LogicalVolume(World,Nitrogen,"World_log");



FIG. 2. Sample generated source code for a simple geometry.

As user interface for the compiled model GAG (Geant4 Adaptive GUI) from Naruto University, Japan [6] is used. For the graphical output of detector elements and trajectories VRML is used. Fig. 3 shows an application of this prototype 3D user interface for an electron beam vault design study.



FIG. 3. Treatment vault design study.

5. MONTE CARLO CODE FOR PROCESS DEVELOPMENT

An open program framework like GEANT 4 allows addressing specialized problems, which are difficult to attack other packages.

For a process development study, the spectrum of 500 keV electrons after travelling a certain distance in nitrogen was of interest. Because of the open source code of the critical routines the solution was straight forward and easy to implement (Fig.4).



Energy Spectrum in Air

FIG. 4. Energy spectrum of 500 keV electron after passing a certain distance of nitrogen.

6. RPSMUG

Modelling and simulation tools like Monte Carlo codes for radiation processing are powerful instruments to predict absorbed doses in order to assist and extend radiation dosimetry. Nevertheless these tools need extensive know-how to set up, provide adequate input information and interpret the output correctly.

With the Radiation Process Simulation and Modelling User Group RPSMUG [7] collaboration with the following mission has been established:

- Help members develop sound practices for using computers and dosimetry to solve processing problems
- Provide education and training for our members
- Provide software tools and training applicable to process design and simulation
- Provide group-sponsored publications (position papers, other coyrighted materials)
- Provide benchmark calculations and measurements
- Provide case studies of end-used experiences
- Provide industry management with information regarding the quality, profitability and saftety gains that come from simulation and modeling
- Help industry members communicate their needs to National Laboratories and other agencies

RPSMUG is a user group formed in a rather early stage of simulation and modelling to coordinate definitions, methods, verification and validation issues, training and other topics related to

radiation processing on an international basis. Hence there is hope that efforts are synchronized and time consuming and complex harmonization in a later stage may be avoided.

CONCLUSION

Monte Carlo methods provide important tools for process development and control and have proven their benefit for problems in industrial irradiation applications. For one-dimensional problems like depth-dose distributions the TIGER code of the ITS package is a relatively easy to use tool. For complex geometries a prototype of a 3D-front end for the GEANT 4 package has been developed at Mediscan which demonstrates the flexibility of an open program environment. RPSMUG, a newly formed users group represents a promising platform to coordinate activities in the emerging field of simulation and modelling for radiation processing.

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QUALITY ASSURANCE IN RADIATION PROCESSING. DOSIMETRY AND CONTROL METHODS FOR RADIATION PROCESSING

F. Kuntz

AERIAL, Schiltigheim, France

Abstract. In the domain of radiation processing, the process control of the treatment is vitally important. In addition to the irradiation parameters checks, the main control in this process is the dose measurement in the three major aspects of quality control as described in different standards or guides: Irradiation facility qualification, Process validation, Routine process control. In this paper, an example is given for each aspect of the quality control for E-beam, X-Rays and gamma irradiation plant as well. These examples show that the use of a traceable and validated dosimetry system is the appropriate tool to realize irreproachable controls. The choice of the dosimeter type and its calibration against traceable laboratories is also essential to prove the quality of the radiation treatments. Concerning the dosimetry readout equipment, a flexible dosimeter analysis and computerized equipment is developed by Aérial in order to bring a major contribution to the quality control in the field of radiation processing. New developments for industrial ESR dosimetry readout equipment are presented. The adapted ESR software analyses the measured spectrum, checks the measurement parameters, and calculates the dosimetry system response, absorbed dose and associated uncertainty as well.

1. INTRODUCTION

Radiation processing is based on the notion of dose deposited in a product. A dose is the quantity of energy deposited in the product, by mass unit. Its measuring unit is the Gray (Gy).

Dosimetry plays an important role in the quality control procedure of radiation processes. In industry, quality control systems are founded on several standards or guides, especially ISO 11137, EN 552, ISO/ASTM 51xxx, [1], [2], [3] This is the case for food irradiation, sterilization, pharmaceutical or polymer treatment as well as for other applications of radiation processing. In most cases, knowledge of the product absorbed dose is of the utmost importance, since methods are often defined in term of the absorbed dose. For example, the processing of foodstuffs requires an exposure radiation dose corresponding to very precise specifications. Measurement of the absorbed dose, in addition to the irradiation parameter controls, provides proof that the radiation processing is done according to the required specifications.

Using the above mentioned example about foodstuff irradiation, the three main phases of the quality control procedure are described and show the role of dosimetry.

2. THE THREE MAIN PHASES OF THE QUALITY CONTROL PROCEDURE

2.1. Irradiation facility qualification

Radiation treatment of foodstuffs must be performed in well characterized irradiation units. This means that the units are able to deliver a certain dose of radiation to a product and that the key parameters for controlling and maintaining the dose are well known and can be reproduced within the specified limits.

Dosimetry is the starting point of the characterization procedure. In fact, the measurement of the relationship between the absorbed dose and the unit's key parameters, as well as the measurement of the dose distribution in reference homogeneous products enable global characterization of the radiation unit to be attained. Figures 1, 2 and 3 show examples of dose distributions with gamma, X-rays and electron beams.



FIG. 1. X-ray (10 MeV) Dose rate mapping emitted by a Rhodotron at 7 cm from the target.



FIG. 2. Electron beam energy determination using the Aer'ODE equipment with the "wedge technique" ISO/ASTM 51649.



FIG. 3. Dose mapping in a single use surgical dress treated with 2 MeV electrons.

2.2. Process validation

When the unit is fully characterized, the next step is to analyze more specifically the product to be routinely processed.

To that effect, dosimetry must give evidence of the fact that the minimum dose required has actually been exceeded and that the maximum dose deposited causes no degradation of the foodstuff.

For this purpose, the product to be processed is submitted to a validation exercise that makes it possible to pinpoint the areas with maximum and minimum doses as well as variations of the absorbed dose in the same areas. From this data, a nominal dose value and a set of control parameters of the radiation unit can be determined so as to guarantee routine radiation processing within the specified dose limits.

2.3. Routine process control

Applied to radiation processes, the routine control consists in checking that the reference (nominal) dose, obtained through the process validation, is within the required limits. This can be done by controlling the radiation unit's key parameters (i.e. radiation time, conveyor speed for a 60 Co unit, power, electron energy, scanning length, conveyor speed for an e-beam radiation plant) and dosimetry makes it possible to carry out the conformity test of the radiation process.

3. SELECTION, CALIBRATION OF A DOSIMETRY SYSTEM AND ASSOCIATED UNCERTAINTY CALCULATION

In the three phases of the control quality procedure, measurement of the dose requires the following steps to be undertaken:

- Selection of a dosimetry system
- Calibration of a dosimetry system
- Uncertainty calculation

3.1. Selection of a dosimetry system

A dosimetry system comprises the dosimeter, its response function, directions for use and the connected measurement instrument. Dosimeter systems can be divided into two categories: physical and chemical dosimetry methods [4].

Calorimetry (a physical method) is a primary dosimetry method that enables direct determination of the dose (D = E/m) by measuring the quantity of heat deposited in a heat isolated absorbent. Consequently, this method requires no calibration on another radiation measurement tool. However, if the concept of the calorimeter is relatively simple, it must be noted that its development and implementation are quite complex. This is the reason why calorimeters are marketed and used for such specific applications as the calibration of secondary dosimeters for electron beams, or for more routine applications of the e-beam radiation processing.

Secondary dosimeters (those that require calibration) mainly belong to the chemical method group and are usually more simple to put into operation.

Such is the case of reference dosimetry systems (characterized by great precision, stability and reproducibility). They are supplied and analyzed by reference laboratories and used for the calibration of radiation fields. Examples of reference dosimeter systems are the Fricke dosimeters, the Dichromate (liquid dosimeter systems) or the alanine (solid system) system.

Routine dosimetry systems are mainly optical absorbent solid systems (radiochromic, CTA, PMMA) or luminiscent solid systems (Sunna-LiF). Of course, the selection of such a routine tool for dose measurement depends on the dose range of the dosimeter, on the stability and reproducibility of the measure [5], but also on its ease of use and above all on its utilization cost (cost of the dosimeter and of the analysis system).

3.2. Calibration of a dosimetry system

In the routine applications of radiation processing, absorbed doses are measured with secondary dosimeters (FWT 60.00, PMMA, CTA, Gex, PVC ...) that, de facto, have to be regularly and frequently calibrated on reference systems. Laboratories that perform calibrations are, among the best known : US National Institute of Standards and Technology (NIST), UK National Physical Laboratory (NPL) and the French Laboratory known as the Laboratorie National Henri Becquerel – (LNHB). Their radiation sources (gamma and electron), which are calibrated by reference dosimeters, and the transfer dosimetry systems (Alanine, Dichromate, FWT 60.00) they use, enable calibration of routine dosimeters by following three different procedures (as shown in the figure 4 [4], [5]) :



FIG. 4. Calibration of routine dosimeters.

In the first procedure, the transfer dosimeters that are calibrated in the reference laboratory, are sent to the industrial radiation plant and exposed to radiation together with the routine dosimeters which are to be calibrated, in the plant calibration source.

The second procedure implies applying dosimetry to foodstuffs processed in the industrial unit using the transfer dosimeters that are supplied and analyzed by the reference laboratory.

In the third procedure, routine dosimeters are sent to the reference laboratory (if they are stable enough), where they are exposed to radiation at known doses. This procedure allows the operator of the radiation unit himself to calibrate the dosimeters he will use on a routine basis.

As a general rule, regardless of the selected calibration method, the dosimeter laboratory of an irradiation plant must operate in conformity with a national or international dosimetry reference. In this way, the operator lends more credit to the routine dosimeter measurements and, consequently, to the radiation processes performed.

3.3. Absorbed dose uncertainty calculation

The determination of the absorbed dose is carried out starting from measurements of several parameters, of calibration data and corrective factors related to the conditions of irradiation and readout. The absorbed dose by a dosimeter i, is then given starting from the various parameters (measured, calculated or established beforehand) bound by a relation of the type: $D_i = f(X_1, X_2, ..., X_n)$. Each one of these parameters is estimated by a value xi with which is associated a u(xi) uncertainty. The results of measurement Di are thus a random variable, with which a composed uncertainty $u_c(Di)$ is associated. The composed uncertainty $u_c(Di)$ is obtained by applying to the u(xi) uncertainty the law of propagation and this, which they are of type A or type B. If the u(xi) are independent :

$$\boldsymbol{\mathcal{U}}_{c}(Di) = \sqrt{\sum_{i} \left(\frac{\partial f}{\partial x_{i}}\right)^{2} \times u^{2}(x_{i})}$$

4. A FLEXIBLE AND INDUSTRIAL DOSIMETRY READOUT EQUIPMENT

A flexible dosimeter analysis instrument – Aer'ODE - has been developed by Aérial [8] in order to bring a major contribution to the promotion of quality control in the field of radiation processing. This instrument is an automatic dose measurement and control system (see figure 5). It is PC controlled and makes it possible to obtain the dose by optical absorption dosimeters (Harwell, FWT, Gex, CTA, GAF Chromic, PVC...) in a few seconds.



FIG. 5. Aer'ODE, a flexible and industrial equipment for optical dosimetry readouts.
The Aer'ODE Software:

- controls totally the equipments (settings of equipments, readings, storage of data in Database)

- performs dosimetry system calibration, single or strip measurements with uncertainty calculation, graphics, profiles, process validations (loading pattern, plans definition and report edition)

- performs equipment verifications (Absorbance, Wavelength, thickness) needed to certify the absorbed dose measurements

-Special developments and software customising can also be performed according to specific industrial demands

The global Aer'ODE equipment is validated and meets the quality assurance requirements according to FDA part 11, ISO, EN, ASTM, guidance or standards

Alanine/ESR method as a routine and industrial dosimetry system for radiation processing controls is under development at Aérial as well. The equipment links an ESR spectrometer to an adapted dosimetry software (based on the Aer'ODE software features) in order to propose to the radiation processing community a global package for high quality routine dosimetry. This industrial tool analyses the measured ESR spectrum, checks the measurement parameters as well as the operator's authorizations and calculates the dosimetry system response, absorbed dose and associated uncertainty. By using this new equipment on a routine bases, the irradiation facility operator will diminish the uncertainty on the absorbed dose calculation of about 3 % and even more.

5. CONCLUSION

The main components of a quality assurance plan in an industrial irradiation unit have been detailed above. Characterization of the radiation field of the facility, validation of process and routine process control by means of traceable dosimeters to national or international standards are the three most important contributions of dosimetry to the quality control of irradiation processes. Training of laboratory staff and compilation of a "dosimetry laboratory operations guide" also enable the operator of an irradiation facility to guarantee the quality of its dosimeter measurements and, consequently, of the performed radiation treatments.

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RADIATION SAFETY AT GAMMA AND ELECTRON IRRADIATION PLANTS

A. Kovács

Institute of Isotope and Surface Chemistry, Chemical Research Center, Hungarian Academy of Sciences, Budapest, Hungary

Abstract: Radiation safety of gamma- and electron irradiation facilities is of basic significance concerning the safety of personnel involved in operation, which of the products treated as well as the environment of the facility. The safety measures with respect to the design and construction of the facilities, requirements concerning the basic parts and the operation of the plants, the reliable control of running irradiation facilities and the role of emergency planning and the role of licences are discussed based on the Safety Guides of the International Atomic Energy Agency.

1. INTRODUCTION

Radiation processing has become a worldwide accepted and established industry during the past few decades both in developed and in developing countries. The most widespread applications involve radiation sterilization of medical products and pharmaceuticals, food irradiation, polymer modification, environmental and biomedical applications. These technologies utilize either gamma- or electron irradiation resulting nowadays more than 200 high activity gamma irradiation facilities and about 700 electron accelerators in operation worldwide.

Due to the nature of ionizing radiation used in radiation processing safety considerations have got basic importance with respect to licensing, design, construction, operation and maintenance of irradiation facilities as well as to transport, loading and unloading of radiation sources. These safety requirements involve precautions to protect the personnel of irradiation facilities as well as customers and inhabitants living in the neighbourhood of the facilities, since severe radiation exposure may result from loss of control of the radiation source. Safety measures are needed to protect the products to be treated and to ensure safe operation by protecting the basic parts of the irradiation facilities including control and auxiliary instrumentation and equipments. It is of basic importance since e.g. damage to the radiation source can lead to dangerous contamination.

The responsibilities of the operating organization as well as all relevant parties taking part in the design, construction, import and installation to establish an irradiation facility have to be clearly identified and documented [1]. Special programmes - like a regulatory programme enforced by the local competent authority and an emergency response planning - must be prepared and documented.

This paper in general follows the structure of the Safety Series No. 107 on "Radiation Safety of Gamma and Electron Irradiation Facilities" edited by the International Atomic Energy Agency [2].

2. IRRADIATION FACILITIES

2.1. Gamma irradiation facilities

The activity of the radioactive source in an irradiator ranges from a few terabecquerels (1 TBq = 27 Ci) to more than 100 PBq (3 MCi). With respect to the design of irradiation facilities four general categories are defined as follows:

Category 1: An irradiator in which the sealed source is completely enclosed in a dry storage container constructed of solid materials and shielded at all times, and where human access to the sealed source and the volumes undergoing irradiation is not physically possible in the designed configuration. Such irradiation facilities are usually used e.g. in research laboratories, where small volumes of products are to be treated.

Category 2.: A controlled human access irradiator in which the sealed source is enclosed in a dry container constructed of solid materials, and the sealed source is fully shielded when not in use and it is exposed within a radiation volume that is maintained inaccessible during use by an entry control system. Such an irradiation facility is usually located within a concrete biological shield providing the necessary radiation protection during operation.

Category 3.: An irradiator in which the sealed source is contained in a water filled storage pool and is shielded at all times, and where human access to the sealed source and the volume undergoing irradiation is physically restricted in the designed configuration and proper mode of use. Such design is usually employed in a pool irradiator in which the product to be irradiated is lowered in a water tight container to the bottom of the pool among or next to the source.

Category 4.: A controlled human access irradiator in which the sealed source is contained in a water filled storage pool, the sealed source is fully shielded when not in use and the sealed source is exposed within a radiation volume that is maintained inaccessible during use by an entry control system. The irradiation volume is typically contained within a concrete biological shield providing the radiation protection during operation or exposure of the radiation source. This type of irradiation facility can be either batch or continuous type and the latter is usually equipped with a product transport system.

Beside these general categories the irradiation facilities can also be categorized according to the storage type of the radiation source (dry or wet), to the radiation volume (closed or panoramic) or to the product transport system (tote, carrier, pallet, bulk or no transport system).

