ISSN 1977-5296

Number 38
June 2008

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Manuscripts are to be sent to the Editor following the 'instructions for authors' available on the 'Bulletin' page of the ESARDA website. Photos or diagrams should be of high quality.

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ESARDA Bulletin is published jointly by ESARDA and the Joint Research Centre of the European Commission.
It is distributed free of charge.

The publication is authorized by ESARDA.

Cover designed by
N. Bähr
EC, JRC, Ispra, Italy

Printed by
Ragusa Grafica Moderna srl – Bari, Italy

ESARDA
European Safeguards Research and Development Association

Bulletin

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Editorial

A word from the ESARDA Editorial Committee

B. Autrusson, Chairman

More and more, ESARDA is considered as a European forum / think tank for safeguards, non proliferation and related fields. This makes effective communication one of the most important roles for ESARDA. For this reason, an Editorial Committee (Working Group) was established, to be responsible for the following communication activities:

- the editorial policy of ESARDA (subject to approval of the Executive Board);
- preparation and publication of the ESARDA Bulletin;
- maintenance of the ESARDA Website (to include WG annual reports, Technical Sheets and updates, etc.);
- provision of a Scientific / Organising committee, to organise the ESARDA Annual Meetings (internal or symposia), including the selection of papers, programme elaboration and feed back; and
- organisation or support to other meetings such as Topical Meetings, joint ESARDA-INMM, etc.

The 2007 Symposium, the largest ever event of ESARDA, was hosted by IRSN in Aix en Provence, France. Between 22 and 24 May 2007, the meeting attracted 260 participants to three parallel sessions dealing with technical, policy and review papers including, for the first time, sessions on dual use, communication and training. Around 150 papers were presented in a total of 30 sessions, whilst the opportunity was also provided for participants to study 23 posters and discuss them with their authors. The proceedings of the ESARDA 2007 Symposium were distributed to participants in September 2007.

Looking ahead, the 2009 Symposium will be held in Lithuania and hosted by VATESI, providing ESARDA with the opportunity to honor a recent member of the European Union.

The objectives of the ESARDA Bulletin, first published in October 1981, have been maintained and developed by the Editorial Committee. The Bulletin is now published at least twice a year, and includes a recently-introduced section for peer-reviewed papers. During 2007, three issues of the Bulletin were published, including the following new sections:

- N° 35 – January 2007 – some news from ESARDA members
- N° 36 – July 2007
  - Summary report of ESARDA working group activities
  - Tribune and opinions of individual members, with papers from DG/TREN and the president of the Swiss Nuclear Forum
- N° 37 – December 2007 – new technical sheets prepared by the working groups

The Bulletin editor can be contacted at the newly established email address: **esarda-bulletin@jrc.it**.

The internet website is considered as a live publication of ESARDA, updated regularly by the Secretary. During 2007, the new version of ESARDA course material, the edited Bulletins, technical sheets and all other information provided by the working groups were added to the website. The website also presents valuable information on safeguards concepts, methods and equipment to those who may be unfamiliar with safeguards, and includes the texts of founding treaties, national laws and a glossary.

The website also includes sections providing news and notification of annual meetings, conferences and symposia. Indeed, the website provides the primary means for communication on such meetings

and will include essential practical and organizational information. ESARDA members are encouraged to visit the website regularly.

The website is also the collective memory of what has been done. It is a data base of all material ever published by ESARDA, and the Editorial committee has the ambition that the website's "Library" will become a reference centre of data in the field of nuclear safeguards.

Of course, the Editorial Committee can only work with the information provided by ESARDA members. The Editorial Committee appreciates greatly the time spent by those who have contributed to the success of ESARDA publications over the last year, and would encourage everyone to maintain an active involvement. The Editorial Committee need your feed back, and any suggestion is welcome.

ESARDA News

Safeguards in Norway – Experiences with Integrated Safeguards

Tonje Sekse
Norwegian Radiation Protection Authority

Abstract

Norway was among the first countries to have a nuclear research reactor in operation. JEEP I, a joint project between the Netherlands and Norway, went into operation in 1951.

In the early years of nuclear power there were no safeguards bureau. However, Norway had in the late 1950's signed an agreement with USA concerning peaceful uses of nuclear energy. The agreement secured USA that the nuclear facilities in Norway were not used for military purposes, and provided Norway with uranium and other vital equipment for the reactors.

IAEA safeguards was implemented in Norway 1 March 1972. Norway is not part of EURATOM, thus only the bilateral safeguards agreement with the IAEA is enforced.

For Norway the Additional Protocol came into force in 2000, while Integrated Safeguards was implemented in 2002. As one of the first countries to have Integrated Safeguards implemented, Norway has a broad experience in this field. This paper will present the nuclear facilities in Norway and discuss the experiences with Integrated Safeguards and compare it to traditional safeguards.

1. Nuclear facilities in Norway in the past

Norway established in 1934 a plant for the production of heavy water in the small industrial centre of Rjukan. The heavy water plant was in operation until 1988, when it was permanently shut down. The plant was of particular interest to the Germans during the World War II. Despite the efforts from the Germans to maximize production and then transport the heavy water to Germany, the heavy water never reached Germany. The Norwegian resistance managed to sabotage the plant in 1943 and the transport in 1944. Today 40 barrels containing heavy water is still resting on the bottom of Tinnsjøen, 400 m below the surface.

The Institute for Atomic Energy (IFA) was established in 1948. In 1951 Norway was the sixth country in the world to have a research reactor in operation. Only the Nuclear Weapon States (minus China) and Canada were earlier. The reactor, JEEP I, was a

Figure 1: The Halden boiling heavy water reactor. (photo: IFE)

joint project between the Netherlands and Norway. Yellow cake from the Netherlands, hidden away in Utrecht during the war, was exchanged with pins of metallic uranium from the United Kingdom. The metallic uranium pins were used as fuel. The capacity of the reactor was 0,2 MW and it was located at Kjeller, just 20 km north-east of Oslo.

In 1959 the second research reactor came into operation in Norway. The reactor was a 25 MW boiling heavy water reactor aimed at research on natural uranium with heavy water power production. It is located in Halden, 130 km south of Oslo and close to the Swedish border. Figure 1 shows the reactor hall at present.

There was high activity in the nuclear field in Norway in the 1960's. A new research reactor, NORA, and a small uranium reprocessing plant came into operation at Kjeller in 1961. NORA was a 0 MW research reactor built for experiments in reactor physics. The IAEA performed its very first safeguard inspection at NORA in 1962.

The last reactor built in Norway was JEEP II, which came into operation in 1966. JEEP II is a 2 MW pool research reactor located at Kjeller. JEEP II was built to keep up the demand for isotopes for medical and technical purposes, and improve studies in neutron physics.

Since the 1980's the discussion concerning nuclear power in Norway has been non existent. Why is that? The Three Mile Island and Chernobyl accidents did certainly not boost the interest for nuclear power, but Norway is also a country rich in natural energy resources like hydroelectric power, oil and gas.

2. Status of nuclear facilities in Norway today

Today there are two research reators in operation in Norway. They are both owned and operated by the Institute for Energy Technology (IFE), former IFA. Figure 2 indicate the localization of the research reactors.

The Halden boiling water reactor (HBWR) is an international OECD project that contributes to improve the safety at nuclear plants around the world through investigation on material science and high burn-up fuel performance. JEEP II produces isotopes for medical and technical purposes. JEEP II is also used for neutron transmutation doping of silicon and research in neutron and materials physics.

Both JEEP I and NORA have been decommisioned. JEEP I was decommisioned in 1969, and NORA in 1979 although operation for both ceased in 1967-1968. The uranium reprocessing plant was shut down in 1968 and finally decommisioned in 1995.