2.2. Electron beam facilities

With respect to induced radioactivity electron accelerators of energies equal or less than 10 MeV are considered in the present discussion. The main differences concerning the different types of electron accelerators are due to the mode of accelerating the electron beam and to the method of producing the necessary high voltages. For the present purpose the electron irradiation facilities are categorized as follows:

Category 1: An integrally shielded electron beam unit with interlocks, where human access during operation is not possible due to the configuration of the shielding. These facilities operate usually with electron energies lower than 0.5 MeV, although there are a few examples of such facilities operating with much higher energies due to recent designs.

Category 2: An electron beam unit located in shielded rooms that are maintained inaccessible during operation by an entry control system. These facilities operate usually with electron energies higher than 0.5 MeV.

3. SAFETY DESIGN PRINCIPLES OF IRRADIATION FACILITIES

3.1. Radiation safety objectives

In the design, installation, commissioning, operation, maintenance and decommissioning of an irradiation facility the following radiation safety objectives have to be taken into account:

a. During normal operation, maintenance and decommissioning as well as in emergency situations radiation protection has to be optimized ensuring that the radiation exposure of both workers and the public is kept as low as reasonably achievable, economic and social factors are taken into consideration (ALARA principle).

b. It is necessary to ensure that during normal operation, maintenance and decommissioning as well as in emergency situations the radiation exposure of both workers and the public is kept below the relevant dose limits as given in the Basic Safety Standards for Radiation Protection [3].

c. It has to be ensured that the probability of events giving rise to significant exposures as well as the magnitude of such exposures are kept as low as reasonably achievable, economic and social factors are taken into account.

3.2. Safety philosophy

Since the design of an irradiation facility depends on its planned operation as well as on its category [4, 5] it is not possible to suggest specific design of the required degree of safety, but to present several design principles to be used - if necessary in combination - to achieve and maintain the required reliability. These basic design principles are the following:

3.2.1. Defence in depth concept

This concept should be applied to all (organizational, design related, behavioural) safety activities to ensure that they are covered by overlapping provisions so that if a failure should occur, it would be compensated for or corrected.

The design procedure shall incorporate this concept so that multiple levels of protection have to be provided and the necessity of human intervention has to be minimized. In the course of the design process e.g. a series of levels of defence (in terms of equipment and procedures) is provided to prevent accidents or to mitigate their consequences in the event that preventive measures fail. The first level of defence is to prevent deviation from the normal operation to be achieved by soundly and cautiously designed, constructed and operated facility, by establishing and maintaining suitable quality assurance programme. The second level of defence is to detect and respond to deviations from normal operating conditions to prevent anticipated operational occurrences from escalating into accident conditions. The aim of the third level of defence is to mitigate the consequences of an accident and through achieving stable and acceptable conditions.

Irradiation facilities should only be operated if all levels of defence are in place and functioning.

3.2.2. Redundancy

It is the use of more than the minimum number of items needed to accomplish a given safety function. It enables the failure or unavailability of one item to be tolerated without loss of the function. A redundant component can also be considered as such an item, which is not really needed, but built in case of failure of another component of similar purpose.

3.2.3. Diversity

The reliability of some systems can be enhanced by using the principle diversity. It is applied to redundant systems or components that perform the same safety functions by incorporating different attributes (like different principles of operation, different operating conditions) into the systems or components.

3.2.4. Independence

Independence is achieved in the design of systems by applying functional isolation and physical separation. The reliability of redundant systems can be improved by using independent components or systems thus avoiding the failure or loss of other components, safety function or equipment designed to mitigate the effect of incidents in the case of the failure of one component or system. The use of this concept improves the reliability of systems also by maintaining independence of systems or components of different importance to safety.

3.2.5. Programmable electronic systems

Programmable electronic systems [6] are in use in safety control applications with potential problems related to the integrity of the hardware and validation of the software leading to faults in the system. Any alterations to the software can be made only in case of having authorization from a competent authority.

3.2.6. Safety analysis

A formal method of safety assessment – e.g. a hazard analysis technique like a probabilistic safety analysis – should be used. Each component within the system should be considered in turn. The potential types of failure and their consequences for the system as a whole should be taken into account considering the reliability of the safety dependent operating procedures and should encompass both inadvertent and deliberate failure to follow procedures. The operating organization has to demonstrate to the competent authority how the irradiator design and the related operational procedures will contribute to the prevention and mitigation of accidents. This information has to be provided in the form of a documented safety analysis as part of the licensing document submitted to the regular authority.

4. SAFETY DESIGN REQUIREMENTS

With respect to the design of irradiation facilities guidance is needed for the designers in terms of dose or dose rate objectives since these parameters are in relation with the planned use of the facility. The permissible radiation levels outside the biological shield to which employees, customers or the public may be exposed during operation of the facility has to be based on the ALARA principle taking into account of any additional dose constraints that may have been specified for the purpose by the local competent authority. Experiences with respect to the operation and maintenance of similar facilities should be collected and used in specifying the levels of exposure that are achievable in practice. Such experience originating from many countries has shown that irradiation facilities can be designed and operated in such a way, that radiation workers are exposed to radiation levels significantly less than 5 move per year. The basic design parameters and related safety features concerning the main parts of irradiation facilities (radiation source, biological shield, product transfer system, control and safety systems, and auxiliary systems) are discussed briefly below.

4.1. Radiation source

4.1.1. Design of sealed sources

The radiation source in case of gamma irradiation facilities in most cases is ⁶⁰Co isotope, while in a few cases it is ¹³⁷Cs isotope. These sources are double encapsulated sealed sources and the general requirements for such sources are given in ISO Standard 2919 [7, 8]. In addition to this standard the manufacturer and the user have to take into account the possible effects of fire, explosion, corrosion, and other aspects related to the continuous use of sealed sources (e.g. leak tests). Specific requirements are needed for radiation sources kept in water (wet storage conditions). Both the source manufacturer and the user have to maintain suitable certification and documentation of the sealed sources for their own purpose (record keeping) as well as for the competent authorities for licensing.

4.1.2. Internal design

The design of the source holder and rack has to ensure the fixed positioning of the sources to avoid their dislodging. The source must be provided with suitable mechanical protection to avoid damage or interference by the product boxes or product carriers. The product positioning system shall be provided with controls to detect any malfunction of the system resulting in the source to automatically become fully shielded.

4.2. Biological shielding

Direct radiation exposure from the operation of irradiation facilities shall be limited by appropriate shielding using materials of well established characteristics [5, 9]. Biological shield provides protection outside of the irradiation room (radiation volume) during operation and provides safe storage for the radiation sources. The amount of shielding should be determined by reference to any dose rate requirements specified by the competent authority. Shielding calculations for design purposes shall be carried out by specialists. In the case of wet storage irradiation facilities special care should be taken to pool integrity.

4.3. Control, safety and auxiliary systems

These systems, devices and instruments are designed and used to control the safe operation of the irradiation facility (e.g. controlling the safe operation of the speed of the product transport system, position of product boxes, movement and position of the source hoist), as well as to control the safety systems (like dose level, water level and condition in the pool, ventilation, power supplies, etc.). These safety features are discussed according to the categorization of the IAEA Safety Series No. 107 [2].

4.3.1. Access to the radiation source and interlocking system

Special attention – and suitable interlocking system - is needed to avoid personnel access into the irradiation room while the radiation source is in exposed position or the source hoist mechanism is energised. Interlocking systems and monitors are required at the personnel access door, at the product entry/exit ports as well as at the removable radiation room shield plugs.

Fixed radiation monitor (with visible and audible alarm signals) shall be provided to detect the radiation level in the radiation room. Interlocks shall be provided to control the source position and exposure system. In case of any malfunction access into the radiation room shall be prevented.

At wet storage facilities fixed monitoring systems (with audible alarm) are provided to control the columns of the water treatment system and to detect contamination originating from source leakage. The monitor shall be interlocked with the irradiation controls to ensure that the source returns to its shielded position, the water circulation is stopped and warning signal is given.

In case of fully shielded facilities (categories I and III for gamma and I for electron beam units) the irradiator shall not be operable until all shielding is in place and all other safety devices are actuated.

4.3.2. Control console

Each irradiator shall have a master control (key operated switch, padlock, etc.) for use to prevent unauthorized operation. In addition means shall be provided to terminate an irradiation and return the irradiator to its safe ("source not in use") status at any time, and an emergency stop device should be available at the control console.

4.3.3. Radiation room

The radiation room shall be equipped with a safety delay timer to automatically generate visible and audible signals to alert persons that the source exposure has begun. To protect persons inadvertently shut inside the radiation room an emergency exit or a low dose location should be provided. An emergency stop device should also be located inside the irradiation room to terminate radiation or to prevent the source from moving from storage to exposed position.

4.3.4. Wet storage irradiators

In these irradiation facilities the integrity of the pool and the use of corrosion resistant pool components have to be ensured. Water level control with visible and audible signals has to be provided for such cases when the water level falls to more than 30 cm below the normal level. Furthermore automatic water replenishment, water cooling, water conditioning system with conductivity control, external high flow rate water supply have to be provided. To protect personnel a physical barrier has to be built along the pool.

4.3.5. Geological site considerations

Geological features that could adversely affect the integrity of the radiation shields have to be evaluated considering the physical properties of materials under the irradiator site and its environs. The irradiator has to be equipped with seismic detector in the case of possible severe seismic activity.

4.3.6. Fire protection

Heat build-up originating from anomalous operation of the facility can lead to combustion, which makes necessary the use of heat and smoke sensing devices with visible and audible alarms. The triggering of these devices shall cause the source automatically to become fully shielded and the product positioning and the ventilation system to shut down. A fire extinguishing system – not containing chemicals or corrosive materials – should be provided in the irradiation room.

4.3.7. Power failure

Means shall be provided that in case of both electrical and non-electrical (e.g. pneumatic or hydraulic) power failure the source shall automatically be returned to the fully shielded position and the irradiator shut down. The safety control system has to be designed so, that it will remain in operation in the case of power failure.

4.3.8. Ventilation

Ventilation system – which creates a negative pressure in the irradiation room - shall be used to protect personnel against exposure to concentrations of harmful gases (ozone, nitrous gases and other noxious gases) - above the prescribed threshold limit values – produced upon irradiation. This type of forced air system – with continuous air flow monitoring - should be interlocked so, that failure of the system will automatically shut the plant down together with the source moving to the fully shielded position. A time delay interlock mechanism should be introduced to prevent personnel from entering the irradiation room immediately after irradiation.

4.3.9. Warning signs and symbols

Clearly visible signs bearing the radiation symbol and warnings must be located at the personnel access door leading into the irradiation room. Clearly visible irradiation status indicators shall be provided at the control console to indicate the status of the source and these shall be visible at each personnel or product entry/exit location. Audible signals designed into the irradiator control system shall be distinct and loud enough to gain immediate attention of persons in the area.

Category I gamma irradiation facilities shall have clearly visible labels showing the contained radionuclides, their activities and the related dates.

5. SPECIAL SAFETY REQUIREMENTS FOR ELECTRON ACCELERATOR FACILITIES

5.1. Safety design considerations

An objective of manufacturers of industrial accelerators is to design EB sources for simplicity and reliability of operation. With respect to potential radiation accidents care should be taken due to the possibility of generating X-rays from dark currents, if the acceleration capabilities of the facility or its subsystems remain because of improper or partially disabled accelerating stages. To reduce the chance of hazardous occurrences the following features should be considered in the course of design:

- a. / Positive means of disabling the main acceleration system;
- b. / Built-in machine parameter monitoring;
- c. / Built-in remote machine diagnostics;

5.2. Shielding

Due to the differences in the nature of electron radiation and X-ray radiation generated only the latter should be considered when carrying out shielding calculations. These calculations must be performed for the maximum energy and current that the accelerator can produce. To minimize the generation of X-ray radiation low atomic number materials should be used as far as possible. Except for self-shielded accelerators and special purpose machines operating at higher energies ordinary concrete is preferred as shielding material. To calculate the shielding thickness different mathematical methods and special programmes are available [5, 10].

5.3. Other requirements

The operating parameters of accelerators (voltage and current) should be interlocked with the product transport system. Testing and commissioning of the accelerators should be carried out at maximum operating parameters and with product transport system under the beam as close as possible to actual operating conditions.

6. REGULATORY CONTROL

6.1. Regulatory programme

In the use of ionizing radiation regulatory programme – enforced by the local authority - is an important part of radiation protection [11]. This programme is intended to control irradiation facilities by systems of notification, registration or licensing, dependent on the legislation in any given country. The most common system of regulatory control is that of licensing or approval as described briefly here.

It is important to note that the main responsibility for safety belongs to the person(s) carrying out the given tasks (design, installation, operation, maintenance, decommissioning).

It is also essential that a regulatory system must be in place in any country before approval of an application to build an irradiation facility.

6.2. The approval process

The control of radiation safety concerning the sitting, design, construction, commissioning, operation, maintenance and decommissioning of an irradiation facility is carried out by governmental licences or approvals, which authorize actions and place conditions on the applicant. The local competent authority has to review and assess the application to decide whether or not the approval is granted.

6.2.1. Functions of approvals

This official document authorizes a certain activity in connection with all procedures relevant to the establishment and use of an irradiation facility and establishes requirements how to perform these activities.

6.2.2. Stages of approval process

The main stages of this process have to include the regulation of sitting, design, construction, commissioning, operation, maintenance and decommissioning. Before approval is issued a radiation safety analysis (case) – as part of the overall licensing document - must be prepared by the applicant including the complete description of the facility with details of all important factors affecting its operation and safety

6.2.3. Requirements for the applicant

The applicant has the primary responsibility to ensure safety for all stages of establishing and operating an irradiation facility. The applicant has to demonstrate to the competent authority that workers, customers and public will be adequately protected. The manufacturer of the facility will also provide all necessary information (e.g. training of workers to operate the facility, removal of spent radioactive sources, construction and installation corresponds to the specifications, etc.) for the applicant. Other requirements involve the appointment of radiation protection personnel, assessment of hazards, periodic testing and surveys of the radiation protection and safety aspects of the facility, regular reports on senior staff changes, radiological data like personal dosimetry, contamination monitoring.

6.2.4. Responsibilities of the competent authority

Based on the applicant's technical submissions the competent authority has to determine whether the design and the operating procedures comply with the actual safety objectives and requirements and whether the facility can be established and operated.

6.2.5. Review and assessment of the application

The basis for the review and assessment of the radiation safety implications of the proposed facility are those documents, safety reports, etc., which have been submitted by the applicant.

6.2.6. Programme of review and assessment

The competent authority has to prepare a programme of review and assessment which comply with the stages of the approval process. It has to follow as closely and continuously as possible all stages of the establishment and operation of the facility, the commissioning programme proposed by the applicant and all necessary changes must also be reviewed before permitting their implementation.

6.2.7. Approval decisions

The review and assessment process will result in a number of regulatory decisions. At certain stages of the approval process the competent authority will take official actions resulting in the granting or the refusal of an approval.

6.2.8. Review of approvals

Granted approvals can be re-examined by the competent authority in case of new information, which can affect safety considerations.

6.3. Regulatory inspection and enforcement

6.3.1. Objectives

The principal objectives are to ensure that:

- a. the persons responsible for the establishment and operation of the facility are competent;
- b. the required quality and performance of components, structures and systems are achieved;
- c. any deficiencies in equipment and procedures are corrected;
- d. the competent authority will be informed about experiences concerning operation, decommissioning;

6.3.2. Responsibilities

The competent authority is responsible for regulatory inspections according to a specified programme to assure that all safety related activities, systems as well as personnel competence satisfy the requirements according to approval.

Special regulatory inspections shall be carried out in case of abnormal occurrence requiring immediate investigation.

The competent authority should send a written warning or directive to the responsible organization in case of minor deviations from or violations of approval regulations specifying its nature and the necessary corrective actions including its timing.

In case of severe violations an immediate action will be taken by the authority requiring the operating organization to curtail activities.

In the event of very serious or chronic non-compliance with approval conditions or regulations the authority shall suspend or revoke the approval.

6.3.3. Inspection functions

Regulatory inspections shall be performed in all areas of the regulatory responsibility with respect to sitting, construction, operation, quality assurance programmes, reviewing results of periodic tests, preparing reports on regulatory inspections, checking emergency plans, etc.

6.3.4. Enforcement

The competent authority must have suitable power to enforce compliance with the relevant regulations and approvals.

6.4. Existing facilities

Facilities built before the adoption of these recommendations should also be subject to regulatory control (and regular inspection) and the operating organization has to demonstrate to the competent authority that the facility achieves an acceptable safety standard.

7. RESPONSIBILITIES OF THE OPERATING ORGANIZATION

The operating organization responsible for the possession and use of the irradiator shall obtain from the competent authority approval, permit or authorization necessary for the acquisition, storage and use of the irradiator. The operating organization shall be responsible for the operation of the irradiator according to the given approval, permit or authorization.

The operating organization has the following responsibilities:

- To appoint radiation protection advisers in case of not having the necessary in-house radiation-technological expertise.
- To appoint at least two radiation protection officers with duties of ensuring the implementation of written administrative procedures and of assisting the organization to comply with the requirements of the approval.
- To assure that qualified operators ensure the safe operation of the facility having appropriate certificate of competence and approved training.
- To ensure regular staff training.
- To ensure personal monitoring (even for visitors), the availability of portable monitors and area monitoring with suitable instrumentation.
- To ensure regular testing and maintenance of safety instrumentation, interlock systems, control equipments. A programme shall be established and performed including weekly, monthly and semi-annual tests.
- All tests, maintenance tasks, modification or changes to the irradiator shall be recorded.
- Regular maintenance for the whole facility shall be done according to the manufacturer's instructions. Modifications can be carried out only after approval from the competent authority and by qualified persons.
- To ensure safe operation clearly defined operational procedures laid down by the manufacturer and approved by the competent authority have to be followed.
- To ensure that all product positioning system components, product boxes or carriers continuously meet the design specifications.

8. RESPONSIBILITIES OF OTHER RELEVANT PARTIES

- 1. The designers and manufacturers of the irradiation facilities shall ensure that the facilities meet the radiation safety objectives and any specific safety requirements of the competent authority.
- 2. The importers and suppliers shall ensure that the facility is of safe design and that information on safe operating procedures is passed on to the operating organization.
- 3. Constructors and suppliers shall ensure that their work does not compromise the safety aspects of the facility and they fully comply with the requirements of the designer and the manufacturer.

9. THE TRANSPORT, LOADING AND UNLOADING OF SOURCES

9.1. Transport

All transport of radioactive sources shall comply with the requirements of the IAEA Regulations for the Safe Transport of Radioactive Material and any existing national legislation [12].

A competent authority shall be appointed to set up and execute a programme for monitoring the design, manufacturing, testing, inspection, maintenance of packaging as well as to implement a system of documentation for the handling and storage of packages containing radioactive material by consignors and carriers.

In the case of transport accidents available recommendations should be used as guidance [13].

Due to recent events stricter regulations have been introduced with respect to the safe surface transport of radioactive sources. These regulations include stricter and more frequent reporting before, during and after the transport and the release of limited information about the shipment.

9.2. Loading and unloading of sources

Due to the potential hazard these operations shall be undertaken under close radiation protection supervision.

These operations may result in higher exposure to persons as compared to the conditions in normal operation of the facility, thus an evaluation is needed beforehand to keep the potential exposure as low as reasonably achievable.

Recent regulations allow the presence only of the skilled and designated staff at such operations.

10. EMERGENCY RESPONSE PLANNING

The operating organization is responsible to prepare an adequate written emergency procedure (for each type of emergency foreseen) after having carried out a formal assessment of hazards [13]. In the case of an accident the operating organization shall initiate the emergency procedures and shall inform the competent authority and the radiation protection advisers.

Any incident shall be reported to the competent authority with a time schedule described in the approval and depending on the severity of the incident. Conclusions should be drawn to improve safety and to learn lessons with special attention to previous radiation accidents taken place earlier.

Extreme care should be exercised in the events of special problems with gamma facilities like the removal of damaged or leaking source, removal of contaminated material and actions under increased radiation levels.

11. CONCLUSIONS

Taking into account the regulations described in the relevant national and international safety standards and recommendations we can conclude that no severe changes have been made with respect to the safety considerations of the gamma and electron beam irradiation facilities. Recent regulations concentrate on increasing the security at major nuclear facilities by introducing closer personal control and by limiting the access of individuals. Stricter regulations have been introduced, however, concerning the transport, as well as the loading and unloading of radioactive material.

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RADIATION PROCESSING OF POLYMERS

A. Zyball

BGS Beta-Gamma-Service GmbH & Co. K, Wiehl, Bruchsal, Saal a.d. Donau, Germany

Abstract: The irradiation of plastics is used in the manufacturing of shrinkable products. In the cable industry, cable insulation made of cross-linked polyethylene, polyvinyl chloride, and polyurethane is used. Cross-linking allows the addition of inorganic fillers to plastics, acting as flame-retardants, while improving the mechanical properties at the same time. This is the only way to realize halogen-free, flame retardant insulations. A cross-linking by means of irradiation improves the stress cracking resistance of PE. This property is required for tubes that are used for radiant heating systems as well as water and gas supply. New irradiation facilities enable the processing of longer materials, such as large diameter tubes with a length of up to 12 m. Such tubes can be used for long-distance gas supply. Due to the cross-linking, these tubes can be installed without a sand bed, or even using a trench less installation method. Glass fiber reinforced polyamide that is cross-linked by irradiation is successfully used in the electrical and automobile industries. The ability to cross link thermoplastic elastomers (TPE) by means of irradiation allows the manufacturing of complex seals that are also able to resist high temperatures.

1. INTRODUCTION

The irradiation of polymers has been performed since the 1950s. It was discovered that plastics could be cross-linked by means of irradiation. In addition, plastics can be degraded as a result of irradiation, and it is possible to graft low molecular weight substances onto a polymer chain.

Extensive research work in the area of irradiation has been performed in the past 50 years. Some results from this work have been implemented in industrial applications. A lot of results, however, have not yet achieved industrial maturity. It can also be assumed that a lot of possible applications that can be realized by means of irradiation have not yet been developed.

2. IRRADIATION SYSTEMS

The development of irradiation systems that can be used industrially, i.e. economically, has made substantial progress in the past 20 years.

A Van-de-Graff Accelerator dating back to the 60s of the past century is installed as a museum piece on the premises of BGS Beta-Gamma-Service. This system provided energy of 1 Me and a power of 250 Watt. The radiation room in which this electron accelerator was originally installed had dimensions of 5 m x 4 m.

Alongside this system, an electron accelerator with a maximum energy of 10 Me and a maximum power of 200 kW is running today. The dimensions of the irradiation room are $55 \text{ m} \times 45 \text{ m}$. 11,500 cubic meters of concrete were processed for the construction of the irradiation room.

During the last 50 years, a lot of experience regarding the operation of irradiation systems was gained. Thanks to this experience, today's irradiation systems can be installed and operated while meeting the highest safety requirements.

3. ACCEPTANCE

These findings have not only contributed to the safe operation of irradiation facilities, but also to the confidence of the population in this technology and in the products that are manufactured using irradiation technology.

Certainly, this aspect has not yet been finally concluded. Even today, operators of irradiation systems are asked again and again which safety measurements the client or the consumer must meet with respect to the handling of the irradiated products.

And still, no distinction is made between an irradiated product and a product emitting rays. This continues to hamper the breakthrough of irradiation technology in industry.

The irradiation sterilization of medical products already finds full acceptance, whereas irradiation cross-linking is partly looked upon with skepticism, and the irradiation of food finds practically no acceptance at all.

4. IRRADIATION ENERGY

As already mentioned, irradiation technology is accepted in the area of sterilization. This does, however, not apply to the other possible industrial fields of application. A lot of instructional work has to be done in the next years.

It is the task of the system manufacturers and the operators of irradiation systems to perform such instructional work. The advantages, but also the limitations, i.e. the risks of irradiation systems, must be clearly presented. Safety has utmost priority.

4.1. What are beams? Beams are energy carriers!

If the correct energy of the beam is chosen, the irradiated products will not be activated as a result of the irradiation. Chemical reactions are triggered by the energy that is absorbed in the irradiated product.

Thus, beams can be compared to heat, which also is an energy carrier. Which technical processes can do without heat, without energy?

This perspective clearly shows the potential of irradiation technology. In many cases, the energy required for the chemical process can be put into a product more specifically, effectively and, thus, also more economically, by means of irradiation than by means of heat.

5. INDUSTRIAL APPLICATIONS

The following sections mainly focus on the irradiation cross-linking of plastics. Other applications in the plastics industry are only touched upon.

In industry, the degradation of plastics by means of irradiation is used for the depolymerization of polytetrafluoroethylene (PTFE). Polypropylene (PP) is also degraded by means of irradiation, and irradiated PP is used as a nucleating agent for PP in the field of injection molding. Cellulose is degraded by means of irradiation to avoid chemical processes having a high environmental impact.

The grafting of plastics with low molecular weight substances by means of irradiation is scientifically known. The industrial applications still have to be developed. This holds a large potential for the future that can be industrially utilized.

5.1. Shrinkable Products

Shrinkable products are the first cross-linked products that were used in industrial applications (see Figure 1). Polyethylene (PE) is cross-linked by means of irradiation. If the crystallite melting temperature, i.e. 140°C, is exceeded the irradiated PE is not a melted mass, but an elastomer. The part

that has been irradiation cross-linked (tube, section, or an injection molded part) can be stretched up to the elongation at break point of cross-linked PE. Depending on the PE used, the elongation at break can be up to 400%. In the further stages of the manufacturing process, the product that has been stretched under heat is cooled down, whereby the elongation is maintained. The crystallites in the PE redevelop, and a "frozen" rubber is thus formed at room temperature. If the tension with which the product was stretched is released, the product does not collapse. The crystallites maintain the shape. If the product is, however, heated and the crystallites melt again, the product "remembers" its rubber-like state and can shrink back to its original dimensions.

This memory effect was initially used in electrical technology, subsequently in the automobile industry, and today in many other technical sectors.

Materials with different properties have been developed for various fields of application. Using a shrinkable product, a part with differing dimensions can be encased in an optically perfect manner. The results are much better than painting or taping. Connections, e.g. from cables to cable lugs, can also be stabilized. There is a broad range of applications.

Application: Shrinkable Products



FIG. 1. Shrinkable products (Memory-Effect).

5.2. Cables

The irradiation cross-linking of plastic cable insulation is a further field of application. The initial requirement was to transfer more power with a cable. This means that the temperature of the conductor increases. The cable insulation must not melt in this case, and the insulation must not shift from the conductor.

Cable insulation made of polyethylene (PE), polyvinylchloride (PVC), polyurethane (PUR), and other plastics can be cross-linked. Cables that are insulated with cross-linked PE can be used at temperatures of more than 200° C. As is well known, the melting temperature of PE is 140° C. Therefore, a short-term operation at 60° C above the melting temperature is possible.

Irradiation cross-linked PVC is used as power cords for irons. It is possible that the iron comes into contact with the cable insulation at a temperature of 250° C, and the insulation does not melt off. In general, such insulations are used for heat-generating appliances (toasters, ovens, hair dryers), but also in the automobile industry as cable insulation for the welding robots in the manufacture of the car body, and in the car itself in the engine compartment.

In the course of the last years, further requirements for cable insulation have emerged. Insulation must be flame retardant, but halogen-free at the same time. Plastic compounds having a high amount of inorganic fillers must be used. Thus, the mechanical properties are reduced. An irradiation cross-linking can compensate for the loss of these mechanical properties.

The main property improvements of a cross-linked cable insulation:

- improvement of the thermal properties
- improvement of the chemical properties (resistance of cable insulation in an aggressive environment)
- improvement of mechanical properties, such as strength, elongation, but also the mechanical flexural fatigue strength

The automobile industry is currently working on the transition from PVC insulation - in part cross-linked - to cross-linked PE insulations. Due to environmental criteria, in this case with respect to the noise level, the engine compartment is now better insulated. This does, however, also mean that the temperatures in the engine compartment increase and, therefore, cable insulation with a better temperature resistance is needed. In addition, the consumption of electrical energy in a car increases due to the higher comfort. This leads to an increase of the power that must be transmitted through the cables. The pending adaptation to a 42-volt network in the car is to be mentioned in this respect.

For reasons of quality assurance, irradiation cross-linking offers advantages compared to chemical cross-linking methods. The irradiation cross-linking process can be uniquely traced.

5.3. Tubes

The stress cracking resistance of PE is improved by means of irradiation cross-linking. This means that tubes made of cross-linked PE (see Figure 2) can be used in corrosive environments and under high mechanical stress.

Application: Tubes produced from

PE, EVA, PA, TPE, PU, PVC



FIG. 2. Tubes produced by irradiation cross-linking (PE,EVA,PA, TPE,PU,PVC).

Possible fields of application include:

- tubes for floor heating systems
- tubes for the connection of radiators
- tubes for warm water supply
- tubes for gas supply.

PE tubes that are cross-linked by irradiation have been used for floor heating systems since 1970. Since that time, the following development has taken place:

In the beginning, tubes coated with Ethylene-Vinyl-Alcohol-Copolymers (EVAL) were installed. The reason for this coating is to prevent the diffusion of oxygen into the water carrying tube.

Special adhesives and also special extrusion methods are required for the manufacturing of such tubes. The irradiation cross-linking also requires special know-how since a gas (hydrogen) develops during the cross-linking (see Figure 3). In addition, the mixture of hydrogen and oxygen results in an oxyhydrogen developer, i.e. a highly reactive mixture from which water eventually develops.



FIG. 3. Cross-Linking of Polyethylene (PE).

The further development in the last years showed a trend towards the deployment of these tubes for drinking water supply. This means that it was necessary to cross-link tubes with larger dimensions. To this purpose, electron accelerators with higher energies and larger handling systems are required.

The next requirement was the irradiation cross-linking of multi-layer tubes made of aluminium (AI) and polyethylene (PE). This presented a new challenge for the operators of irradiation facilities. The adhesion between AI and PE must not be lost due to the irradiation. During irradiation, AI heats up much more than PE. The internal tube must be cross-linked in any case. The multi-layer tube is much more rigid than a pure PE tube and, thus, presents additional requirements. These requirements have been met, the problems are solved.

The current requirements in this area are the following:

- Multi-layer tubes become more rigid with increasing outside diameters.
- It must be possible to irradiate the tubes as long sections.
- It must be possible to weld the cross-linked PE tubes together.
- Connectors are made of plastics.
- The tolerances with respect to the dimensions are becoming tighter.

These are some of the challenges to the operators of irradiation facilities, and each requirement is connected to a lot of additional minor requirements.

BGS was the first company worldwide to put into operation an electron accelerator able to irradiate parts with a length of up to 12 m (see Figures 4). The production has already started. Products, including long tubes, are successfully irradiated by BGS.



Application: Irradiation of tubes up to 12 m length (multi-layer-tubes, water supply, gas pipes)

FIG. 4. Irradiation of tubes with a length of up to 12 meters.

The possibility to irradiate these long PE tubes opens up a new market for irradiation technology: VPE tubes for gas supply.

Today, tubes, and also cables, are often not installed in an open trench. Instead, these tubes or cables are pressed in, i.e. the tubes/cables are installed using a trench less method. This requires a high stress cracking resistance of the tubes/cables. This type of installation causes an enormous load on the tubes/cable insulation, resulting in grooves, scratches, and damages, which may lead to subsequent failures if the tube/the cable insulation is not cross-linked.

In addition, tubes or cable insulation made of cross-linked PE can be installed in trenches without sand bedding, which is much more cost-effective.



FIG. 5. Irradiation cross-linking of corrugated tubes made of thermoplastics.

5.4. Corrugated Tubes

A corrugated tube is a flexible tube (see Figure 5). During use, it is subject to a flexural load. The irradiation cross-linking of corrugated tubes made of thermoplastics increases the number of flexural cycles, thus increasing the lifetime by many factors.

5.5. Injection Molded Parts

Injection molded parts made of PE have been cross-linked by irradiation for decades. The applications are locks, shrinkable products (heat-shrinkable end caps), parts that are subject to mechanical tension, gaskets and seals requiring a specific hardness or a specific temperature resistance, and many more.

We have irradiated parts made of cross-linkable, glass fiber reinforced PA since approximately 15 years (see Figure 6).



3D-MID-Parts, Coil former and Electronic Component produced fromPA and PBT

FIG. 6. Cross-linked glass fibre reinforced Polyamide parts.

The first applications originated from the electrical industry. Further applications from the automobile industry were added during the last years. Glass fiber reinforced PA that is cross-linked by irradiation has partly replaced the thermosets. The possible fields of application have certainly not been fully explored yet.

Polybutylene terephthalate (PBT), which is a plastic material for the electrical industry, can also be cross-linked by means of irradiation. Cross-linked PBT can be used with higher temperatures (see Figure 7). If you consider that solders must be lead-free in the future for environmental reasons, and that the temperature in the soldering bath increases, you can see that irradiation cross-linking can be the solution for a lot of these applications in the near future.



FIG. 7. Cross-linked PBT can be used with higher temperatures.