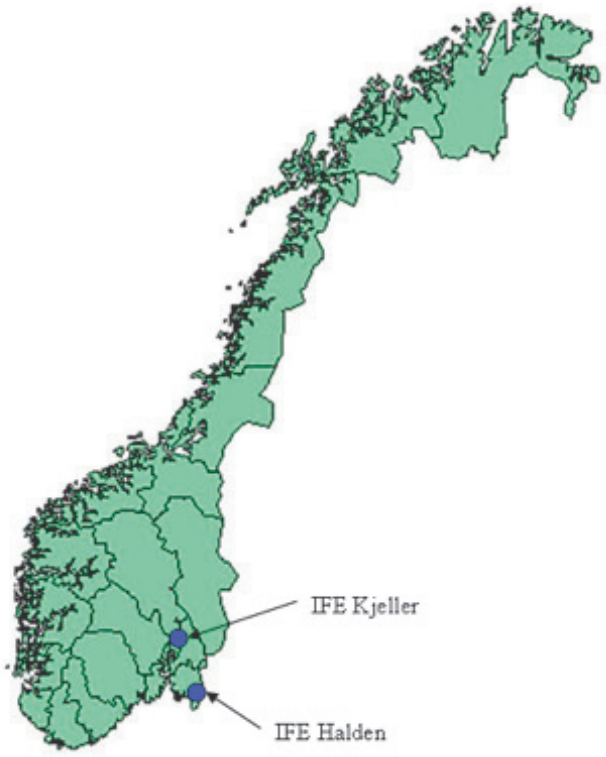

Figure 2: Localization of the present nuclear facilities in Norway.

The heavy water plant at Vemork, Rjukan, was permanently shut down in 1988. Today this area is a museum, the Norwegian Museum for Industry Workers.

Most of the nuclear material in Norway is located at IFE at Kjeller and in Halden. There are also small amounts of nuclear material in other parts of the country, generally consisting of depleted uranium being used for shielding.

3. Material balance areas

Four material balance areas are established in Norway. The JEEP II reactor at Kjeller is material balance area NOA-. NOB- is the rest of the Kjeller area, including among other a pellet production plant, metallurgical laboratory, and storages for fresh and spent fuel. The Halden reactor with facilities like storages for fresh and spent fuel and an instrumentation workshop is organized in NOC-. NOD- is the rest of Norway, and includes a lot of small Locations Outside Facilieties (LOFs). The LOFs are spread all over Norway. Most of these LOFs are industrial radiography companies which uses depleted uranium as shielding. In addition there are some small amounts of nuclear material at the University

of Oslo, the Norwegian Radiation Protection Authority and the Norwegian Museum for Industry Workers at Rjukan.

4. Norwegian legal framework

Norway has two main laws that apply to nuclear energy and radiation protection. *'Act on Nuclear Energy Activities 12 May 1972'* regulates the licensing regime, general requirements for licences, inspection regime and the legal basis for the regulatory body.

'Act on Radiation Protection and Use of Radiation 12 May 2000' constitutes the legal basis for regulating the use of ionising and non-ionising radiation, radiation protection requirements, medical use of radiation, contingency planning, waste management and discharges to the environment.

There are two main regulations issued pursuant to the Act on Nuclear Energy Activities;

'Regulations 2 November 1984 on the Physical Protection of Nuclear Material' and '*Regulations 12 May 2000 on Possession, Transfer and Transportation of Nuclear Material and Dual-use Equipment'*.

According to the act, there shall be an accounting system for nuclear materials in Norway. IAEA safeguards replaced the enforcement of the agreement between Norway and USA when it was implemented in Norway 1 March 1972. As Norway is not part of EURATOM, only the bilateral safeguards agreement with the IAEA is actively enforced. However, the agreement with USA is still valid.

The regulations on Possession, Transfer and Transportation of Nuclear Material and Dual-use Equipment was issued to facilitate the Additional Protocol coming into force.

From 1969 IFA was responsible for safeguarding the nuclear material in Norway through an agreement with the Ministry of Foreign Affairs. This arrangement was terminated in June 2004, and the responsibility for safeguards, State System Accountancy, was transferred to the NRPA.

5. Experiences with the implementation of the Additional Protocol and Integrated Safeguards

For Norway the Additional Protocol (AP) came into force 16 May 2000. The implementation of the AP caused some major changes in the inspection regime. Unannounced inspections and complementary accesses were introduced. Integrated Safeguards (IS) was implemented in Norway in 2002. Norway was at the time one of few countries in the world to have implemented this regime fully.

With the implementation of IS the combination of the traditional safeguards regime and the AP should be optimized such that both the economic costs and effective supervision of a country's nuclear holdings are optimal.

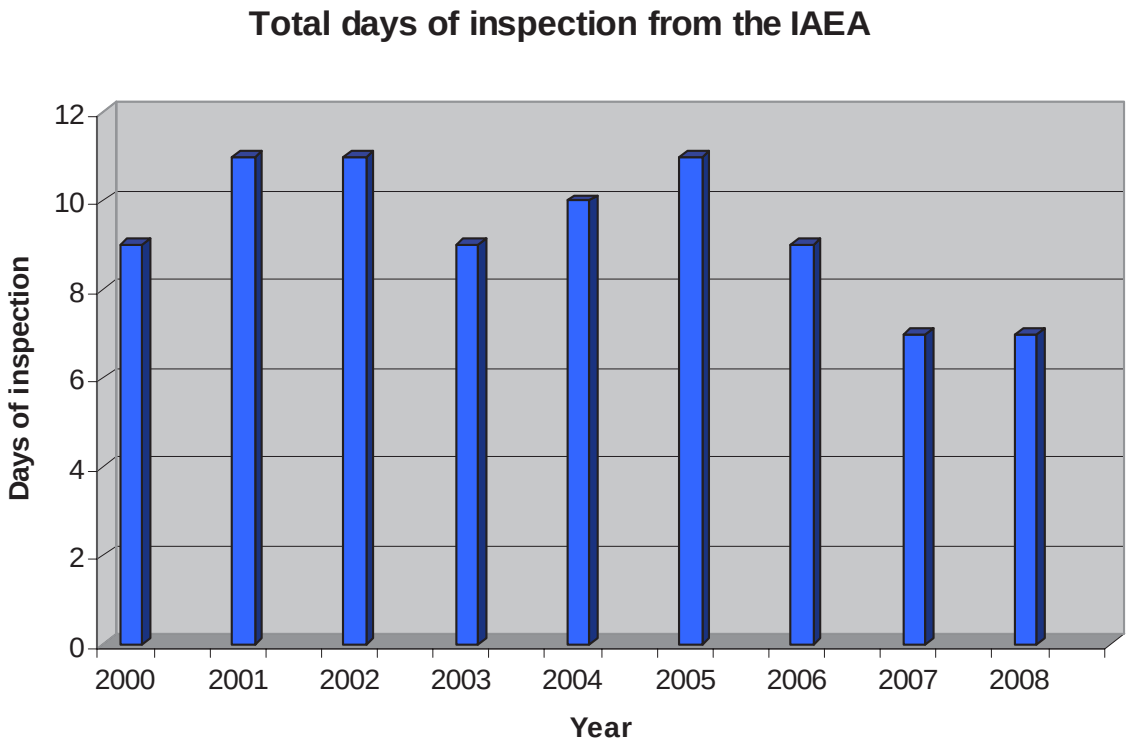

Figure 3: Total days of inspection from the IAEA per year.

The traditional IAEA regime for inspecting Norway consisted in performing physical inventory verifications at each inspection. With this regime the Halden site, NOC-, was inspected 4 times per year. The Kjeller site, NOA- and NOB-, were inspected 3 times per year. As for NOD-, this material balance area was inspected roughly every 3rd year.

With the implementation of AP and IS, the IAEA annually performs 2-3 unannounced inspections to Norwegian facilities, and these may include complementary accesses. There is one physical inventory verification per year at NOB- and NOC-. At NOA- there is at least one physical inventory verification every 4th year, as at NOD-. Concerning the unannounced inspections, there is normally one per year at NOB- and two per year at NOC-.

6. Integrated safeguards – does it work?

If we look at how much time the IAEA has spent inspecting Norway over the last years, we can detect a small reduction the last couple of years. Figure 3 illustrates the total inspection days the IAEA has spent inspecting facilities in Norway in one year from starting and ending with the physical inventory verification.

With the traditional safeguards regime the IAEA spent between 9 and 11 days every year. With IS fully implemented the number of days stayed around the same for the first years, but has been reduced to 7 days for the last two years. It remains to see if this trend will continue.

Before IS was implemented the IAEA and Norway had made some directions concerning the scope of unannounced inspections. Taking the thermal output from the Norwegian reactors into account the timeliness requires to verify spent fuel annualy only. It should then be sufficient to perform physical inventory verifications at the main inspection. In 2007 the IAEA requested to verify spent fuel during unannounced inspections. Norway does not understand the need for this, but such verifications have taken place. It seems to us as if the IAEA is moving back towards the traditional safeguards regime. If that is right, what is then the point of IS? Although the implementation of IS has reduced the number of days spent on Norwegian facilities, at least we knew when to expect the IAEA with the traditional safeguards regime. This is something that would be very interesting to further adress in the IS working group.