We are currently working on individual projects to see to what extent PP compounds can be cross-linked by irradiation. For the automobile industry, PP is a "commodity resin". Also here the idea is to increase the temperature stability of the parts by means of irradiation cross-linking.

Apart from the thermoplastics, the thermosets, and the elastomers, there has been a new "class" of plastics since about 10-15 years, the thermoplastic elastomers (TPE).

These are plastics that can be processed thermoplastically, i.e. they have a melting area, but possess elastomeric properties within the temperature range of their use.

TPE is a two-component plastic, whereby the two "components" may reside within a single polymer chain.

The physical cross-linking points can be crystalline areas or areas of the polymer that are within the temperature range of the application below the glass transition temperature i.e. that is "hard". The "soft" component is elastic, amorphous and not frozen yet. Actually, PE is also a TPE at room temperature. The physical cross-linking points are the crystallites, and the "soft" elastic area is the amorphous part.

These TPEs are almost ideal materials for seals and gaskets as they provide utmost flexibility with respect to their shape. Each sealing shape can be injection molded (see Figure 11). Unfortunately, the seals made of TPE can only be deployed within a very limited temperature range.



FIG. 8. Cross-linked seal produced from thermoplastic elastomer (TPE).

The irradiation cross-linking provides "real" elastomeric properties as chemical points of crosslinking are formed as a result of the irradiation. Especially in this case, the advantage of irradiation cross-linking becomes clear: the finished part, i.e. the injection molded seal, is cross-linked.

5.6. Curing

Fiber reinforced resins can be "cold" cured by irradiation. This results in a higher strength within the parts as delaminating is avoided and the internal tensions during the cooling process after heat curing are avoided. This allows the parts to become smaller for the same load. Irradiation curing leads to a weight reduction.

This technology is already used for satellites and airplanes. In case of further developments, it could also be implemented in "everyday" parts.

6. CONCLUSIONS

Beams are energy carriers. Beams trigger chemical reactions. We must stop people being afraid of beams by means of careful and well-considered handling of this technology. We must develop new applications. Based on these considerations, the acceptance of this technology will be achieved and new applications will be found.

RADIATION PROCESSING OF NANOCOMPOSITES

Khairul Zaman Hj. Mohd Dahlan, Jamaliah Sharif, Nik Ghazali Nik Salleh

Radiation Processing Technology Division, Malaysian Institute for Nuclear Technology Research (MINT), Bangi, Kajang, Malaysia

Abstract. Research on radiation processing of nanocomposites has been initiated at the Radiation Processing Technology Division of MINT in the past two years. The main focus of this research is to utilize indigenous natural polymers for production of nanocomposites materials. Natural rubber/clay composites and thermoplastic natural rubber/clay composites are the important materials that under studied. On the other hand, specialty products such as high abrasion and high scratch resistance materials have many industrial applications and it involved the usage of nano-sized silico-organic particles. In this area, palm oil acrylate is seen to have potential to be utilized in the formation of nano-sized silico-organic hybrids.

1. INTRODUCTION

In recent years there is a wide spread interest in development of nanostructured materials. The vast commercial potential of nanostructured materials has attracted the interest of the industry, academic institutions and government laboratories. The term nanostructured materials usually refer to "solids or thin film in which either the fundamental building block or the microscopic order are nanostructured". Generally, nanostructured material is defined as particles, grains, functional structures or devices with dimensions in the range of 1 - 100 nanometers. The much finer grain size can produce denser materials with greatly improved mechanical properties

The approach to produce nanostructured materials and devices are categorized by many scientists as 'top-down' and 'bottom-up' approached. Bottom-up means building larger objects from smaller building blocks. It involves the initial formation of the nanostructured molecules and atoms and their assembly into the final materials. Self-assembly is one of the most important bottom-up ways of creating nanostructures. Self-assembly consists of the spontaneous integration of the components bouncing in a solution, gas phase or interface until a stable structure is reached. Bottom-up approaches usually take place and inspired by chemical and biological systems.

The other important approach is still now vigorously promoted by scientists is 'top-down'. Top-down refers to the approach that begins with appropriate starting materials (or substrate) that is then 'sculpted' to achieve the desired functionality. This method is similar to that used by the semiconductor industry in fabricating devices out of a substrate by the methods of electron beam lithography and reactive ion etching. Another typical top-down approach is the 'ball-milling' technique, which involves the formation of nanostructure building blocks via controlled, mechanical erosion of the bulk starting substance. Those nano-building blocks are then subsequently assembled into a new bulk material.

Many workers have studied the fabrication of nanostructure materials using electron beam irradiation in particular in the field of nanolithography [1-3]. In this process, the energy of radiation is deposited on the materials via an ionization process. Namely, a molecule is spatially separated into a radical cation and an electron. The electron generated through ionization loses its energy through interaction with surrounding molecules and eventually thermalized. The initial separation distance between the radical cation and thermalized electron on average is approximately several nanometres, and depends on the material used [4]. It was also reported that ion beam produced a non-homogenous field of chemical reactions on polysilane thin film, where the reactive intermediates produced by an incident ion were radially distributed from the projectile [5]. The size of the field depended on LET, and varied from a few nm to several tens nm. The area where the ion beam deposited its energy is crosslinked and produced nano-sized cylindrical structure. When the uncross linked parts were dissolved, isolated nanowires are produced on Si surface. The use of radiation such as ion beam and

electron beam proved to be a great potential for the fabrication of nano-structured materials to be used in the lithography, membrane for ultrafiltration system, membrane with electrical and magnetic properties as a potential for chemical detectors and biosensors.

Radiation processing for cross-linking and grafting can be extended well into the processing of nanostructed materials in particular for producing crosslinked or grafted nanosize materials in a polymeric matrix or onto backbone of polymer chain respectively. In situ intercalative polymerization for poly (methyl methacrylate)/clay nanocomposite by gamma irradiation has been studied [6]. This paper will further elaborate the applications of electron beam processing for crosslinking/grafting of nanocomposites.

2. RADIATION PROCESSING OF NATURAL RUBBER/CLAY NANO COMPOSITES

Among the large number of inorganic layered materials that exhibit intercalation capabilities, layered silicates are one of the most typical because of the versatility of the reactions. In particular, the smectite group of clay minerals such as montmorillonite, sponite and hectorite has mainly been used because they have excellent intercalation abilities. The combination of clay silicate layers and polymer matrices at nanoscale level constitutes the basis for preparing an important class of inorganic-organic nano-structured materials.

The first such clay-based composites were synthesized by a Toyota research group [7, 8] and involved a polyamide as the polymeric phase. This work was expanded rapidly with a wide variety of other polymers including polystyrene [9], polypropylene [10-12], ethylene propylene diene monomer [13], poly (methy methacrylate) [14] and many others. The clay such as montmorillonite consists of a dioctahedral aluminium sheet sandwiched between two silica tetrahedral sheets in a layer structure that is ~ 1 nm thick (Figure 1). Stacking of the layers leads to a regular Van der waal's space between the layers called interlayer. Clay has the ability to undergo extensive interlayer expansion or swelling, exposing a large active surface area, permitting guest molecules to enter into the interlayer. Interlayer cations such as Na+, Ca++, and K+ exist on the internal surfaces but can be exchanged with alkyl ammonium cations to give surfaces that are less ionic or polar [15]. Such organically modified interlayers are more easily penetrated by polymers (either in the molten state or in solution) or by monomers that are subsequently crosslinked or polymerized chemically or by using radiation. In the process of polymer-clay nanocomposite synthesis, it is important to ensure the compatibility of the entering polymer/monomer (hydrophobic) with the clay surface interlayer (ionic) by using intermediate such as exchangeable organic based cations, intercalation agent or compatibilizer. Figure 2 shows the overall process of preparing nanocomposites based on clay and polymer/monomers.



FIG. 1. Structure of montmorillonite [16].



FIG. 2. Schematic view of preparation of methods for polymer intercalation compounds [17].

The entering polymer molecules into the interlayer can either simply increase the distance between the still-parallel layers in an intercalation process or randomly disperse the separate layers entirely in an exfoliation. In conventional composites, the registry of the clay nanolayers is retained when mixed with the polymer, but there is no intercalation of the polymer into the clay structure. Consequently, the clay fraction in conventional clay composites plays little or no functional role and acts mainly as a filling agent for economic considerations. An improvement in modulus is normally achieved in conventional clay composite, but this reinforced benefit is usually accompanied with a sacrifice in other properties such as strength or elasticity.



FIG. 3. Schematic illustration of the structure (a) conventional (b) intercalated and (c) exfoliated [18].

2.1. Preparation of organophillic clay and natural rubber / clay nanocomposites

Organophillic montmorillonite (MMT) nanolayers were prepared by cationic exchange process in an aqueous solution using several ammonium based cations to produce Cetyl trimethyl ammonium MMT (CTA-MMT), dodecyl ammonium MMT (DDA-MMT), hexadecyl ammonium MMT (HAD-MMT) and octadecyl ammonium MMT (ODA-MMT).

Two types of polymer-clay nanocomposites are possible, i.e. intercalated nanocomposites and exfoliated nanocomposites. Intercalated nanocomposites are formed (Figure 3) when one or a few molecular layers of polymer are inserted into the clay interlayer with fixed interlayer spacing. Exfoliated nanocomposites are formed when the silicate nanolayers are individually dispersed in the polymer matrix, the average distance between the segregated layers being dependent on the clay loading. The separation between the exfoliated nanolayers may be uniform or non-uniform. Exfoliated nanocomposites show greater phase homogeneity than intercalated nanocomposites. More importantly each nanolayer in exfoliated nanocomposites contributes fully to interfacial interaction with the matrix. This structural distinction is the primary reason why the exfoliated clay state is especially effective in improving the reinforcement and other performance properties of clay composite materials.

Organophillic clay was swelled in toluene and added to the solution of natural rubber (NR) that dissolved in toluene. The solution blends were stir for 24 hours and subsequently dried to remove the toluene. The natural rubber/clay nanocomposite was subjected to compression molding for physical, mechanical and analytical tests.

The molded test pieces of natural rubber/clay nanocomposites were also subjected to electron beam irradiation at 3.0 MeV, 10mA, at various dosages and followed by similar testing as un-irradiated samples.

2.2. Characteristic of organophillic clay and natural rubber / clay nanocomposites

Table 1 provides the basic information on the interlayer distance of sodium montmorillonite (Na-MMT) and modified MMT. Interlayer distance of Na-MMT expanded after modification with alkyl ammonium from 1.21 nm to about 1.67, 2.92, 3.41 and 2.02 nm for DDA-MMT, HAD-MMT, ODA-MMT and CTA-MMT respectively. Amount of alkyl ammonium intercalated increase with increasing alkyl ammonium chain length.

XRD analysis of unmodified Na-MMT and NR/Na-MMT composite indicate no change in the interlayer spacing of both compounds that is about 1.23m. This is because the unmodified Na-MMT particles are simply incorporated into the NR matrix in an agglomerated state with no intercalation take place. However, NR/modified MMT nanocomposites show higher interlayer spacing compare to unmodified Na-MMT as shown in Table 2.

Type pf clay	Exchange cation	2 θ (degree)	d ₀₀₁ – spacing (nm)	Amount of surfactants (mmol/g)	Content of surfactant (%)
Na-MMT	Na+	7.28	1.21	-	-
DDA-MMT	$C_{12}H_{25}NH_{3}^{+}$	5.26	1.67	0.81	15.2
HAD-MMT	$C_{16}H_{33}NH_{3}^{+}$	3.01	2.92	1.42	34.6
ODA-MMT	$C_{18}H_{37}NH_{3}^{+}$	2.58	3.41	1.51	40.9
CTA-MMT	$C_{16}H_{33}N(CH_3)_3^+$	4.78	2.02	1.38	44.4

TABLE I.	MODIFIED MONTMORILLONITES CHARACTERIZED BY X-RAY
DIFFRACTOME	FER AND CHNSO ANALYZER.

Type pf clay	Exchange cation	2 0	d ₀₀₁ – spacing (nm)
		(degree)	
NR/Na-MMT	Na+	7.20	1.23
NR/DDA-MMT	$C_{12}H_{25}NH_3^+$	2.59	3.40
NR/HAD-MMT	$C_{16}H_{33}NH_{3}^{+}$	2.50	3.50
NR/ODA-MMT	$C_{18}H_{37}NH_{3}^{+}$	2.37	3.72
NR/CTA-MMT	$C_{16}H_{33}N(CH_3)_3^+$	2.46	3.50

TABLE II. XRD ANALYSIS OF NR/MODIFIED MMT NANOCOMPOSITES.

3. ELECTRON BEAM CROSSLINKING OF NATURAL RUBBER/CLAY NANOCOMPOSITE

In this work, NR/CTA-MMT nanocomposite and the others were irradiated using electron beam accelerator, 3.0 MeV, 30 mA at room temperature. NR is an elastomer that can be crosslinked by radiation. The optimum dose for crosslinking of natural rubber is $\sim 200 - 250$ kGy [19]. Since no active reactive is added to NR/CTA-MMT, the optimum dose for crosslinking of the NR based nanocomposites is at 250 kGy as shown in Figure 1. At this dose, the tensile strength and elongation at break show significant increase from 1.0 MPa (un-irradiated) to 10.0 MPa (irradiated) and 600% (un-irradiated) to 750% (irradiated), respectively.

Figure 2 shows the effect of CTA-MMT content on the mechanical properties of NR/CTA-MMT nanocomposites irradiated at 250 kGy and in comparison with non-irradiated sample. The presence of nano-structured intercalated layers of clay in the matrix of NR shows significant improvement of the tensile strength and elongation at break. However, above 3 % of CTA-MMT content, the properties of the irradiated NR/CTA-MMT start to drop that indicate the re-agglomerated of clay layers. On the other hand for non-irradiated NR/CTA-MMT, the elongation at break increases with increasing CTA-MMT content up to 5%. Further studies are being carried out to determine the thermal stability, Tg and other properties of the irradiated NR/organophillic MMT.



FIG. 4. Effect of radiation dose on TS and Eb of SMRL 1 phr CTA-MMT.



FIG. 5. Effect of CTA MMT content on tensile and elongation at break of SMRL irradiated at 250 kGy.

Preliminary study indicates that radiation processing has great potential to be utilized in the processing of nanocomposites materials via crosslinking of the polymer matrix or grafting of the nanoparticles onto the backbones of the polymer molecules. The blend of natural rubber with thermoplastic and nanosized particles (clay or magnetic particles) will add further dimension to the study of radiation processing of nanocomposites based on natural rubber.

4. RADIATION PROCESSING OF NANO-SIZED PARTICLES – ORGANIC POLYMER COMPOSITES

The preparation of nanosized materials can be either 'top down' or 'bottom up' approached. In the case of top down, there are many nanosized particles that are available and being used as fillers, reinforced materials or other specific functions for composites. The use of nanosized silica as fillers for radiation crosslinked polyacrylates is one of the area that has been shown of great success [20,21]. In our laboratory, with the cooperation of IOM, Germany, work on development of radiation curing of silico-organic nanoparticle is in progress. The purpose of this research is to develop coating materials of high abrasion and scratch resistance. Several acrylates and AEROSIL OX50 based nanocomposites were synthesized in the laboratory by the heterogeneous hydrolytic condensation of the silane to silanol groups of the AEROSIL particles. The insitu reaction is proton catalysed and efficiently proceeds at 70C.

In this paper, some of the materials used are AEROSIL OX50 and silane couplaing agents such as VTMOS from Degussa, Germany and polyether tetraaacrylate (EB 40), acrylated trifunctional oligomer (OTA480), dipropylene glycol diacrylate (DPGDA), aliphatic urethane triacrylate with 15% HDDA (EB264), EB254, polyether acrylate (EB80) and epoxy acrylate (EB600) from UCB Chemicals, Belgium. The abrasion and scratch resistance properties of UV and EB curing of the acrylates based nanocomposites are shown in Figure 3 and Figure 4 respectively.

Table 3 indicates the significant improvement of the abrasion resistance of nanocomposites coating materials compared to pure acrylate based coatings. For the UV curing of nanocomposites, the higher the number of acrylate functional groups, the higher will be the abrasion resistance as indicated in Table 3 in the following order, EB 40 (tetra-functional) > OTA 480 (tri-functional) > DPGDA (di-functional).

At a relatively high nanopowder content of nanodispersion, UV induced polymerization proved to be an efficient alternative to EB curing. However, the scratch resistance of the nanocomposites are comparable to the pure acrylate coating materials as shown in Table 4.

This work has been expanded into the development of sol gel nanostructured organic – inorganic hybrid materials from palm oil acrylate. Currently, palm oil acrylate such as epoxidised palm oil acrylate (EPOLA) and Palm Oil based Urethane Acrylate (POBUA) are being synthesized in our laboratory.