With the implementation of AP and IS Norway is also required to report all small LOFs. There are a lot of LOFs in Norway. Most of them hold depleted uranium. From my personal point of view it is a bit odd to neatly count every kg of depleted uranium in Norwegian shielding containers, when depleted uranium sometimes is dispersed all over the battlefields where anyone can pick it up.

Application of Safeguards in Switzerland

Bärbel Leibrecht
Swiss Federal Office of Energy SFOE

Abstract

In Switzerland the responsible body concerning nuclear material is the Swiss Federal Office of Energy SFOE. The SFOE is enacted as the responsible body for Safeguards since about three decades. The strengthening of safeguards has been stipulated also with the engagement and contribution of Switzerland.

This paper is aimed to give a brief overview of the nuclear environment and of the safeguards activities in Switzerland.

1. Introduction

Switzerland signed the Comprehensive Safeguards Agreement with the International Atomic Energy Agency IAEA in the late 1960's and put it in force on September 1978. Since the very beginning the State System of Accounting for and Control of nuclear material SSAC was entrusted to the Swiss Federal Office of Energy SFOE which is responsible for all questions concerning energy policy, use and supply. The Additional Protocol was signed in 2000 and ratified on February 2005. The Swiss Federal Office of Energy acted as the leading federal body for implementation, execution and supervision of this Agreement.

Although Switzerland is a small country, its the nuclear environment is quite manifold: five nuclear power reactors, three research reactors, two intermediate storage installations, one national research centre and one international research centre all are in operation. Moreover, new duties had to be assumed and several decommissioned nuclear facilities and locations require more concentration, since the Additional Protocol went into force.

Beyond this, the Swiss SSAC fulfills the following tasks: participation in elaboration of laws and ordinances, formulation of directives and guidelines, involvement in export control subjects, granting of transport authorization and co-operation during negotiation of bilateral agreements with other states.

2. Federal Administration

Switzerland has a unique democratic system. The Government is composed of seven councilors, each of them being head of a separate Department (Ministry). The Federal Department of Environment, Transport, Energy and Communications DETEC is responsible for providing vital public services like transport routes, communications and electricity networks. Every Department has various Federal Offices with expertise for solving the different technical questions. The Swiss Federal Office of Energy SFOE is providing its expertise on the field of energy and is the entrusted body for the fulfilling of obligations resulting from the Non Proliferation Treaty, the Comprehensive Safeguards Agreement and the Additional Protocol.

Several reorganizations were made inside of the SFOE since the starting of the accountancy and control of nuclear material. They were reflecting modification of the main objectives. During the elaboration of nuclear acts and ordinances SSAC was embedded in the Division of Legal Services. With the increasing activities in international and bilateral relations, Safeguards became part of the International Energy Affairs Section belonging to the Division of Energy Economy.

Until the end of 2007 safeguards and nuclear security were grouped under the same Division of Legal Services of SFOE. By decision of the Parliament, and complying with long-standing recommendations by the IAEA and the NEA, a new independent Nuclear Safety Inspectorate will start operations in 2009 – until now the Nuclear Inspectorate has been a unit of SFOE. In the same move, safety and security will be transferred to the newly founded Nuclear Safety Inspectorate.

3. Nuclear Environment

Nuclear Reactors			
Reactor	**Type**	**Power MWe**	**In Operation**
Beznau-1	PWR	365	1969
Beznau-2	PWR	365	1971
Mühleberg	BWR	355	1971
Gösgen	PWR	970	1979
Leibstadt	BWR	1165	1984

Switzerland has five power reactors as mentioned above at four different locations.

Every reactor is a separate Material Balance Area, with the common four Key Measuring Points KMP's: fresh fuel, core and spent fuel and others. At Gösgen an additional KMP is the separate wet storage pond building for spent fuel which just went into operation.

At the very beginning of the accountancy, SFOE prepared the reports for all nuclear installations; then the operators took over their obligations, which is the major part of reporting commitment. Now SSAC processes the reports into the state's accounting system and onto IAEA.

Research Reactors

Reactor	Type	Power	Location
Proteus	Possibility for irradiation of fuel elements	0-power	Research Centre Paul Scherrer Institut PSI Würenlingen
Crocus	Self-construction	0-power	Swiss Federal Institute of Technology EPFL Lausanne
AGN 211-P	Swimming-pool	0-power	University of Basel

There are three research reactors with little output.

Proteus will be reconstructed for new research projects.

The other two reactors have no recharges of fuel assembly due to the small capacity.

Due to the little content of nuclear material Physical Inventory Verification PIV is the sole inspection per year. Reporting was and is carried out by SSAC.

Research Centers

National Research Centre
Paul Scherrer Institute PSI,Würenlingen

Facility	Type	Location
Reactor Proteus	Possibility for irradiation of fuel elements	PSI
Vault Saphir	Storage of un-irradiated nuclear material	PSI decommissioned reactor
Hot Labor	Laboratories, hot cells, storage for irradiated samples	PSI

International Research Centre
CERN, Geneva

Facility	Type	Location
Building 607	Storage for out-of-use nuclear material components before shipment	CERN

Switzerland has one research centre of its own and hosts together with France the International Research Centre CERN.

The vault Saphir is inspected every month whereas the Hot Labor twice a year. CERN is inspected only once a year.

During the fist two decades the SSAC carried out reporting for the national research centre, since few years the three sites have assumed their obligations. Reporting of CERN – nuclear material is on the Swiss side only – is carried out by the Swiss SSAC.

Intermediate Storage Facilities

Facility	Type	Capacity	Location
Zwilag	Storage for spent fuel and HLW 1 Hot cell	200 casks	Würenlingen
Zwibez	Storage for spent fuel	36 casks	Beznau

Two intermediate storage facilities are in operation. Zwilag is authorized for storage of all kinds of nuclear and radioactive material. The hot cell is foreseen for the loading of spent fuel from one cask to another e. g. in case sealing system of the lid is losing tightening function. At the moment it is used for transshipping of spent fuel from a small shuttle into a big storage cask, as the big cask can not be loaded in the nuclear power plant directly.

Zwibez is the storage for spent fuel casks from the two Beznau reactors only and has just started operation.

Accounting records of Zwilag were prepared for some years by Zwibez and has now been handed over to the operator, while Zwibez is now preparing reporting for their own storage facility.

4. Remote Monitoring System

The Remote Monitoring System RMS does not record the pictures of the surveillance cameras on a chip, but sends them directly to IAEA headquarters in Vienna. Three quarterly inspections are omitted but one or two unannounced inspections are carried out instead during one calendar year. Reporting obligations increase: inventory, general ledger and operational report have to be sent every month instead of quarterly frequency with the standard surveillance system.

Switzerland has been since the middle of the 1990's the test bed for the RMS; both systems – standard surveillance system and RMS – running in parallel. Mühleberg was the first reactor ever officially oper-

ating with RMS since 2001. In the following year the other four nuclear reactors were switched solely to the new system while standard surveillance system was removed.

In two other facilities, Vault Saphir and Zwilag, the RMS is installed, but neither have the inspection intervals nor the reporting frequency changed.

The IAEA informed us recently, that only two failures occurred requiring the re-verification of spent fuel pond. Disturbances, which demand remedial action, are still quite common.

5. The way towards Integrated Safeguards

The Comprehensive Safeguards Agreement and the Additional Protocol provide the direction towards Integrated Safeguards.

In the middle of 2005, the SSAC provided the Initial Declaration of Switzerland pursuant to the Additional Protocol and renews it with annual updates. Requests for clarification fill existing gaps of information. Namely questions concerning historical issues require thorough and time consuming researches and only few questions have not yet been answered completely.

But the IAEA already signaled that conclusion could be drawn that neither enrichment nor reprocessing activities have ever been carried out in Switzerland. This is a big step ahead.

6. Conclusions

Starting point of Safeguards was back in 1979. Since then, safeguards in Switzerland have developed together with the renewal of legislation and the entry into force of the Additional Protocol in 2005. Big steps forward to Integrated Safeguards could already been achieved and Switzerland is looking forward that broader conclusions will be drawn in the near future.

Peer reviewed section

C/S in Final Disposal Processes – Swedish and Finnish Perspectives

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Abstract

Sweden and Finland have the same strategy for the treatment of the spent fuel from their nuclear power plants: its direct disposal in geological repositories deep in the bedrock. An important consequence of this strategy is that once the spent fuel is emplaced in the repositories, it will be highly inaccessible. This trait is unique among the nuclear facilities that are under safeguards today. For the credibility of safeguards, the safeguards authorities must ensure that the fuel to be disposed of is verified and that the continuity of knowledge is securely maintained, since no re-verification is feasible if the continuity of knowledge is lost. This highlights the importance of a robust and reliable system of containment and surveillance.

This paper identifies the boundary conditions of such a system, discusses the role of the safeguards authorities and suggests ways of implementation of containment and surveillance in the final disposal processes of Finland and Sweden.

Keywords: Containment and Surveillance; Final Disposal; Geological Repositories

1. Introduction: final disposal in Finland and Sweden

Sweden and Finland are two countries with similar strategies within the field of nuclear power production. For example, both Sweden and Finland implements the once-through fuel cycle where no fuel is reprocessed. This strategy, and the fact that the Fennoscandian peninsula has a stable crystalline bedrock, makes geological repositories the natural choice for the disposal of spent nuclear fuel. The similarities between the two countries make close cooperation not only possible but also clearly advantageous. For this reason, the companies responsible for final disposal, Posiva in Finland and SKB in Sweden, have had research cooperations and jointly funded projects for years.

Sweden and Finland plan to encapsulate the spent fuel in cast iron enforced copper canisters, which will be emplaced in the geological repository. Finland has decided on the repository site, while Sweden is in the phase of site selection with two candidate sites left.

The final disposal procedures in Sweden and Finland will eventually render the spent fuel unavailable for verification and identification. The degree of inaccessibility after disposal in the geological repository will be unique among all facilities presently under safeguards in the world and will present an unprecedented challenge for the design of a credible safeguards system. An important consequence of the inaccessibility of the spent fuel is that all safeguards issues must be resolved prior to emplacement and backfilling.

The disposal process will be continuous with a number of fuel assemblies being prepared for encapsulation and final disposal each week. Since no such operations take place in the nuclear power industry in Sweden or Finland today, the present safeguards system needs to be adjusted to handle this new situation. Although continuous processes like enrichment and reprocessing exist today in other countries, the methodologies there cannot be directly transferred onto the final disposal process. This is due to an important difference: the geological repository does not have an outflow that can be investigated to confirm the presence of all nuclear material. This feature makes it necessary to create a new safeguards approach for final disposal of spent nuclear fuel.

In Finland and Sweden alike, there is a common understanding that comprehensive non-destructive-as-

say (NDA) verification should be performed prior to encapsulation. NDA measurements provide the operators, the safeguards authorities and future generations with understanding of the properties of the disposed material, and allow for an objective assessment of the correctness and completeness of the State's declarations. The continuity of knowledge (CoK) must be vigilantly maintained after the NDA verification. Hence, a robust and reliable system of Containment and Surveillance (C/S) is of vital importance.

The safeguards challenges of spent nuclear fuel disposal were identified by the International Atomic Energy Agency (IAEA) already at the end of the 1980's, and generic safeguards requirements were presented. [1] However, these did not take into account the site-specific requirements and implementation practices, e.g. DIQ (Design Information Questionnaire) forms and inspection routines. In crystalline bedrock, the basic safeguards elements are the jointly applied DIV (Design Information Verification) and C/S functions. The design information of the repository will vary with the progressing disposal operation and the locations of the C/S instruments at the potential pathways to the repository may change with time as well. Therefore, the CoK of both design information and of the spent fuel is essential.

This paper addresses the generic application of C/S procedures to spent fuel. It is based on the current views and plans for final disposal in Finland and Sweden.

2. The final disposal process

The Swedish and Finnish final disposal processes both consist of three blocks [2, 3]:

1. Wet interim storage,
2. Encapsulation plant and
3. Geological repository

Presently, the spent fuel is stored in wet interim storages. These facilities will constitute the starting point of the final disposal process. The encapsulation facility is either adjacent to the interim storage, within the same nuclear site, or further away in which case fuel transport to the encapsulation facility is necessary. In Finland the transport is inevitable, since the Loviisa NPP fuel needs to be transported to Olkiluoto, where the encapsulation facility and the final disposal site is located. Sweden has one central interim storage facility (Clab in Oskarshamn) and the main alternative in Sweden, as presented by the operator, is that the encapsulation facility should be constructed in direct connection to Clab, within the same perimeter.

In the encapsulation facility the spent fuel assemblies will be placed in sealed copper canisters. From a safeguards point of view the encapsulation is a re-batching process where 12 (BWR, VVER) or 4 (PWR) fuel assemblies (nuclear fuel items) are placed in one canister. The canister thus forms the new fuel item of interest for the safeguards system and must therefore have a batch ID that can be read manually or using adequate technical equipment. In this paper it is proposed that the new fuel item, the canister, is defined to be created immediately after the canister has been filled and sealed by the welding of the lid.

After encapsulation, the spent fuel canisters will be transferred or transported to final disposal tunnels and emplaced in disposal holes, which will be backfilled. The operators want to keep the open rock volume as small as possible, so the excavation of new disposal tunnels and the backfilling of old ones is an ongoing process, concurrent with final disposal operations.

3. Boundary conditions for the safeguards system

A few boundary conditions for the safeguards system have been identified:

- The system must be able to create accurate information about the spent fuel. After emplacement and backfilling the fuel cannot be re-verified, therefore the information and the safeguards conclusion drawn from it must be clear, unambiguous, accepted by all parties and well documented. This requirement gives the guidance that:
  - all spent fuel should be subject to verifying measurements. The NDA technique used should be able to provide credible assurance that no diversion of nuclear material from the measured assembly has taken place. This implies that the spent fuel should be verified on at least partial defect level, with acceptable measuring uncertainties. If no such NDA technique is approved for safeguards use at the time a repository is taken into operation, the best available technique should be used. In this case, the safeguards system should be prepared, both technically and conceptually, for incorporation of new, better technologies, as they evolve.
  - after the NDA verification, the CoK should be well maintained. In case of failure, the CoK must be re-established by use of an adequate method, for example NDA.

  - there should be a mechanism created through which the relevant parties (operator, safeguards authorities, the IAEA) can give their statements and clearances before the material becomes difficult to access, i.e. before encapsulation. This brings in the requirement of a sufficient buffer capacity between the final verification and the encapsulation/disposal process.

- The safeguards approach should be feasible in relation to the use of resources. For instance, the system should not require constant physical presence by the safeguards authorities or the IAEA.
- The safeguards system should not interfere with the operator's regular activities, implying that the system and its components must be robust in the operating environment. Sufficient amounts of spare parts and components should be made available for all relevant technical systems. To minimise the intrusiveness of the safeguards system on operations, a buffer of verified fuel assemblies ready for disposal is beneficial. With a buffer, a failure in the NDA equipment could be handled without causing a disruption in the encapsulation process.
- The present three month or one year inspection interval is not a functioning strategy for the final disposal process, since the material will become difficult to access continuously. This makes verification of inventory impossible.
- Backflow of the material in the process chain could severely disturb operations. Therefore, the IAEA and the safeguards authorities should make a great effort to ensure a.) that the C/S system is designed with sufficient robustness and redundancy so that the probability of a loss of CoK is minimised, and b.) that, in case of a loss of CoK, it could be restored by NDA measurements without the need for a backflow of material. Having provided a safeguards system with the capabilities listed in a.) and b.) the IAEA and the authorities will have reduced the probability of a situation requiring a re-take of material. However, if the operator creates conditions such that safeguards cannot be effectively maintained, the situation cannot be excluded. For such an event, The IAEA should retain the option to re-verify material that has not been emplaced and backfilled.

4. Authority requirements

The Swedish and Finnish safeguards authorities have not yet issued any requirements for the safeguards system for the final disposal processes. Under no circumstance can these requirements be less stringent than those (still undefined) of the IAEA. However, some guidelines can be found in the results of the IAEA Consultants Group Meeting of 1995 [3]. The meeting recommendations include: "spent fuel […] be disposed of only as verified nuclear material on which continuity of knowledge (CoK) has been maintained". In the safeguards terminology, CoK is considered maintained as long as the C/S system that has been applied on the nuclear material can be evaluated as acceptable. Due to the intrinsic redundancy of a dual C/S system, it produces an acceptable result also when one part of it fails. A dual C/S system is therefore the reasonable choice and should be required from the authorities. In this context it may turn out to be feasible to re-define dual C/S in such a way that it requires two conclusive positive results in order for a conclusion of maintained CoK, making it an "enhanced dual C/S".