TABLE III.	ABRASION RESISTANCE OF THE POLYMERIC NANOCOMPOSITES AND PURE
ACRYLATES W	/ITH FILM THICKNESS OF 50 μM.

Lacquers	Compositions	Weight Lost from Taber Abrasion (mg)		
		UV Irradiation	EB Irradiation	
Lack 1	30% AEROSIL OX50/ 61% EB	13.7	30.8	
	40/ 9% VTMOS			
Lack 2	30% AEROSIL OX50/ 61%	20.7	30.1	
	OTA 480/ 9% VTMOS			
Lack 3	30% AEROSIL OX50/ 61%	22.5	31.2	
	DPGDA/ 9% VTMOS			
EB40	100% EB40	55.7	-	
OTA480	100% OTA 480	65.5	-	
DPGDA	100% DPGDA	46.3	_	
EB254	100% EB254	_	41.5	
EB264	100% EB264	_	41.9	
EB80	100% EB80	_	61.6	
EB600	100% EB600	_	43.4	

TABLE IV. SCRATCH RESISTANCE OF THE POLYMERIC COMPOSITES WITH FILM THICKNESS OF 50 $\mu M.$

Type of Needles for	Lack				Pure Acrylates			
Scratch Test	1	10	11	12	13	DPGDA	OTA48 0	EB40
Diamond tip, 90°/N	3.0	3.0	3.0	3.0	2.0	3.0	3.0	3.0
Steel ball, Diameter	>10	>10	>10	>10	>10	>10	>10	>10
1 mm/N								

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EMERGING TECHNOLOGY REQUIRES AN EASY PROCESSING

H. de Rocquigny

COFRAR et RADIENT, Versailles, France

Abstract. The EBeam technology is becoming a mature energy moving out of the "research and development" stage to be installed as an industrial tool for the production of new material or new process within the respect of the environment. If this new energy in itself is well known, it remains a specialist business. The aim of the present report is to demonstrate that its development will pass by its vulgarisation and standardization making its use and its maintenance as easy as possible. The specialists have to develop systems which will allow the implementation of this technology at the end of the production line and even in the coming future on line with the production. This requires equipment standardization together with performing tools insuring the treatment reliability and traceability even being under the normal personal responsibility.

1. INTRODUCTION

EBeam ionization application started in the 1950 by sterilization application. Since then this technology is moving forwards supported by facilities installation and accelerators developments covering energies from some KeV to 10 MeV and Beam power from some kW to hundreds of kW.

Premium research got published updating the industrials knowledge on the electrical treatment and facilities management demonstrates its feasibility. Many applications are running in service centres, there will be transferred by number on the production line (or at the end of a production line) as soon as the industrial get a full confidence in the technology and in its key systems.

A quick resume on the existing experiences will allow us to detail some ways to follow up this development. I will mainly focus on the 10 MeV energy applications as these types of facilities seem to be the future due to accelerator development and its physical property and effect.

2. EXISTING TREATMENT FACILITIES AND ACCELERATORS SUPPLIERS

2.1. Actual installed facilities

Since the beginning of 1900 10 MeV industrial facilities are within the range of 40 units and some more in Russia. 21 are in Europe, 17 being multi-purpose, 1 belong to a central logistic centre, 3 are installed "at the end of a production line" for medical sterilization or composite polymerisation.

2.2. Main 10 MeV Accelerator suppliers.

Referring to these facilities, there are some 5 main suppliers of 10 MeV accelerators, 2 being European, 1 American, 1 Japan and 1 Russian. Most of them use the technology: "linear acceleration though multi-cavity" called "Linac", one use "multi pass through a single cavity" technology called Rhodotron. To resume, 10 MeV Linac accelerators deliver some 1 to 60 kW of power, 10 MeV Rhodotron are more likely to deliver some 25 kW up to hundreds even as I understood thousand of kW

2.3. Facility design and installation

Referring again to the European facilities, 50% of them are design and built by its owner with advices of the accelerator supplier. European engineering company, specialized in such design and installation, get involved with most of the others.

3. EXISTING AND EMERGING EBEAM APPLICATIONS

EBeam applications sectors may be listed together with some of their expecting emerging applications or concepts. Micro-processor treatment and some others exist generally covered by confidentiality agreement.

3.1. Medical products sterilization

It follows a high development since 20 years against the Ethylene oxide which remains particularly active in the "complete set" sterilization due to inhomogeneous packaging. Corresponding emerging application or concept may be in house sterilisation, inhomogeneous package treatment and on line treatment

3.2. Food decontamination

This activity is close to zero in Europe due to product "labelling". Large development is experienced in US for meat pasteurization. A successful marketing of "Electrical sources" for food treatment may transform develop this sector under the request of "high precaution principle".

3.3. Polymers treatment: tubes, ducts, conduits, wires protection

This sector is a strong support of the EBeam technology due to its advantage such as minimizing the material degradation, oxidation and heat transfer by time reduction and dose delivery accuracy. One of the merging applications could be the treatment in thickness with "10 MeV energy".

3.4. Main polymers pieces or large scale structure for cars, trucks, planes and railways builders.

These types of treatments are forcing their way out of the R&D department for most of the major industrial. These types of application are industrially new and will require particular concept to face huge treatment capacity and large scale elements.

4. TREATENT FACILITY SYSTEMS REQUIREMENT

Four main systems are required to develop an "electrical source ionization unit"; their interface may vary according to the applications.

a. The handling and moving system insures the quality and the reproducibility of the treatment by controlling:

The movement of the product under the beam to insure the quantity of energy delivered into the product matter or packaging.

The multi pass (transfer and turn around) of the product, insure the productivity

The regulation of the product loading and unloading into the treatment cell controls the treatment capacity, and its viability.

The cost of this system rises according to the product dimension and EBeam power.

b. The accelerator delivers a nominal Electron Beam at the exit of the window horn. Its power must be reliable and sized as required by the application and production rate. Normally unique on site, its maintenance capability is a key point of decision. The cost is function of the power and of its technology. An easy energy variation would be a real plus for applications processing.

c. The security system may be divided in two parts – One is almost a constant. It covers the cell shielding access, video, security equipment, procedure and control. The second is the concrete treatment cell; its geometry and sizes are to be adapted to the specific application.

d. The treatment supervision system guaranties the treatment and its reproducibility. It mainly consist of two computer systems insuring respectively the *treatment order* with its *supervision* and the

control of the *treatment reproducibility* by recognition of each piece before and after their treatment together with the process parameters versus the product validated treatment data.

e. Standard 10MeV & 10 kW EBeam treatment unit cost: Scale for in house production, the direct cost of a standard in house electrical sterilization facility (normal treatment units size) is within the range of 2,6 Million \in . It can be divided in 4 main lots:

|--|

Handling system to place and move objects in front of the EBeam	12%
EBeam Accelerator and its annexes	48%
Security system and appropriate concrete cell and equipment	28%
Production treatment supervision and traceability.	12%

5. INDUSTRIAL DEVELOPMENT REQUIREMENT

The requirement for existing applications is mostly known by specialists. But emerging applications will be developed at large scale if they are installed close to the production lines. At first, industrials have to be convinced that this technology is easy, reliable and cheap and does not belong solely to "research and development" departments

5.1. The processing operation has to be handy!

a. Handling system must be practical. It is the key factor to perform quality on high production rate.

Its design should be adapted to the dedicated application. Its operation must be reliable as any adjustment or repair takes time due to security access.

Accordingly, the handling system has to be simplified in its:

Concept by a succession of small and dedicated handling elements.

Design by choosing each element as standard mechanic unit and assembled on resistant structure.

Speed adapted to the treatment capacity and operator capability

Maintenance to be assumed by standard personal.

b. Accelerator must work easily, EBeam accelerator power dictates the treatment speed and the productivity. Its design must be standard so that the maintenance can be minimize and placed under the responsibility of normal maintenance personal.

Defaults have to be quickly detected by increasing the number of checking points.

Monitoring must be design to fit with the main "operation system"

The Beam power should be adapted to the industrial production rate. Any increase in the power largely influences the handling system, its control and the treatment cell dimensions. A production increase would preferably be covered by a second facility installation which will increase the whole production flexibility.

c. Treatment cell geometry and security has to simplify operation as security, handling and maintenance are directly pending upon.

Coordinated with the cell access, the treatment cell must be adapted to the application, the handling device and the process quality.

"Making operation easy" means:

Full radiation security with a minimum of controller apparatus and a maximum of fix security screens.

Easy access to the treatment cell for personal and equipment

Sufficient spaces for equipment maintenance.

d. Make the Reproducibility easy by means of proper equipment design to eliminate any operator misleading.

Such objective requires:

To design the various systems so that they works individually under an automatic coordination

To design the various computer controller systems so they are integrated in one unique supervision system 100% secured.

e. Facility specialization makes all systems easier as all of them are totally delegated to one particular industrial application.

5.2 **Processing has to be cheap**

The figures presented for a standard investment are still high when applied to an in house production even if the gain in time, logistics cost and production breakage is taken in account.

One way to prevent such cost during the first years is for the industrial to operate in partner ship with a specialist firm. Within the market, this partner may use the unit for some external treatment. The partnership stops when the internal production takes the full capacity of the facility. This evolution combines an easy operation starting and a positive technology transfer. COFRAR experienced such development with one leader in laboratory tools manufacturer in France. This experienced will end in May 2003. A similar experience will start this year.

Another way is to reduce the price of each system by developing standard equipment. We experienced also some system standardization like access cell, security, dosimetry and boxes handling systems. Some more will be developed soon.

The total investment will then be the addition of 2 different parts:

- A standard package for the access cell, accelerator casemate, security, accelerator with services and supervision.
- A specialized package for the treatment system and the operation control

6. CONCLUSION

The electric sterilization seems to move forward through in house "medium power, high energy EBeam" standard facilities. They are accepted by industrials as a competitive treatment concept easy to operate, able to trace unit by unit the treatment quality with even a possible individual labelling input which covers the "individual legal treatment guarantee", a real plus!

The same can be applied to R&D facilities and to other emerging applications as we are experiencing some with success.

With the help of the EBeam specialists together with industrials and customers, we think that for most of the emerging applications the "Medium Power EBeam technology" will be granted as an easy, reliable and cheap investment supporting the "Electrical Source Radiation process".

DEMONSTRATION PLANT FOR ELECTRON-BEAM TREATMENT OF TAEGU DYE INDUSTRY COMPLEX WASTEWATER

I.E. Makarov^a, A.V. Ponomarev^a, Bumsoo Han^b

^a Institute of Physical Chemistry of Russian Academy of Sciences, Moscow, Russian Federation ^b EB-Tech Co., Ltd, Daejeon, Republic of Korea

Abstract. The main features of basic design of the plant for industrial wastewater treatment using electron beam are described, including chemical aspects of the process, process technology, and building construction. The plant with treatment capacity 10,000 cubic meters of wastewater a day is proposed to be located at the area of existing wastewater treatment facility of Dye Industry Complex in Taegu city, Korea. Total area required for the plant is 220 m². Total electric energy consumption is inside the range 764 (min) and 1265 (max) kW. For electron-beam treatment 1 MeV-400 kW accelerator will be utilized. Besides conventional purposes, the plant will be used also for collection of new data on radiation treatment of wastewater under real industrial conditions and as a demonstration facility for training and for popularization (advertising) of radiation technology.

1. INTRODUCTION

According to construction schedule for Technical Co-operation Project of IAEA "Construction of Demonstration Facility for Industrial Wastewater Treatment with Electron Beam", the first stage consisted of basic design of the plant. The plant is proposed to be located on the area of existing wastewater treatment facility in Taegu Dye Industry Complex (TDIC) and to have treatment capacity 10,000 cubic meters of wastewater a day. The process of electron-beam treatment of wastewater has not been implemented anywhere, as yet, at such a large scale. So, the purpose of basic design was to estimate the following principal parameters of the plant:

- area required for building and communications;
- features of building construction;
- features of processing (technological scheme);
- nomenclature and location of the equipment;
- electric energy and cooling water demand.

Basic design should serve as a source for detail design of the plant. In this article chemical aspects (backgrounds) of the process, process technology, and building construction are described.

2. CHEMICAL ASPECTS OF THE PROCESS

In the process of electron-beam treatment of wastewater there are utilized chemical transformations of pollutants induced by ionizing radiation. At sufficiently high absorbed doses these transformations can result in complete decomposition (removal) of the substance. Under real conditions, i.e., at rather high content of pollutants in a wastewater and economically acceptable doses, partial decomposition of pollutant takes place as well as transformations of pollutant molecules that result in improving subsequent purification stages, efficiency of the process being notably influenced by irradiation conditions and wastewater composition.

2.1. On the TDIC Wastewater Composition

Characteristics of TDIC wastewater undergo both short-term and long-term variations, the former being equal to 10-13 % while the latter amounting up to 20 % of mean value. Overall characteristics of influent wastewater, such as 5 day's biological oxygen demand (BOD₅), chemical oxygen demand measured by dichromate method (COD_{Cr}), chemical oxygen demand measured by permanganate method (COD_{Mn}), and total organic carbon (TOC) have been reported [1] to be as presented in Table 1.

1.2. PARAMETER	BOD ₅	\textbf{COD}_{Cr}	\textbf{COD}_{Mn}	TOC
Min. value, mg/l	1320	2240	520	860
Max. value, mg/l	2340	3870	840	1564

TABLE I.CHARACTERISTICS OF TDIC WASTEWATER

Results of more recent measurements were inside the min-max ranges listed in Table 1. Content of suspended solids (SS) was measured to be near 100 mg/l. Initial temperature of the wastewater is 40-50 °C and after cooling tower is reduced to 32-35 °C. Initial pH, before neutralization, is 10-13.

Chemical composition of the wastewater also is not constant. Data of chemical analysis showed that averaged chemical composition of dissolved organic impurities in influent wastewater is approximately as following (in weight percent of total dissolved organics):

- terephtalic acid in dissociated or semi-dissociated form (TPA) from 75 to 85 %
- ethylene glycol (EG) from 15 to 18 %
- organic dyes less than 1 %
- surfactants less than 1 %
- other organic compounds less than 2 %

TPA and EG are definitely the major components of the wastewater pollutants. Organic dyes and surfactants, even at comparatively low concentration, determine such objectionable properties of the wastewater as color and foaming, so concentration of these compounds should be substantially reduced. Among other organic compounds there are: hexane, carboxymethyl and hydroxymethyl cellulose, phenols, starch, waxes.

Inorganic compounds are presented mainly by sulfate anion and sodium cation (as result of pH adjustment) and small amounts of chlorides and carbonates. Besides, some amount of hydrogen peroxide may be present. The latter, unlike other inorganic compounds, can take part in radiation induced transformations of organic compounds.

Both major dissolved polluting components of wastewater, i.e. TPA and EG, are biodegradable and both of them are oxidized by dichromate while only EG is oxidized by permanganate. Thus, BOD, COD_{Cr} , and TOC values are caused, mainly, by both TPA and EG while COD_{Mn} characterizes only content of EG (and some other compounds). Molar concentrations of TPA and EG in TDIC wastewater are close to each other, being in the range from 6 to 9 mmol/l.

Composition of suspended solids can not be determined precisely. Usually, those contain undissociated TPA, polymers, insoluble inorganic compounds, etc.

2.2. Primary Radiation-Chemical Processes in the System

Total concentration of soluble compounds in the wastewater does not exceed 0.3% (mass), their electron fraction being less than 1/3000. Therefore, direct action of ionizing radiation on these compounds may be neglected, i.e. more than 99.7% of accelerated electrons, as well as secondary electrons arising in the medium, interact with water molecules producing chemically active short-lived particles: hydrated electrons (e_{aq}), atoms H, and radicals OH, their initial radiation-chemical yields, $G_{e_{aq}}$, G_{H} , and G_{OH} , being equal to 0.27, 0.06, and 0.28 µmol/J, correspondingly [2]. Also, hydrogen peroxide which can take part in some of reactions with solute is produced as primary product of water radiolysis with the yield $G_{H_2O_2}$ =0.07 µmol/J [2]. When combined electron-beam and ozone treatment
is used, additional amount of hydroxyl radicals is produced in reactions of dissolved ozone with hvdrogen peroxide^{*}:

$$\begin{array}{rcl} O_3 \ + \ H_2 O_2 \ \longrightarrow \ HO_3 \ + \ HO_2 \\ HO_3 \ \longrightarrow \ OH \ + \ O_2 \end{array}$$

Rate constants for reactions of hydrated electrons, H atoms, and OH radicals with most of organics present in the wastewater, including TPA and EG, are high enough^{**} to prevent reactions between short lived particles. Stationary concentration of the latter at dose rate 50 kGy/s and concentration of organic pollutants in water near 0.008 mol/ 1^{***} is equal to 5.10⁻⁷ mol/l. Also, it should be taken into account that both types of reducing particles, i.e., e_{aq}^{-} and H-atoms, react rapidly with dissolved oxygen producing peroxide radicals. Because concentration of dissolved oxygen in naturally air saturated water is slightly higher than $1 \cdot 10^4$ mol/l and rate constants of its reactions with e_{aa} and Hatoms are near $2 \cdot 10^{10} \, \text{I mol}^{-1} \, \text{s}^{-1}$ [3], these reactions are competitive with reactions of e_{aq} and H-atoms with dissolved pollutants at absorbed doses up to ca. 0.5 kGy.