On a national level the authorities' concern is to provide future generations with credible assurance that the nuclear material declared to be disposed of is actually emplaced in the declared location and has the declared properties. This means that correct and complete records have to be maintained over centuries, which implies two challenges:

- to ensure that correct and complete documentation of all nuclear material is created, and
- to preserve this documentation so that it is accessible to future generations.

The authorities should thus require that only verified material, on which continuity of knowledge has been kept, is allowed to pass through the disposal process.

The archival challenge must also be met, not only in the sense that the data and information must be available during an unforeseeable time-span but also with consideration of the authentication of the data.

An important question for the safeguards authorities to consider is the ownership and maintenance of safeguards equipment: should the equipment belong to the authorities or the operators? When balancing these alternatives, the authorities will have to balance the integrity of the collected information against the intrusiveness on operations and resource efficiency. If the authorities own, operate and maintain for example the NDA equipment, it will be highly resource demanding since the equipment will be used continuously. Remote operation of the equipment poses large initial costs but could be

more economical in the long run. However, in case of a failure, the operators have to halt operations until personnel from the authority arrive and repair the apparatus. On the other hand, the integrity of the implemented safeguards system could be questioned if the operators own, operate and maintain the measuring equipment. A related issue is whether the IAEA can use information from the national authorities' safeguards systems. These issues should be carefully considered by the authorities and the IAEA.

5. C/S functions

Possible diversion scenarios are described in the SAGOR reports [1]. To protect against the diversion scenarios we propose the following functions for the C/S of a final disposal safeguards system. It should be noted that C/S functions are not meant to be the only safeguards measures but are complemented with other methods.

1. The assurance of integrity and identity of the fuel elements after the final verification until encapsulation is achieved by the constant surveillance and appropriate sealing of items. Dual C/S (or perhaps "enhanced" Dual C/S) is recommended, as losing CoK at this stage would interrupt the encapsulation process and may require re-verification. A fingerprinting system of fuel elements would make confirmation of CoK more robust and, consequently, requirements of the C/S systems could be relaxed. If this phase includes fuel transport over long distances, a tracking system may be appropriate.

2. The encapsulation process is performed under constant, uninterrupted surveillance. For monitoring the flow in the encapsulation facility, portal monitoring is a viable option.

3. The CoK must be maintained also in the tunnel system of the repository. A risk assessment analysis is necessary to find the best way to achieve this. Environmental conditions in the disposal tunnels excavated in the crystalline bedrock in Sweden or Finland are expected to be such as to allow for the use of portal monitoring systems or other forms of robust radioactivity measurements. Monitoring of only the access points to the repository (ramp, lift and ventilation shafts and drill holes from the site investigation) while viewing the tunnel system as a "black box" is a desirable safeguards approach because of the relatively low costs for equipment and labour. However, a risk assessment may give the result that the "black box" approach is not sufficiently secure. In that case it should be complemented with additional safeguards activities underground. An example is that the emplacement vehicle could be equipped with a fingerprint reader and a positioning and radiation monitoring system to detect any replacement attempts. After the canister is emplaced, non-C/S activities like inspections and environmental sampling could reveal any undeclared opening of canisters. For the purpose of monitoring activities in the final disposal tunnels, the performance of portal monitoring should be investigated. The monitoring should be sensitive enough to detect the diversion of a shielded canister if it is to be of use for safeguards purposes.

6. Process models

The requirements and the performance of a safeguards system can be analysed only if the disposal process is known. At the moment the process is not completely defined either in Finland or in Sweden. Since the Finnish plans are more evolved, this preliminary model is created from the Finnish case.

6.1 Analysis of process model "FIN-1"

The first safeguards concept for the Finnish final disposal process is proposed in a STUK report [5]. The concept is produced for the process model FIN-1, which is based on the following assumptions:

- There are interim storages, one of them being within the same site as the planned encapsulation plant and geological repository.
- Final disposal tunnels are within the same site as the encapsulation plant.
- Encapsulation is an ongoing process with a capacity of 2 canisters/week.
- Final NDA verification will be performed at the interim storages, which have separate pool areas where verified fuel is stored. These pools serve as a buffer zone.

Early NDA verification is a benefit to the safeguards system because it provides large buffer storages of verified fuel ready to be introduced into the final disposal process. A long analysis period also follows from the early measurements, which means that there is time to draw safeguards conclusions before encapsulation. This makes the system non-intrusive.

After the NDA verification the challenge is to maintain the CoK. We propose to develop special fingerprinting methods, which would be used simultane-

ously with the NDA verification. A "fingerprint" would be a specific pattern based on a unique physical characteristic of the fuel assembly. This pattern should be practically impossible to counterfeit. An example of such a method is reading the microstructures of the fuel item surfaces [6]. The selected method could be combined with weighing and radiation pattern measurements of the fuel assembly to detect if the integrity of the item has been breached.

The fingerprinting method would make the C/S system more robust since the fingerprint would provide identification of the item and evidence that item integrity is intact. However, no such system is in use today. Potential candidates exist, like surface scanning methods, which can be used to uniquely identify surfaces or bolts of the fuel assembly. The reading of fingerprints would detect the replacement of an assembly, as well as the removal of bolts. A similar fingerprinting method could be also used to verify its identity and integrity of the disposal canister.

All buffer storages and fuel movement operations should be continuously monitored with a camera. Sealing may be used as a complementary method. For fuel transports the use of a tracking system is proposed.

Underground operations are challenging to monitor. If all access points to the repository can be comprehensively monitored by radiation detectors, the detector network could be a backbone of the safeguards system. However, the detection capability of the radiation detectors should be investigated. The need for other C/S methods or even the use of novel technologies can be analysed when the process is better known.

The safeguards processes of FIN-1 model are described in Table 1. Some of the proposed methods are optional. Further analysis is needed.

Table 1: C/S and other safeguards methods in final disposal process model "FIN-1".

Process	Purpose	Technical method	Operational requirement
NDA verification	To verify the correctness and completeness of the State's records.	Tomography or another partial defect method.	Reliability: medium (MTBF = few years). Servicing time less than 1 month. A spare system may be required. Remote monitored.
Assembly fingerprinting	To create an unambiguous fingerprint of the assembly for later identification. The fingerprinting and NDA verification are simultaneous operations. Fingerprinting could also detect if the integrity of the assembly is compromised.	Surface fingerprinting (or some other identification method). Radiation fingerprinting. Measuring the assembly weight. Further research is required.	Reliability: medium (MTBF = few years). Probability of false positives or false negatives lower than 10^{-6}. Servicing time less than 1 month. A spare system may be required. Remote monitored Integration with NDA verifier (fingerprinting and verification are simultaneous operations).
Buffer storing of verified assemblies	To maintain CoK during the cask loading.	Camera surveillance.	Camera reliability medium (MTBF = few years). Remote monitoring desirable.
Transport cask loading	To maintain CoK during the cask loading.	Authenticated fingerprints maintain CoK. Camera surveillance.	Camera reliability medium (MTBF = few years). Remote monitoring desirable.
Cask transport/transfer	To maintain CoK during the transport and to detect covert opening and substitution.	Authenticated fingerprints maintain CoK. Seals.	Seal reliability high (MTBF = several years). Remote monitoring desirable.
Cask storing	To maintain CoK during the cask storing and to detect covert opening and substitution.	Authenticated fingerprints maintain CoK. Seals.	Seal reliability high (MTBF = several years). Remote monitoring desirable.
Cask unloading	To maintain CoK during the cask unloading and to detect diversion and substitution.	Authenticated fingerprints maintain CoK. Camera surveillance.	Camera reliability high (MTBF = several years). Remote monitoring essential.

Encapsulation process	To verify CoK of the assemblies.	Verification of fingerprints. Remote monitoring cameras, inspector presence, portal monitors. All diversion routes should be covered.	Overall reliability: high (MTBF = several years). Servicing time: few days. Remote monitoring system required.
Canister fingerprinting	To create an unambiguous fingerprint from the canister for later identification. Fingerprinting could also detect if the integrity of the canister is compromised.	Surface fingerprinting (or other identification method) or other novel method. Radiation fingerprinting. Measuring canister weight. Further research is required	Overall reliability: high (MTBF = several years). Servicing time: few days. Remote monitoring system required. Probability of false positives or false negatives lower than 10^{-6}.
Canister transfer to the emplacement hole	To maintain CoK during the transfer.	Authenticated fingerprints maintain CoK. Portal monitors. All diversion routes should be covered.	Overall reliability: high (MTBF = several years). Servicing time: few days. Remote monitoring system required.
Canister emplacement	To verify CoK of the canister.	Verification of fingerprints. Remote monitoring cameras, inspector presence, portal monitors. All diversion routes should be covered.	Overall reliability: high (MTBF = several years). Servicing time: few days. Remote monitoring system required.
Backfilling	To detect undeclared removal of the canister	Cameras, inspector presence, portal monitors. Novel methods.	Overall reliability: extremely high (MTBF = tens of years, Dual systems & redundancy). Servicing time: few days.
After backfilling	To detect undeclared access to the repository/canister.	Sealing, geophysical methods, satellite monitoring. Novel methods.	Passive systems desirable.

7. Challenges

This paper has identified a number of challenges in the disposal process in geological repositories in Sweden and Finland that can be resolved either by research efforts or by analyses and decisions made by the safeguards authorities:

- An NDA verification system that would satisfy the needs for partial defect level verification does not exist today. Development of a tomographic verifying technique could potentially solve this problem. Such work is done under the IAEA Support Program task JNT 1510, with active participation by Finland and Sweden.
- The performance of portal monitors for use in the final disposal tunnels needs investigation. If the radiation detectors used as portal monitors can be made sensitive enough to detect the diversion of a shielded canister they could be implemented with the purpose of monitoring activities in the repository.
- A fingerprinting method to be used on fuel assemblies before encapsulation, and on the closed disposal canisters after encapsulation, is desirable. The method should be fast and with a very low probability of false identification.
- For a resource efficient operation of safeguards equipment, remote monitoring will be needed. This concept will have to be developed further, including extensive reliability testing.
- A lot of effort (see for example [7]) is being put into finding a secure way of transferring information to future generations. This research should be closely watched by the national safeguards authorities, whose task it is to inform the coming generations of the contents of the geological repositories. A method to authenticate the infor-

mation to be placed in archives is also needed since the correctness of the information is as important as its completeness.

- The question of ownership of safeguards equipment should be resolved. The IAEA also needs to decide on the possibility of using information created by the national safeguards authorities.
- As has been stressed above, the C/S system needs to be both robust and reliable. If the C/S system were to deliver inconclusive results the operation of the final disposal process would be affected negatively. A methodology of determining the C/S system's ability to deliver conclusive results is therefore desirable. At Uppsala University in Sweden, a research project is planned that aims at quantifying the ability of a C/S system to detect an anomaly and to reveal the course of actions leading to the anomaly. Such quantification would provide a possibility to logically connect the important concept of CoK to measurable entities. Furthermore, in the ESARDA Working Group on Containment and Surveillance a qualitative method of determining a system's Assurance and Performance is being developed.
- In Sweden and Finland today, the safeguards system does not operate on continuous processes like the future final disposal process. This means that the methods used today are not well adapted to the future situation where the encapsulation plant and the repository are in operation. Some methods used abroad in safeguarding continuous processes, like enrichment or reprocessing, could possibly be implemented but there is one very important difference: the enrichment and reprocessing plants have an out-flow that could be compared to the in-flow to verify that all nuclear material is accounted for, while the material emplaced in the repository will not be accessible for verification. This is a completely new trait, unique among all nuclear facilities in operation today, and it will demand a concentrated planning and design effort from all instances involved in safeguards in Sweden and Finland. This means that close co-operation at all levels, including the authorities SKI (Sweden) and STUK (Finland), would be of great benefit for both countries.
- Connected to the previous item is the issue of measuring strategy, measuring accuracy and confidence levels. To obtain a cost effective safeguards system with predictable properties it is important to define exactly what should be measured, and for what purpose. The measurements should thus be performed with rationally defined limits on accuracies in order to be able to quantify the performance of the implemented system as a whole. This is important in order to be able to determine with which confidence one may state that the final disposal processes run according to regulations and declarations. Hence, we propose that a project addressing these issues is initiated.

8. References

[1] *Safeguards for the final disposal of spent fuel in geological repositories,* IAEA STR-312, 1997

[2] J.Tanskanen, *Facility Description 2006,* Posiva Working Report 2007-19.

[3] SKB: *Programme for research, development and demonstration of methods for handling and final disposal of nuclear waste, including societal research*, 2004

[4] *Report of the consultants group meeting on safeguards for the direct final disposal of spent fuel in geological repositories,* STR-305, IAEA, 1995

[5] J. Rautjärvi, A. Tiitta, J. Saarinen, *Preliminary concept for safeguarding spent fuel encapsulation plant in Olkiluoto, Finland.* Phase III report on Task FIN A 1184 of the Finnish support program to IAEA Safeguards. STUK-YTO-TR 187. STUK, Helsinki 2002.

[6] J. Buchanan, *'Fingerprinting' documents and packaging*, Nature vol. 436, 2005

[7] Y. de Lusenet, *Preservation of digital heritage*, European Commission on Preservation and Access, 2002

3D Reconstruction in Nuclear Security

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Abstract

Accurate modelling is gaining increasing importance in security applications. Indeed, realistic and dimensionally accurate models of critical areas can be used for prevention and simulation exercises as well as for planning emergency responses once an attack is perpetrated. CEA-DAM and EC-JRC engaged in a joint exercise involving the simulation of a terrorist attack in an urban area with possible release of radiological substances. JRC was responsible for creating a dimensionally accurate (centimetre accuracy) 3D model from the urban area "as-is" before and after the attack. Further tests involved the automatic 3D detection of changes in both indoors and outdoors environments. The paper describes the principles and technologies behind the generation of photo-realistic and accurate 3D models of wide areas "as-is", and will discuss the use of those technologies for nuclear security applications.

Keywords: Nuclear Security, 3D Reconstruction, Data Integration, Change Detection

1. Introduction

The French Commissariat à l'Energie Atomique, Direction d'Applications Militaires – CEA-DAM – and the European Commission's Joint Research Centre, Institute for the Protection and Security of the Citizen – JRC-IPSC – engaged into informal discussions on technologies relevant for security research. From these discussions, JRC's three dimensional laser-based technologies were identified as of interest to future security applications.

CEA-DAM invited JRC-IPSC to participate in a joint exercise involving the simulation of a terrorist attack in an urban area with possible release of radiological substances. JRC was responsible for creating a 3D model from the urban area "as-is" before and after the attack. To this effect a JRC team travelled to the exercise site and made a photo-realistic and dimensionally accurate 3D reconstructed model using laser range and photography scanning. 3D Data acquired from different viewpoints were registered and integrated with the photographic records into a single geometric model with centimetre accuracy. Following the "attack", specific spots of the urban area were again scanned and new models created. Several practical applications of these 3D models are currently being considered:

a) Documentation of the area under attack – before and after;

b) Detection of changes;

c) Geometric input to atmospheric dispersion models;

d) Calibration of atmospheric dispersion models in what concerns the propagation and diffusion of smoke plumes and clouds;

e) Generic platform for the intuitive presentation and integration of data from multiple sources and time-frames (e.g., ground penetrating radar, satellite and airborne images and 3D scans, radiation and thermal images, CAD data, etc.)

This paper describes the investigations done, including the results obtained in the experimental campaign. It also looks forward in terms of future security applications.

2. 3D reconstruction

Three dimensional reconstruction – 3D Reconstruction – refers to the techniques that allow the creation of dimensionally accurate geometric models of real environments both indoors and outdoors, including small objects. For several years JRC has been developing techniques to this effect [1, 2]. Most of the techniques are based on the use of laser based instrumentation to generate 3D cloud of points of the object or environment to reconstruct. These clouds of points contain the 3D information as perceived from the sensor. In general, to reconstruct an environment more than one single-view capture point is necessary as some objects hide – i.e., occlude – others.

Figure 1: (left) Laser scanner on tripod with dolly for indoor operation; (right) Laser scanner mounted on vehicle for outdoor operation.