At total absorbed dose about 3 kGy and irradiation time much less than 1 s (that makes oxygen impossible to diffuse from surface to the bulk of liquid under irradiation), fraction of e and H-atoms participating in reactions with oxygen is estimated to be less than 30%. So, G-values for OH-radicals, H-atoms, and hydrated electrons reacting with dissolved organics in the bulk of solution may be estimated to be near 0.3, 0.2, and 0.04 μ mol/J, correspondingly; total G-value for peroxide radicals $(O_2^- \text{ and } HO_2)$ being equal to about 0.12 μ mol/J. The latter are in acid-base equilibrium.

$$HO_2 \leftrightarrow O_2^- + H^+ \qquad \{pK_A \ 4.7\}$$

and, mainly, combine with each other producing hydrogen peroxide and molecular oxygen:

$$\begin{split} &HO_2 + HO_2 \longrightarrow H_2O_2 + O_2 \\ &O_2^- + HO_2 + H_2O \longrightarrow H_2O_2 + O_2 + OH^- \\ &O_2^- + O_2^- + 2H_2O \longrightarrow H_2O_2 + O_2 + 2OH^-, \end{split}$$

or can take part, also, in reactions with organic radicals of solute that results in formation of organic peroxides and, in some cases, cleavage of C-C bonds.

So called "spur" or "track" reactions plays minor role at pollutant concentration near or less than 10⁻³ mol/l, even if its rate constant with primary water radiolysis products are high enough. Increase in G-value of the primary products of radiolysis due to penetration of pollutant molecules into "spurs" would not exceed 10%.

2.3. Radiation Induced Decomposition of TPA, EG, and Some Other Soluble Components of **TDIC Wastewater**

Both major components of TDIC wastewater, TPA and EG, efficiently react with radical products of water radiolysis, in the case of TPA the first stage being attack of all radicals on the benzene ring, in the case of EG - abstraction of H atom by H and OH radicals (rate constants from [3]):

^{*} The reaction seems to proceed mainly with dissociated form of H_2O_2 , i.e.: $O_3 + HO_2^- \rightarrow HO_3 + O_2^-$ ** Except reaction of e_{aq}^- with EG and other saturated alcohols

^{***} It corresponds to conditions of TDIC wastewater composition, flow rate 420 m²/h, and e-beam power 400 kW



$$H + HOCH_2 - CH_2OH \rightarrow H_2 + HOCH_2 - CHOH \qquad \{k = 1.4 \cdot 10^{7} \text{ 1 mol}^{-1} \text{ s}^{-1}\}$$

$$\dot{H} + HOCH_2 - CH_2OH \rightarrow H_2 + HOCH_2 - \dot{C}HOH \qquad \{K = 1.8 \cdot 10^{9} \text{ L MOL}^{-1} \text{ S}^{-1}\}$$

Further transformations of TPA radicals are realized (in reactions with other radicals donating H-atoms) in changing benzene ring into cyclodiene structure, formation of phenols and decyclization:



Decyclization process is accompanied by partial decarboxylation, i.e. elimination in some cases one or two carboxylic groups that results in formation of hydrocarbons with lower molecular weight and carbon dioxide. It is confirmed by observed decrease in TOC and COD_{Cr} values upon radiolysis of aqueous solutions of TPA at doses up to 2 kGy (see Table 2). Also, it should be mentioned that oxygen plays important role in radiolytical degradation of TPA, the main type of its reactions being an addition to organic radicals:

$$O_2 + \dot{R} \rightarrow R - O - \dot{O}$$

The yield of TPA degradation by elimination of carboxyl group calculated on the base of COD_{Cr} and TOC measurements are presented in Table 2 together with maximal initial yield of radicals from TPA which corresponds to total yield of primary radicals from water, i.e. 0.54 μ mol/J considering the reactions of primary radicals with oxygen.

Initial concentration of TPA, mg/l	50	100	250	500
$\Delta COD_{Cr} / \Delta Dose (mg l^{-1} kGy^{-1})$	103	140	125	130
$\Delta TOC / \Delta Dose (mg l^{-1} kGy^{-1})$	30	40	75	70
Average yield of decarboxylation (µmol/J)		4.2 ±	0.8	
Average degradation yield (for TPA) (µmol/J)		0.52 ±	0.10	
Max. yield of organic radicals (µmol/J)		$0.54 \pm$	0.05	

TABLE II.YIELDS OF TPA DEGRADATION

Data presented in the Table show that yield of complete TPA decomposition is close to the yield of TPA radicals formed in TPA reaction with primary radicals from water radiolysis, while the yield calculated for C-atoms, i.e., decarboxylation yield, is much higher. It means that some chain processes take place during radiolysis of TPA in aqueous solutions, chain propagation step being reaction of organic radicals with other organic molecule.

Ethylene glycol radicals formed in reactions 9, 10 are known [2] to take part in reactions of combination and disproportion, as well as in monomolecular reaction of dehydration, giving rise to formation of tetraoxybutane, glyoxal, and acetic aldehyde:

$$2 \operatorname{HOCH}_{2} - \dot{\operatorname{C}} \operatorname{HOH} \longrightarrow \operatorname{HOCH}_{2} - (\operatorname{CHOH})_{2} - \operatorname{CH}_{2} \operatorname{OH}$$

$$2 \operatorname{HOCH}_{2} - \dot{\operatorname{C}} \operatorname{HOH} \longrightarrow \operatorname{HOCH}_{2} - \operatorname{CH}_{2} \operatorname{OH} + \operatorname{HOCH}_{2} - \operatorname{CHO}$$

$$\operatorname{HOCH}_{2} - \dot{\operatorname{C}} \operatorname{HOH} \longrightarrow \operatorname{H}_{2} \operatorname{O} + \dot{\operatorname{C}} \operatorname{H}_{2} - \operatorname{CHO} \xrightarrow{\operatorname{H}} \operatorname{CH}_{3} \operatorname{CHO}$$

In the presence of oxygen fast reaction of its addition to the radicals proceeds:

resulted in organic molecules oxidation and decarboxylation. Processes of oxidation and decarboxylation (like in the case of TPA solutions radiolysis) proceed, as well, in reactions of peroxide radicals and hydrogen peroxide with organic radicals. Yields listed in Table 3 show that chain processes in this case also take place.

Initial concentration of EG, mg/l	50	100	250	500
$\Delta COD_{Cr} / \Delta Dose $ (mg l ⁻¹ kGy ⁻¹)	35	80	70	20
$\Delta \text{COD}_{Mn} / \Delta \text{Dose} \text{ (mg } l^{-1} \text{ kGy}^{-1} \text{)}$	40	70	60	25
$\Delta TOC / \Delta Dose (mg l^{-1} kGy^{-1})$	30	15	20	25
Average yield of decarboxylation, µmol/J		1.6 ±	0.6	
Average degradation yield (for EG), µmol/J		$0.72 \pm$	0.27	
Max. yield of organic radicals, µmol/J	0.54 ± 0.05			

TABLE III. YIELDS OF EG DEGRADATION

Organic dyes are easily decomposed in aqueous solutions upon irradiation. It is due to effective destruction of chromophoric groups and related system of conjugated double-bonds (that cause intensive light absorption in visible range) by radical products of water radiolysis. Usually, on the first stage of radiolysis, in reactions of dye molecule with e_{aq} and H-atom the dye radical named semi-reduced form of dye is formed, while in reaction with OH-radical semi-oxidized form is formed. Reactions of electron and/or hydrogen transfer between semi-reduced and semi-oxidized forms of dye result in partial reparation of dye molecules. Such reactions between two semi-reduced or two semi-oxidized forms lead to degradation of dye. Decoloration yields for aqueous solutions of soluble organic dyes, as a rule, are equal to 0.1-0.3 μ mol/J, depending on the structure of dye, i.e. some less than total yield of primary radicals of water radiolysis because of reparation reaction, or may be higher in some cases due to chain reactions.

Figures 1 illustrates the results of electron-beam treatment of aqueous solutions of one of organic dyes, being characteristic of TDIC wastewater: reactive blue dye RBR BB. It is seen that almost complete decoloration is observed at absorbed dose less than 7-9 kGy. Presence of dissolved oxygen increases decoloration rate; however, at high decoloration efficiency, as in the case of reactive blue dye, the effect of oxygen on decoloration process is small.



FIG. 1. Degradation of reactive blue dye RBR BB in aqueous solutions (50 mg/l) upon electron-beam treatment: **a** - decrease in relative absorbance at 510 nm with dose in deaerated (1) and aerated (2) solutions; **b** - decrease in COD_{Cr} (1) and TOC (2) with dose in aerated solutions. Insert in **a** – optical absorption spectra of the solution before and after irradiation at 7 kGy.

Together with decoloration of dyes, total degradation of organic molecules into carbon dioxide and water takes place. It follows from presented in the figures data on changes in COD_{Cr} and TOC values of dye solutions upon electron-beam treatment, where substantial decrease in both COD_{Cr} and TOC is observed in absorbed dose range of several kilograys.

Synthetic surfactants, like other organic compounds in aqueous solutions, are decomposed in reactions with primary radicals from water followed by reactions of organic radicals, oxidation and decarboxylation. Initial yield of decomposition of conventionally used surfactants in aqueous solutions is about 0.2 μ mol/J [4]. It is decreased with increasing the absorption dose. Thus, total dose 300 kGy was found to be required for complete decomposition of such surfactant as isobutylnaphtalene sulfonate at initial concentration near 300 mg/l [5]. Usually, there is no necessity to decompose all the substance, since concentration of surfactant in wastewater, comparing to other organic compounds, is relatively low and surfactant is not extremely toxic itself. Elimination or transformation of functional group is sufficient for deactivation of the surfactant or reducing its surface activity. It makes also the molecule biodegradable and does not require high absorption dose.

Radiation induced degradation of other organic compounds presented in TDIC wastewater, like saturated hydrocarbons, phenols, derivatives of cellulose, etc., proceeds approximately in the same way as that of described above. There are two principal stages in degradation process: a) reactions of primary radicals from water (e⁻_{aq}, H, OH) with the compound and with the products of its transformation - formation of organic radicals; and b) reactions of organic radicals including reactions with organic molecules, oxygen, hydrogen peroxide and peroxide radicals, combination and disproportion reactions; the reactions being accompanied by C-C bond rupture resulted in decrease of mean molecular mass, decyclization, and decarboxylation.

Radiation stability of the compounds does not differ too much, and initial yields of degradation are in the range of $0.05-0.3 \ \mu mol/J$, unless radiation induced chain processes take place. It is important to stress, however, that in the case when the compound is present in solution as a minor component of mixed solute, its initial degradation yield is lower than it would be if the compound were present in solution alone. The decrease in initial degradation yield of minor component is approximately proportional to ratio of concentrations of major and minor components.

2.4. Total Changes in Wastewater Characteristics under EB Treatment

The main changes in TDIC wastewater resulting from electron-beam treatment concern transformations of TPA and EG molecules as major components of soluble pollutants. Mean yield of complete degradation of the compounds, as follows from Tables 2 and 3, is equal to about 0.6 μ mol/J. It means that less than 10 % (mass) of organic substance will be completely decomposed into water and carbon dioxide at initial concentration near 1500 mg/l and absorbed dose up to 3 kGy. Numerous experiments on electron-beam treatment of TDIC wastewater showed, indeed, that no significant decrease was observed for parameters characterizing total content of organic compounds, such as TOC, COD, and BOD, after irradiation at several kGy absorption dose. The most notable effect of just irradiation was decrease in color of the wastewater as a result of dyes molecules decomposition.

High yields of decarboxylation (4.2 μ mol/J for TPA and 1.6 μ mol/J for EG, see Tables 2, 3) demonstrate, however, that major part of organic molecules initially contained in solution, from 50 to 90 %, undergo structural transformations. These transformations have an effect on efficiency of the further wastewater treatment processes, like chemical coagulation, settling, and, mainly, biotreatment (see Fig. 2).



FIG. 2. Effect of electron-beam treatment on biological treatment of TDIC wastewater: \mathbf{a} - kinetics of biotreatment of irradiated (1) and unirradiated (2) wastewater; \mathbf{b} - absorbed dose effect on combined electron-beam/biological treatment.

From results of laboratory and pilot plant experiments it follows that optimum absorbed dose while electron-beam treating TDIC wastewater, that provides appreciable improvement of subsequent biological treatment, is 3 kGy. At this dose additional decrease in TOC, COD, and BOD after biological treatment of irradiated wastewater, comparing to unirradiated one, may achieve 30-60 %, and hydraulic retention time of biotreatment may be reduced by factor of two at the same biotreatment efficiency [6].

2.5. Influence of Ozone on EB Treatment Process

Ozone is formed in the air upon irradiation of wastewater. It can be used for additional treatment of the wastewater by bubbling irradiated air through wastewater before or after its electronbeam treatment.

Interaction of ozone with organic pollutant dissolved in water proceeds in two ways, relative efficiency of each one being dependent on conditions:

- indirect reaction, i.e., transformation of ozone into radicals in reaction with water and subsequent reactions of appeared radicals with organic molecules.
- direct reaction of ozone with organic molecules;

The first way is favored by increased pH-value of the solution and relatively low reactivity of particular organic molecules to ozone. Higher pH-values promote transformation of ozone into radicals because of higher efficiency of ozone reaction with hydroxy-anions comparing its reaction with water. The following ozone reactions in water have been found to proceed (rate constants from [3, 7, 8]):.

PEROXIDE RADICALS FORMATION

 $\mathbf{O}_3 + \mathbf{OH}^- \longrightarrow \mathbf{HO}_2 + \mathbf{O}_2^- \qquad \{k = 70 \ 1 \ \mathrm{mol}^{-1} \ \mathrm{s}^{-1}\}$

OZONIDE RADICAL-ION FORMATION

$\mathbf{O}_3 + \mathbf{O}\mathbf{H}^- \longrightarrow \mathbf{H}\mathbf{O}_2^- + \mathbf{O}_2$	$\{k = 40 \ \mathrm{M}^{-1} \mathrm{s}^{-1}\}$
$\mathbf{O}_3 + \mathbf{HO}_2^- \longrightarrow \mathbf{HO}_2 + \mathbf{O}_3^-$	$\{k = 2.2 \cdot 10^6 \mathrm{1 mol^{-1} s^{-1}}\}$
$\mathbf{O}_3 + \cdot \mathbf{O}_2^- \longrightarrow \cdot \mathbf{O}_3^- + \mathbf{O}_2$	$\{k = 1.6 \cdot 10^9 \mathrm{l mol^{-1} s^{-1}}\}$
$\cdot \mathrm{HO}_3 \longleftrightarrow \cdot \mathrm{O}_3^- + \mathrm{H}^+$	$\{pK_A 8.4\}$

HYDROXYL RADICAL FORMATION

OTHER REACTIONS OF OZONE AND OZONIDE RADICAL-ION

$\mathbf{O}_3 + \mathbf{e}_{aq} \longrightarrow \mathbf{O}_3^-$	$\{k = 3.6 \cdot 10^{10} \mathrm{l mol^{-1} s^{-1}}\}$
$\mathbf{O}_3 + \mathbf{\cdot} \mathbf{H} \longrightarrow \mathbf{\cdot} \mathbf{O} \mathbf{H} + \mathbf{O}_2$	$\{k = 3.8 \cdot 10^{10} \mathrm{l mol^{-1} s^{-1}}\}$
$\mathbf{O}_3 + \mathbf{O}\mathbf{H} \longrightarrow \mathbf{H}\mathbf{O}_2 + \mathbf{O}_2$	$\{k = 1.1 \cdot 10^8 \text{ l mol}^{-1} \text{ s}^{-1}\}$
$\cdot \mathbf{O}_3^- + \cdot \mathbf{O}\mathbf{H} \longrightarrow \cdot \mathbf{H}\mathbf{O}_2 + \cdot \mathbf{O}_2^-$	$\{k = 6 \cdot 10^9 \mathrm{l \ mol^{-1} \ s^{-1}}\}$
$\cdot \mathbf{O}_3^- + \cdot \mathbf{O}\mathbf{H} \longrightarrow \mathbf{O}_3 + \mathbf{O}\mathbf{H}^-$	$\{k = 2.5 \cdot 10^9 \ 1 \ \mathrm{mol}^{-1} \ \mathrm{s}^{-1}\}$
$\cdot OH \leftrightarrow - O^- + H^+$	${pK_{\rm A} \ 11.9}$
$\cdot \mathbf{O}_3^- + \cdot \mathbf{O}^- \longrightarrow \cdot \mathbf{O}_2^- + \cdot \mathbf{O}_2^-$	$\{k = 7 \cdot 10^8 \ 1 \ \mathrm{mol}^{-1} \ \mathrm{s}^{-1}\}$
$\cdot \mathbf{O}_3^- + \cdot \mathbf{O}^- \longrightarrow \mathbf{O}_4^{2-}$	$\{k = 8 \cdot 10^8 1 \text{mol}^{-1} \text{s}^{-1}\}$
$\cdot \mathbf{O}_3^- \longrightarrow \cdot \mathbf{O}^- + \mathbf{O}_2$	$\{k < 10^5 \text{ s}^{-1}\}$
$\cdot 0^{-} + 0_{2} \longrightarrow \cdot 0_{3}^{-}$	$\{k = 3.6 \cdot 10^9 \ 1 \ \text{mol}^{-1} \ \text{s}^{-1}\}$

Efficiency of direct reactions of ozone with organic compounds depends on structure of the compound. As strong nucleophilic reagent, ozone reacts presumably with aromatic and unsaturated aliphatic compounds, the reaction being more efficient in the presence of electron donor groups in the compound. Besides, ozone is highly reactive to some specific functional groups, e.g., -S-, -NH.