Figure 1 shows two commercial, off-the-shelf, laser range scanners that are used for 3D data collection. Whereas the laser on the left (Fig. 1.a) is mainly used for indoor applications the laser on the right (Fig. 1.b) is generally used for outdoor applications. This division is mainly the result of considerations on accuracy, spatial resolution and maximum measured range. Table 1, illustrates the main features for both instruments. The reconstruction of small objects is mainly obtained using triangulation based laser scanners though the details are beyond the scope of this paper.

Once an environment is scanned there is the need to:

a) Get a single coordinate frame to the data from single-view scans – registration:

b) Combine the data from multiple scans and, eventually, discard 3 data

c) Integrate other types of data into the 3D reconstructed model. Important pieces of data are photos from the environment to create visually realistic models

All the above operations are done using JRC's *3D Reconstructor*® software package. Figure 2 describes the architecture/paradigm behind the complete and realistic 3D reconstruction of environments. The *3D Reconstructor* package implements the different processing blocks.

To illustrate a practical use for *3D Reconstructor* technologies one should mention its routine use by the International Atomic Energy Agency (IAEA) for design information verification (DIV) purposes. DIV aims at accurately mapping interiors of complex nuclear facilities under safeguards. DIV activities are performed during the complete life cycle of a nuclear facility to confirm that it operates as declared and is not misused for undeclared nuclear activities [3].

3. Joint nuclear security exercise

CEA's emergency response project organized an internal – to the CEA – security exercise involving the participation of radiological and nuclear experts from several sites. The exercise was triggered by an

Table 1: Main features of two laser scanners used for 3D modelling.

	Z+F Imager 6	Riegl Z 420i
Range	1 – 79 m	2 – 250m (> 10%) or 800m (> 80%)
Maximum Acquisition Rate	500,000 points per second	12,000 points per second
Accuracy	Better than 4 mm	10mm
Scanner Angular View	310° (vert.) by 360° (horiz.)	80° (vert.) by 360° (horiz.)
Spatial Resolution – Vertical	111.1 samples/° (34,444 samples)	125 samples/° (10,000 samples)
Spatial Resolution – Horizontal	111.1 samples/° (40,000 samples)	100 samples/° (36,000 samples)
Output	range and reflectance	range and reflectance
Eye Safety (Laser Class)	Class 3R	Class 1

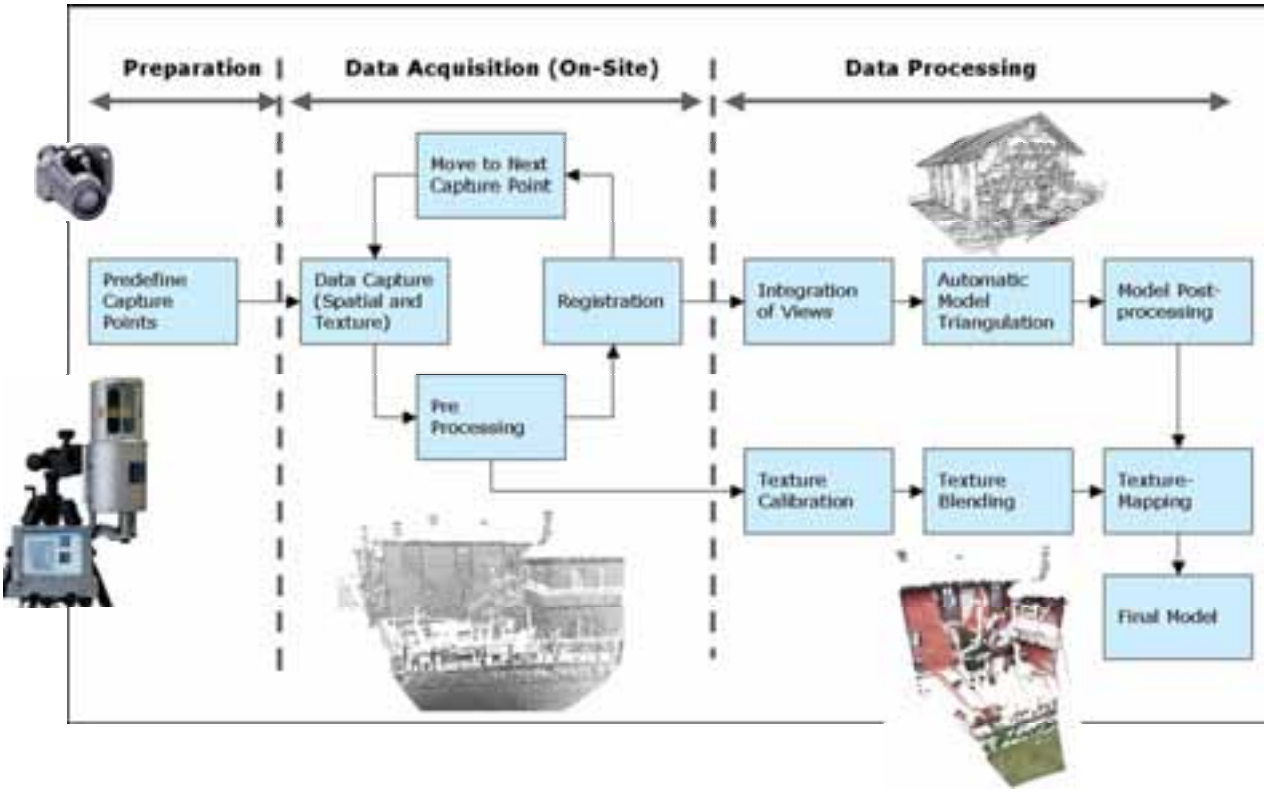

Figure 2: System's Architecture of JRC's *3D Reconstructor*®.

'alarm' indicating that a "terrorist accident with release of radioactive/radiological substances had taken place at the CEA-CESTA research centre."

The "attack" consisted of having two cars burn/explode in an urban area, with simulated release of several samples of radiological and/or radioactive material. Different teams from different CEA sites immediately travelled to CESTA. The teams brought with them a van with NDA and radiological detection equipment, as well as a car equipped for detecting radioactive substances in urban environments. All measurements are geographically referenced using on-board GPS.

JRC's participation: During the exercise the JRC team created 3D models from the urban environment before and after the simulated terrorist attacks. This included scans from:

a) an indoor environment – warehouse – to document the premises and detect changes;

b) the urban environment – outdoors – for 3D site reconstruction;

c) the burning vehicles – outdoors – to investigate the use of 3D information to model the smoke plumes and clouds.

The following sections will detail the three independent exercises

4. Indoor environment

The objectives of this exercise were twofold:

i) to create a dimensionally accurate and visually realistic model of an indoor environment that could be used for documentation purposes;

ii) to detect minor changes occurring in this quasi-static environment

The warehouse was scanned from ten different positions with the Z+F laser scanner. For each scan a set of seven photographs were taken (total: 70 photos). With both the range and photo images a millimetre accuracy model of the environment was created. Figure 3 illustrates the 3D model of the warehouse.

Two more scans were done to detect minor changes. It should be noted that none of the changes introduced in the environment were known to the team. All changes were detected. Figure 4 shows some of the changes detected in red/orange. These include (a) changing the position of a mobile phone on a shelf; (b) changing the positions of a few boxes on the shelves; (c) rotating a pot with a plant; (d) changing the position of the crane; (e) changing the position of the forklift vehicle.

The total time for acquiring the 3D and visual data for ten modelling scans and two verification ones,

Figure 3: View of the 3D reconstructed model of a warehouse. Ten laser scans were used to build this model, taking approximately 60 minutes. All points were measured with an accuracy of 4mm. Light blue indicates areas for which no 3D information is available (i.e., have not been scanned).

(a) (b) (c)

Figure 4: Detection of changes inside the warehouse. Changes are indicated in red/orange: (a) changes in the shelves area; (b) changes in warehouse's main hall – three objects were detected: a mobile phone on the shelf (bottom left), the crane cable (top right) and the forklift (centre-right); (c) false colour code – red corresponds to distance changes larger than 10cm; change in the object in (a) is about 6.5cm.

was about 90 minutes. Processing time amounted to about 3 hours.

5. Outdoor urban environment

In this case, and considering the dimensions and complexity of the wide area environment the work was divided in two parts:

Part I: Build a general overview model of the site – this objective involved the creation of a 3D model of the site layout as "seen" from the existing water tower which was then updated by local scans acquired on the ground. Figure 5 shows three snapshots of the site model. The model integrates data acquired from the water tower and 7 scans acquired at ground level.