Combined electron-beam and ozone treatment of wastewater polluted with organic compounds is often characterized by synergistic effect. From the data presented in Table 4 it follows that combined treatment is more efficient than summarized efficiency of only radiation treatment and only ozone treatment.

TABLE IV. COMPARATIVE EFFICIENCY OF RADIATION, O₃, AND COMBINED TREATMENT

Compound to be removed	Removal degree, %			
Compound to be removed	Radiation treatment	Ozone treatment	Radiation/O ₃ treatment	
Tetrachloroethylene [9]	70	~0	85	
Phenol [10]	5	45	96	

High efficiency of combined radiation and ozone treatment is explained by promoting chain reactions due to ozone induced conversion of relatively unreactive peroxide radicals from water into hydroxyl radicals and organic peroxide radicals into much more reactive alkoxy-radicals:

$$\mathbf{RO}_2 \cdot + \mathbf{O}_3 \longrightarrow \mathbf{RO} \cdot + \mathbf{2O}_2$$

and further formation of organic radicals R.

 $\cdot OH + RH \longrightarrow H_2O + R \cdot$

 $RO \cdot + RH \longrightarrow ROH + R \cdot$

which react with oxygen forming again organic peroxide radicals.

Application of ozone for TDIC wastewater treatment appears to have positive effect in either of two variants:

- after electron-beam treatment to induce additional degradation of organics;
- before electron-beam treatment to promote chain processes.

3. PROCESS TECHNOLOGY

Technology of the process was designed for plant capacity 10,000 tons of wastewater a day. According to the data obtained in laboratory and pilot plant experiments with TDIC wastewater, the optimum absorbed dose for electron-beam treatment was chosen to be near 3 kGy. For that purpose 400 kW electron accelerator with three separate irradiators was proposed as a source of ionizing radiation.

3.1. General Technological Scheme

Total technological scheme of the installation on of electron-beam treatment of TDIC wastewater is presented in Figure 3. It includes three principal technological chains: wastewater flow, cooling/ozonizing air flow, and ventilating air flow. Coordinated functioning of those chains is assured by monitoring and control systems.



FIG. 3. Simplified technological scheme of the plant. In the scheme (see Basic equipment): F1-F4 – Air fans, P1-P2 – Water pumps, D1 and D2 – Diffusers, A – Accelerator, R – Reactor, B1 and B2 – Primary and secondary basins.

The process of wastewater treatment consists of the following steps:

- collecting the inflow wastewater in primary (stock) basin;
- pumping the wastewater from primary basin to reactor;
- irradiating the wastewater inside reactor, cooling air being also irradiated;
- collecting irradiated wastewater in secondary basin;
- bubbling irradiated air (containing ozone) through the wastewater in a basin;
- pumping the wastewater from secondary basin to outlet line.

3.2. Wastewater Flow

Wastewater flow passes the following elements (in series): Inlet System – Primary Basin – Water Pump 1 (P1) – Nozzles – Reactor – Secondary Basin – Water Pump 2 (P2) – Outlet Line. All the steps of wastewater flow chain are correspondent to flow rate 420,000 kg/h (about 10,000 m^3 /day).

3.3. Air Flows

There are two separated air flows in technological scheme. Function of the first one (Air Flow I) is to supply cooling air to electron beam output window and to supply ozone formed inside reactor with the rate 3.6 kg/h to wastewater for additional ozone treatment, three variants of ozone treatment being proposed: in secondary basin, in primary basin, and in both of them. Flow rate is 5,760 kg/h.

The second flow (Air Flow II) is ventilating flow for generator and reactor rooms. Organizing air flows in such a way makes it possible to minimize total air flow rate and substantially reduce ozone content in exhaust air by using it for additional treatment of wastewater (ozonizing). Flow rate is 2,880 kg/h. Total air flow rate is 8,640 kg/h.

The pressure inside Reactor should be slightly lower than that in Reactor Room to prevent penetration of irradiated air into Reactor Room. Therefore, slight air flow (up to $0.014 \text{ m}^3/\text{s}$) from Reactor Room into Reactor can appear because of imperfect airtight of the Reactor.

3.4. Basic Equipment

Basic technological equipment used in the process is listed in Table 5.

TABLE V.BASIC TECHNOLOGICAL EQUIPMENT

No	Name	Characteristics
1	Electron Accelerator	1 MeV, 400 kW beam power. With 3 irradiators.
2	Reactor	Volume 6.5 m ³ . Air-tightened. Equipped with 12 nozzles.
3	Water Pump 1	Capacity 0.117 m ³ /s at pressure drop up to 4 kg/cm ² .
4	Water Pump 2	Capacity 0.117 m^3 /s at pressure drop up to 3 kg/cm^2 .
5	Air Fan 1	Capacity 1.33 m ³ /s at pressure drop up to 400 mm Hg.
6	Air Fan 2	Capacity 1.35 m ³ /s at pressure drop up to 460 mm Hg.
7	Air Fan 3	Capacity 1.35 m ³ /s at pressure drop up to 230 mm Hg.
8	Air Fan 4	Capacity 0.67 m^3 /s at pressure drop up to 100 mm Hg.
9	Diffusers	Two groups. Each one for total diffusion area 7 m^2 at porosity 20% or 5 m^2 at porosity 30%.

View of the reactor is presented in Figure 4. The walls of the reactor (its base) are made of concrete, interior surfaces being covered by 0.5-1.5 mm stainless steel. Over the walls on a steel onlay there are situated two hatches, six side shields, and two arches, which are connected with bottom part of irradiators and make the reactor air-tightened, preventing penetration of irradiated air to reactor room. The pressure inside reactor should be slightly lower than that in reactor room to ensure slight air flow from reactor room into reactor for the case of imperfections in reactor airtight. Bottom part of the reactor (collector) is deepened under the ground. It is connected by two channels, equipped with hydraulic hitches, with secondary basin for passing irradiated wastewater. Over collector, on the ground level, there is installed horizontal grid plate for mounting and repair work inside reactor.



FIG. 4. View of Reactor.

Table 6 contains data on electric energy consumption by all the equipment used in a technological process.

TABLE VI.	ELECTRIC ENERGY	CONSUMPTION	(MINIMAL)	/MAXIMAL)
			\	

No	Equipment	Energy consumption, kW
1	Electron Accelerator	600 / 800
2	Water Pumps	44 / 144
3	Air Fans	120 / 321
1.2.1. Total		764 / 1265

3.5. Monitoring and Control Systems

Control system includes controlling pressures, flow rates, levels of liquid in stock reservoirs, and electric engines conditions.

Pressure Monitoring. Along the wastewater flow pressure is measured before and after each of two water pumps P1 and P2. Air pressure along air flow I is measured after air fan F1 as well as before and after each of two air fans F2 and F3. Air pressure along air flow II is measured before air fan F4 that corresponds to the pressure in irradiation room.

Flow Rate Control. Flow rates of wastewater are controlled on inlet of the plant, i.e. before primary basin, before each branch of nozzles (to equalize flow rate distribution in reactor), and on the outlet of the plant, i.e. after water pump P2.

Level Control. The levels of wastewater in both primary and secondary basins are monitored by level gauges and controlled to be constant by changing appropriate flow rates.

Electric Engine Control. All electric engines of water pumps and air fans are standard controlled.

Each water pump or air fan unit (P1, P2, F1-F4) consists of two pumps or fans (supplied with input and output valves) connected in parallel and paralleled with bypass pipe that includes precision regulating valve.

4. BUILDING CONSTRUCTION

While projecting the building construction, following circumstances were taken into account:

sufficient fastness to carry heavy equipment; compact disposal of the equipment and minimizing pipelines combined with sufficient space for montage and repair works; sufficient radiation protection.

4.1. Basic Layout

The building consists of two floors: the ground and the first ones. Profile views of the building are presented in Figure 5. Following notations are used: 1 - Accelerator/Generator Room, 2 - Reactor Room, 3 - Reactor, 4 - Collector, 5 - Reactor Output Channels, 6 - Ceiling Window, 7 - Instruments Room, 8 - Montage Area 9 - Control Room, 10 - Safe Door (First Floor), 11 - Safe Door (Ground Floor), 12 - Primary Basin, 13 - Secondary Basin. Entrance to building is by staircase to montage area on the first floor. Ceiling window may be used for primary installation of accelerator units into the building as well as for repair needs.



FIG. 5. Profile views of the building.

4.2. Location of the Equipment

On the ground floor there are situated reactor room and two rooms for primary and secondary basins, the basins themselves being deepened under the ground. Following equipment is located on the ground floor (and under the ground level):

- Diffusers located on the bottom of primary and secondary basins.
- Reactor located inside reactor room, collector part of the reactor is deepened under the ground.
- Pumps and fans located outside building on special open site. It is possible (as a variant of building construction) to reduce area of primary and secondary basin rooms and release area 25.4 m² for location of pumps and some fans inside building.

On the first floor there are situated accelerator/generator room, control room, store&instruments room, and montage area. Following equipment is located on the first floor:

- Accelerator (including high voltage generator) located inside accelerator/generator room.
- Monitoring and control system located inside control room.

Besides, some fans may be located on the first floor inside store&instruments room.

4.3. Radiation Protection

Radiation protection from direct X-ray radiation is ensured by 1200 mm walls, made of monolithic dense concrete, around reactor room. Accelerator/generator room and secondary basin room, adjacent to reactor room, are equipped with shielded doors.

Additional means to ensure radiological safety include:

- monitoring of radiation level inside all the rooms of the building;
- automatics of opening and closing shielded doors;
- automatics to turn on and off ventilation system and cooling/ozonation system;
- electrical and mechanical blocking system;
- visual and sound signaling system;
- access control system.

5. CONCLUSIONS

On the base of the design performed following main characteristics of the demonstration plant on TDIC wastewater treatment with capacity $10,000 \text{ m}^3/\text{day}$ may be presented:

- a. Total area required is 220 m^2 . It consists of area under the building ($13.9 \times 13.6\text{m}$) plus area under communications and special site for pumps and fans location.
- b. The building consists of two floors, total height being 11 m. Under part of building there are deepenings: -3.5 m for basins, -1.7 m for collector of reactor; 390 m³ of dense concrete is required.
- c. Jet method is used for irradiation of wastewater by 1 MeV, 400 kW electron beam inside air-tightened reactor, irradiated air with ozone from reactor being used for additional ozone treatment of wastewater.

- d. Three variants of additional ozone treatment can be realized: I after EB-treatment; II before EB-treatment; III both before and after EB-treatment by dividing air/ozone flow.
- e. Total electric energy consumption is inside the range 764 (min) and 1265 (max) kW.
- f. Optimum utilization of demonstration facility:
- g. for regular treatment of TDIC wastewater;
- h. for investigation of influence of wastewater parameters on purification efficiency under real conditions;
- i. as training center for raising the level of operators' skill for other plants;
- j. for demonstration of electron-beam method and advanced approaches in the field of industrial dosimetry, radiation safety.

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INDUSTRIAL APPLICATIONS OF ELECTRON BEAM FLUE GAS TREATMENT

A.G. Chmielewski^a, A. Pawelec^b, B. Tymiński^b, Z. Zimek^b, J. Licki^c

^aInternational Atomic Energy Agency, Vienna ^bInstitute of Nuclear Chemistry and Technology, Warsaw, Poland ^cInstitute of Atomic Energy, Otwock-Świerk, Poland

Abstract. The electron beam flue gas treatment process is one of the most promising technologies in modern environmental protection. The technology allows for simultaneous removal of acidic pollutants such as SO_2 and NO_x with high efficiency and decomposition of VOC (volatile organic compounds) without generating any wastes. After the laboratory pilot plant experiments the industrial installation has been constructed at EPS Pomorzany in Szczecin, Poland. The installation treats flue gases from two boilers of 65MW_e each. The maximum flow of the gases is equal to 250,000 Nm³/h and the total beam power of accelerators exceeds 1MW that makes it the biggest radiation facility in the world.

1. INTRODUCTION

Fossil fuels combustion is one of the main sources of human origin air pollution. Such pollutants as SO_2 , NO_x , volatile organic compounds (VOC) and others are emitted in these processes. Different technologies have been applied for air pollution control e.g. coal and oil desulphurisation, combustion modification or flue gas purification. The most widely applied systems for coal fired boilers are wet flue gas desulphurisation (FGD) using lime or limestone as a reagent for SO_2 removal [1] and selective catalytic reduction (SCR) for NO_x reduction. VOCs are usually adsorbed on carbon, but this process is rarely used up to now.

All these technologies are complex chemical processes and waste, like wastewater, gypsum and used catalyst are generated. Electron beam technology is among the most promising advanced technologies of new generation. This is a dry-scrubbing process of simultaneous SO_2 and NO_x reduction, where no waste except the by-product is generated. The by-product is fully usable as a fertilizer. The process concept was developed by S. Machi and collaborators [2]. Also, as it was shown by our investigation, VOCs present in flue gas may be reduced in this process.

Two industrial installations using this technology have been constructed in the world, one in China and the second in Poland. The third one in Japan, which is under construction, is going to be put in operation in a short time. Chinese installation is mostly SO_2 removal oriented (there are no NO_x emission limits in China up to now); so Polish one is as a matter of fact the first installation for simultaneous desulphurisation and denitrification of flue gases. The plant located in EPS Pomorzany in Szczecin treats the flue gases emitted from two Benson boilers of 60 MW_e and 100 MW_{th} each. Flue gases of maximum flow of 250,000 Nm³/h are irradiated by four accelerators of 700 keV electron energy and 260 kW beam power each. With the total beam power over 1MW Polish installation is the biggest radiation facility in the world.

2. PROCESS DESCRIPTION

The electron beam flue gas treatment process is designed for simultaneous removal of sulphur and nitrogen oxides. SO_2 and NO are being oxidised and after the reaction with ammonia added to the process and create ammonium salts that are collected by electrostatic precipitator.

After humidification and lowering its temperature, flue gases are guided to reaction chamber, where irradiation by electron beam takes place. Ammonia is injected upstream the irradiation chamber. Fast electrons interact with gas creating various ions and radicals [3]. In the case of high water vapour concentration the oxidising radicals OH⁻ and HO₂⁻ and excited ions as O(³P) are the most important products.

There are known several pathways of NO oxidation. In the case of electron beam treatment the most common are as follows [4]:

$$NO + O(^{3}P) + M \rightarrow NO_{2} + M$$
$$O(^{3}P) + O_{2} + M \rightarrow O_{3} + M$$
$$NO + O_{3} + M \rightarrow NO_{2} + O_{2} + M$$
$$NO + HO_{2}^{-} + M \rightarrow NO_{2} + ^{-}OH + M$$

After the oxidation NO_2 is converted to nitric acid in the reaction with OH^{\circ} according to the reaction:

$$NO_2 + OH + M \rightarrow HNO_3 + M$$

HNO₃ aerosol reacts with NH₃ giving ammonium nitrate that can be written:

$$HNO_3 + NH_3 \rightarrow NH_4NO_3$$

Partly NO is reduced to atmospheric nitrogen [5].

There can be also several pathways of SO_2 oxidation depending on the conditions. In the case of electron beam treatment the most important are radiothermal and thermal reactions [4].

Radiothermal reactions proceed through radical oxidation of SO₂ in the reaction:

$$SO_2 + OH + M \rightarrow HSO_3 + M$$

then HSO₃ creates ammonium sulphate in the following steps:

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2$$
$$SO_2 + H_2O_2 \rightarrow H_2SO_2$$

$$SO_3 + H_2O \rightarrow H_2SO_4$$

$$H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2SO_4$$

The thermal reaction is based on the following process:

$$SO_2 + 2NH_3 \rightarrow (NH_3)_2SO_2$$

$$(NH_3)_2SO_2 \xrightarrow{O_2, H_2O} (NH_4)_2SO_4$$

The total yield of SO_2 removal consists of the yield of thermal and radiothermal reactions that can be written [6]:

$$\eta_{SO2} = \eta_1(\phi, T) + \eta_2(D, \alpha_{NH3}, T)$$

The yield of the thermal reaction depends on the temperature and humidity and decreases with the temperature increase. The yield of the radiothermal reaction depends on the dose, temperature and ammonia stoichiometry. The main parameter in NO_x removal is the dose. The rest of parameters play minor role in the process. Nevertheless in real, industrial process, dose distribution and gas flow conditions are important from the technological point of view.

In the case of VOCs decomposition the process itself is based on the similar principles as primary reactions concerning SO_2 and NO_x removal i.e. free radicals attack on organic compounds chains or rings causing VOCs decomposition.

For chlorinated aliphatic hydrocarbons' decomposition (e.g. chloroethylene), Cl⁻ dissociated secondary electron –attachment and Cl, OH radicals reaction with VOCs play very important roles for VOCs decomposition.

For aromatic hydrocarbons, VOCs decomposition will mainly go through:

1. Positive ions' charge transfer reactions

 M^+ + RH (RH=VOC, eg. Benzene or PAHs) = M + RH^+

Because RH has lower ionisation energy (IE) (Benzene: IE = 9.24 eV; PAHs: IE < 10 eV) than most primary positive ions (IE > 11 eV) formed above, part of VOC will be decomposed by rapid charge transfer reactions.

2. Radical – neutral particles reactions

^OH radical plays very important role for VOC decomposition, especially when water concentration is 10 %. ^OH radicals react with VOC in two ways:

'OH radical addition to the aromatic ring (eg. toluene)

 $OH + C_6H_5CH_3 = R1$

and H atom abstraction (for the alkyl-substituted aromatic compounds) or H atom elimination (for benzene, naphthalene and the higher polycyclic aromatic hydrocarbons)

 $C_6H_5CH_3 + OH = R2 + H_2O$ (H atom abstraction)

 $C_6H_6 + OH = C_6H_5OH + H$ (H atom elimination)

Radicals (R1^{\cdot}, R2^{\cdot}) formed above go though very complex reactions: O₂ addition, O atom release, aromatic –CHO (-dehydes), -OH (-ol) compounds formed or ring cleavage products:

R[°] (R1[°], R2[°]) + O₂ = RO₂[°] 2 RO₂[°] = 2RO[°] + O₂ RO₂[°] + NO = RO[°] + NO₂ RO[°] + O₂ = HO₂[°] + products (aromatic-CHO, -OH) RO[°] → aliphatic products

3. INDUSTRIAL INSTALLATION

The flue gas treatment industrial installation was located in EPS Pomorzany in Szczecin in the north of Poland [7]. The installation purifies flue gases from two Benson boilers of 65 MW_e and 100 MW_{th} each. The maximum flow rate of the gases is 250,000 Nm³/h and the total beam power exceeds 1MW. The installation consists of four main parts:

- flue gas conditioning unit
- ammonia storage and dosage unit
- reaction chambers
- by-product collecting and storage unit.

The computer control and monitoring device was applied (Fig. 1). The scheme of the electron beam flue gas treatment installation is given in Fig. 2.



FIG. 1. Control room at EPS Pomorzany.



FIG. 2. The scheme of industrial plant at EPS Pomorzany.



FIG. 3. Gas cooling tower.

Flue gas stream after electrostatic precipitators is divided into two streams. About half of the total flue gas amount is leaded to the installation while the rest is bypassed to the stack. The part of gas being purified enters a dry bottom spray cooler (fig. 3). In the process of water evaporation the temperature lowered to $65 - 80^{\circ}$ C and humidity rises up to 10 - 14%. The process water is dosed by a system of nozzles so the complete evaporation of droplets takes place. If the humidity is to low, there is also the possibility of adding the steam over the gas outlet from the cooling tower.

Ammonia, which is the main reagent adding in the process, is stored in the ammonia water form and may be dosed in two ways:

- as gaseous ammonia, which is evaporated from ammonia water and then injected upstream to the irradiation chambers (variant I)
- as ammonia water straight to spray cooler using separate system of nozzles (variant II).

There is also the possibility of adding ammonia both in the form of ammonia water and gaseous ammonia (so called mixed variant). The tests carried on the industrial installation showed, that the way of ammonia adding is important especially from the SO_2 removal point of view. The ammonia water consumption is in the range of 150 to 600 kg/h depending on the process conditions.



FIG. 4. Reaction chamber.



FIG. 5. Accelerator.

Formed by-product particles are of the diameter about 1 μ m and sticky. The by-product is collected by the electrostatic precipitator with a flat heated bottom furnished with a scrapping device and after storage is shipped to NPK fertilizers production plant. The by-product yield is up to 300 kg/h. The gas after purification is mixed with the unpurified stream of gases and leaded to stack. The gas temperature after mixing exceeds 110°C that avoids wet stack problems.

4. OPERATIONAL EXPERIENCES

As it was previously mentioned the removal efficiency depends strongly in the process conditions. The highest obtained efficiency for SO_2 reaches 95%, while for NO_x it reaches 70% (figure 6). The obtained results may be compared with previously reported, based on the pilot plant experiments and theoretical calculations, presented in Fig. 7 [8]. The very good agreement between these two figures may be noticed.



FIG. 6. SO_2 and NO_x removal efficiency vs. dose. The results obtained at the industrial installation. (SO_2 inlet conc. $1500 - 1630 \text{ mg/Nm}^3$, NO_x inlet conc. $470 - 540 \text{ mg/Nm}^3$).



FIG. 7. SO_2 and NO_x removal efficiency vs. dose. The results obtained by the pilot plant experiments and theoretical calculations [8].

The data obtained during the operation of the installation confirmed the previously taken theses on the impact of the process parameters on the removal effectiveness. In the case of NO_x removal the most important parameter is the dose. The inlet concentration of NO_x is the second parameter, that impact on this pollutant removal was observed. The correlation is linear and total removal (taken in mg/Nm³) increases with the inlet concentration of NO_x , while the relative removal (in %) decreases with the parameter increase. The ammonia stoichiometry factor has a very little impact on the NO_x removal (figure 8).



FIG. 8. SO_2 and NO_x removal efficiency vs. ammonia stoichiometry. The results obtained at the industrial installation.

In the case of SO_2 removal there is more parameters that impact on the removal efficiency has been observed. The most important parameter is temperature of the gases after the humidification because of the thermal reactions pathway. Afterwards the dose should be mentioned. Although the humidity seems to have the major impact on the process efficiency, it is hard to prove it with no doubt because of the strong correlation between the humidity and the temperature of the process. The temperature lowers during the water evaporation process and lowering the temperature involves the humidity increase. Unexpectedly high impact of ammonia stoichiometry factor on the SO_2 removal efficiency has been observed (figure 4). The other factors as flue gas flow rate and inlet concentration has much less impact on the removal efficiency. During the experiments one more parameter having impact on the whole process has been detected: the ammonia dosing way. It was observed, that the injection of part of ammonia water straight to humidification tower increases the SO_2 removal efficiency. This phenomenon is under research now.

The plant was operated for over 3000 hours with two accelerators and than, since June 2002, with set of all four accelerators for over 2500 hours.

In parallel using laboratory unit and pilot plant [9], tests concerning PAH removal were performed. On the basis of the industrial scale operational experience and laboratory/pilot tests results, feasibility study for the installation for PAH eb treatment is prepared in the frame of EU INCO EBOGEM project. The purpose is possible application in the metallurgical industry, where beside of SOx and NOx, harmful organic pollutants are emitted. This opens opportunity of the process application for the air pollution control at municipal waste incineration, in which case beside of the acidic pollutants, PAH like dioxins are emitted.

5. CONCLUSIONS

The first electron beam flue gas treatment installation for simultaneous SO_2 and NO_x removal has been constructed in EPS Pomorzany in Szczecin, Poland. It is also the biggest radiation facility in the world. Among the other advantages the most important are:

- simultaneous removal of SO₂, NO_x and VOC
- high effectiveness of the process
- dry process
- no wastes generation with agricultural use of by-product.

The described method is easy to realisation, so the installation may be used in most plants, where multipollutant control is essential.

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IRRADIATION ON A NEW SCALE: INTRODUCING BREVION

R.M. Brinston, D.G. Levesque

MDS Nordion, Ottawa, Ontario, Canada

Abstract. Since its very beginnings, the gamma industry has been challenged to develop an economical system for processing products in low volumes. MDS Nordion has solved that longstanding problem with *Brevion*: an innovative, small-scale irradiator designed specifically for operations with annual product-processing volumes of up to 20,000 m3 per year. Achieving good cobalt efficiency and dose uniformity, *Brevion* delivers the performance of a medium-sized facility at a lower capital cost. It is the world's only batch irradiator capable of cartridge changeover in less than five minutes, an advantage that increases cobalt utilization and overall process throughput. Lead-time for construction is considerably shorter, resulting in significantly lower start-up costs.

1. INTRODUCTION

A truly effective irradiation system must offer both technological advantages and demonstrable business benefits, delivering the greatest possible performance economically and efficiently. It was with these criteria in mind that MDS Nordion engineering developed its unique small-scale "cartridge" irradiator, the *Brevion* (Figure 1). Designed for companies with annual processing requirements of 20,000 m3 per year or less, *Brevion* is versatile, flexible and efficient.



FIG. 1. Brevion small scale "cartridge" irradiator.

Representing a low-cost alternative to ethylene oxide sterilization, *Brevion* makes in-house gamma processing both affordable and practical. It is ideal for small-volume manufacturers and for contract irradiation service providers serving growing markets in developing countries. And it's well-suited to a wide range of applications, from medical devices and pharmaceuticals to cosmetics, packaging materials, foods and dried ingredients.

2. BREVION: THE SYSTEM AT A GLANCE

Delivering the performance of a typical medium-scale automatic tote facility at a substantially reduced cost, *Brevion* offers a number of benefits, including:

- 1- Low capital cost
- 2- Short project lead time
- 3- Economical processing of small product volumes

- 4- Flexible processing of products with different densities—enabling efficient batch processing, rapid product changeovers and reducing product hold-ups
- 5- Minimal direct labour requirements for operation
- 6- Easy and quick dismantling to permit relocation of facility

A reliable PLC (programmable logic controller) with redundant safety-interlock features monitors *Brevion's* performance throughout its entire cycling process. This computer-control process is fully validated.

Perhaps the most critical performance characteristic of *Brevion* is its capacity for very rapid batch changeovers. In fact, it is the only system in the world capable of batch changes in less than five minutes—which translates into a considerable competitive advantage for processors.

Brevion performance advantages:

- Rapid cartridge exchange: under five minutes
- 1 MCi (37 PBq) cobalt rack capacity
- Up to 0.4 g/cm3 (maximum tote load 88 kg) product-density range
- Low dose-uniformity ratio (DUR)
- Small total footprint: less than 31 m x 18 m—no interior maze
- Small shield footprint: 12 m x 8 m
- Short installation time: less than four weeks onsite

3. UNIQUE DESIGN FEATURES

Most often, batch irradiators provide good dose uniformity but poor throughput, due to the downtime required for batch change outs. *Brevion* corrects this imbalance by employing a double-cartridge system that permits one cartridge to be processed while the other is loaded or unloaded. Each simple, modular cartridge is free of any drive mechanisms; this contributes to the simplicity of the system as a whole and improves its reliability.

Brevion's unique door design (Figure 2) removes the need for a maze within the system reducing its footprint significantly—and helps accelerate the cartridge-exchange process. Fitting snugly into the irradiator like a plug in a socket, the shielded door slides in and out of position with the source pass mechanism in tow. The irradiated cartridge is removed and the un-irradiated cartridge is loaded into position. After the exchange, the source pass retracts into the chamber and the door slides shut behind it.

Brevion provides an option of two tote sizes; processors can choose whichever is best suited to the dimensions of their products.



FIG. 2. Brevion's unique door design.

Because *Brevion* is compact and modular, it can be built within a few short months and installed in a matter of weeks. Components are shop-tested and optimized prior to shipping, and transported to the construction site partially assembled.

The characteristics that contribute to *Brevion's* ease of construction also make is possible for the system to be disassembled and relocated—giving processors the freedom to follow their markets, remain close to distribution centres, and maintain access to transportation routes. The cost of building a new shield at a relocation site is fairly low due to the small footprint and simple rectangular layout of that component.

4. BUILT FOR EFFICIENCY

Brevion is designed to use MDS Nordion's worldwide-standard C-188 cobalt-60 sources. To achieve maximum cobalt efficiency, totes are stacked two-high, creating product-overlap geometry. The space that would be required customarily for a second-level roller conveyor is consequently eliminated. Stacked totes circulate once around the source pass, and then are interchanged vertically. After the totes circulate a second time, they are removed. Because the vertical exchange occurs automatically inside the cell, total batch cycle time is reduced substantially.

Brevion demands less source-pass maintenance than a traditional two-level tote system due to the fact that it has fewer moving components and just a single level of drives. Conveniently, maintenance can be performed on each cartridge outside the shield; while one is being serviced, the other can remain in use.

Again, because *Brevion* employs a double-cartridge system, one batch of products can be loaded and unloaded by the operator while another is being irradiated. To accelerate batch interchange, loading and unloading occurs at the same station; the two activities are kept separate by a 10-tote tilt station. The operator indexes each cartridge row until all processed totes have been unloaded and then loaded with product for processing. Once the entire cartridge has been reloaded, it returns to the interchange position and waits for the current batch to be completed. Using this procedure, *Brevion* makes it possible for operators to process a significant number of batches per year (Figure 3).



Total Batch Processing Time (25 kGy Dmin with 1 MCi Cobalt-60 installed)

FIG. 3. Total batch processing time.

5. CONCLUSIONS

At roughly one-third of the cost of a conventional irradiator, *Brevion* provides an extremely economical alternative for low-volume processors. Able to process products of different densities, and with a total capacity of one million curies, *Brevion* opens the door to numerous applications—both existing and emerging. Designed to enable rapid cartridge change outs and flexible product changeover, *Brevion* presents not only a cost-effective solution but also a highly competitive one. Companies with low-volume product lines can now enjoy the control—and economic advantage—afforded by in-house processing.

LIST OF PARTICIPANTS

Berejka, A.J.	Ionicorp, 4 Watch Way, Huntington, New York 11743, United States of America
Brinston, R.M.	MDS Nordion, Ottawa, Ontario, Canada
Chmielewski, A.G.	International Atomic Energy Agency
Jongen, Y.	Ion Beam Applications S.A. Louvain-la-Neuve, Belgium
Khairul Zaman, Hj. M.D.	Radiation Processing Technology Division, Malaysian Institute for Nuclear Technology Research (MINT), Bangi, 43000 Kajang, Malaysia.
Kovács, A.	Institute of Isotope and Surface Chemistry, Chemical Research Center,
	Hungarian Academy of Sciences, Budapest, Hungary
Kuntz, F.	AERIAL, 19 Rue de Saint Junien, 67305 Schiltigheim, France
Machi, S.	Japan Atomic Industrial Forum, Inc.
Makarov, I.E.	Institute of Physical Chemistry Russian Academy of Sciences, Moscow, Russian Federation
Mittendorfer, J.	High Tech Consulting, Anton Brucknerstr. 6, A-4600 Wels, Austria
Rocquigny, H.	COFRAR et RADIENT, Versailles, France
Sarma, K.S.S.	Radiation Technology Development Section, Bhabha Atomic Research Centre, Trombay, Mumbai, India
Yousri, R.M.	National Centre for Radiation Research and Technology, AEA, P. O. Box 29, Nasr City, Cairo, Egypt
Zimek, Z.	Department of Radiation Chemistry and Technology, Institute of Nuclear Chemistry and Technology, Warsaw, Poland
Zyball, A.	BGS Beta-Gamma-Service GmbH & Co. K, Wiehl, Bruchsal, Saal a.d. Donau, Germany