Part 2: Document the "terrorist" attack scene – this objective was met by scanning from several viewpoints – to resolve for possible spatial occlusions – the street where the "terrorist" attack took place. This was done before and after the attack. Figure 6 illustrates these scans.

Given that in a few cases the range of distances was inferior to 50meters, both the indoor and outdoor laser scanners (see Figure 1) were used. JRC's *3D Reconstructor* software can easily merge and integrate data from scans with different spatial resolution.

Figure 5: Partial 3D Overview of the CESTA site. Eight laser scans were used to create this model. Each model point has an accuracy of about 2cm. Processing time amounted to about 3 hours.

Figure 6: View of the scene before the 'attack'.

6. Plume detection and modelling

Any explosion attack is normally associated with a fire and corresponding smoke. Depending on the explosion, dangerous chemicals may be freed into the open atmosphere. It is thus of interest to predict how such dangerous cloud will disperse in the air. This knowledge is particularly relevant in urban areas to:

(a) help deciding where to concentrate rescue effort (resource management)

(b) time allowing, alert the population

Cloud dispersion is a complex modelling exercise, especially at short ranges, as it depends on many factors such as, wind direction and speed, air temperature and humidity, gases to be dispersed, and on the specific 3D topology of the environment both where the explosion took place and of its vicinity. During the preparation of this joint exercise it was found useful to make a few experiments in order to provide quality data to assist the atmospheric dispersion modelling.

3D Modelling of the Environment: the laser scanning technique and 3D Reconstuctor software provide easily a complete modelling of the environment "as-is" with an accuracy of about 2cm. Such 3D model can be used as input in any short-range atmospheric dispersion model.

3D Plume Detection and Modelling: Many 3D scans of the plumes were made in order to find out whether the laser scanning equipment could (i) detect the plume itself and (ii) trace its dispersion in terms of both height and orientation.

The two laser scanners – indoors and outdoors – were used in the experiments. It was soon found out the indoors scanner could not always detect the plumes, probably due to the small diameter of the laser beam. Encouraging results were obtained with the outdoor laser.

It was possible to detect the plume, even when it was very dark (i.e., corresponding to poor laser reflectance). It should be noted that with the use of a single laser scanner it is possible to detect and map only one of the sides of the envelope of the smoke plume. In other words it is not possible to measure either the plume diameter (i.e., thickness) or its density. Further, changing wind speeds and the chaotic

Figure 7: Two snapshots of the 3D model of the smoke plumes emanating from the burning car on the left. The laser scanner is located at the right. The dashed lines indicate the scanning field of view (FoV). Only the smoke plumes inside the FoV were captured.

behaviour of the explosion fire makes plume dispersion a highly dynamic four dimensional phenomenon, i.e., the shape and orientation of a plume change in time. Figure 7 illustrates the capability for the 3D detection of the smoke plumes.

7. Future work

Combining 3D models with radiation maps – It is possible to create dimensionally accurate and photo-realistic 3D reconstructed models from real environments both indoors and outdoors. This representation can be used in many ways, including documentation, detection of changes and for training purposes.

It is possible to use the 3D models as a human presentation and interfacing tool by integrating the 3D model with real measurements such as radiation or thermal maps. Figure 9 illustrates the results of experiments made in collaboration with US DoE's Oak Ridge and Lawrence Livermore National Laboratories [4]. In this particular case the 3D model of a room is integrated with both the photographic map and a gamma radiation map. It is possible to see the gamma measurements produced by a radioactive source inside one of the pipes hanging from the ceiling. Such combined representation eases the interpretation of the scene as it allows the easy location of relevant sources, including the accurate and interactive measurements of distances or volumes.

The same principle can be used outdoors integrating 3D and radiation mapping of large and complex environments such as urban areas. Basic 3D data of an urban environment "as-is" (i.e., not an idealised CAD model) is acquired at the ground level, e.g., using the vehicle of Figure 1.b. This data provide good detail of buildings but fail to describe the roofs or building tops. Airborne data – both range and photos – do complement the data acquired at ground, i.e., at the street level, and make possible to build complete 3D urban models. Instruments can be flown aboard a UAV – unmanned aerial vehicles, plane or helicopter [5].

Similarly, radiation data can be acquired both at ground level and airborne. In the field of security of special events, CEA-DAM has used for several years both a vehicle – AutoNuc – and a helicopter – HelicNuc – equipped with radiation detection equipment [6, 7, 8].

Radiation data from AutoNuc and HeliNuc (Figure 10) are geo-referenced based on onboard GPS sensors. These data constitute the radiation baseline for a given scanned area. Indeed, in large and complex environments, such as urban areas, one cannot expect the radiation reference level to remain constant in the entire area. The integration of radiation maps onto 3D urban models allows for the comparison with subsequent verification scans in order to detect structural and/or radiation changes leading to more detailed or targeted investigations.

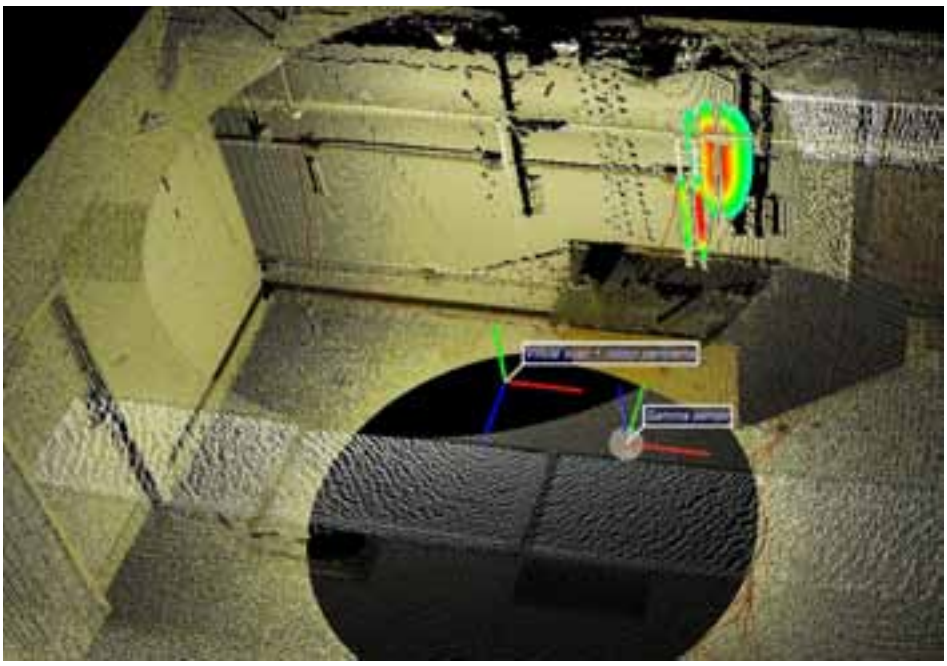

Figure 9: Snapshot of the 3D and gamma mapping integrated model. The laboratory room has two pipes hanging from the ceiling. One of the pipes contains a radiation source inside. The positions of the gamma-ray imager and 3D scanner are represented.

(a) (b)

Figure 10: CEA's AutoNuc Radiation Detection Vehicle and HeliNuc Radiation Detection Helicopter.

The integrated structural and radiation models allow for:

(a) Detection of radiation changes – using the radiation maps as reference;

(b) Detection of structural and geometric changes – using the urban model as reference (i.e., using data from previous 3D scans or from other sources, such as cadastre, GIS or CAD models). Subsequent scans, as part of a verification exercise, can then be used to detect 3D changes occurring at the ground level as well as on building roofs or terraces.

(c) Immediate and easy interpretation of the changes, as the scene is realistically represented by both the geometrical model and its photographic description;

(d) Easy correlation and link to other information sources such as, maps of electricity, gas, water, communication networks or description of buildings' functions.

(e) Finally, accurate location of the areas to be further investigated

Combining 3D models with Ground Penetrating Radar – GPR is gaining an increasing role as a security sensor. Indeed, its potential to detect cavities, hidden rooms or underground cells in buildings or tunnels in public areas is highly relevant to find hideouts or places where suspicious activities may

be planned. As with many other sophisticated sensors, the output of GPR needs to be interpreted by expert staff. Both GPR and 3D laser scanner compute distances from the time analysis of an echo signal and their output corresponds to two datasets of 3D points. This parallel suggests that the integration of 3D laser range data with GPR could be beneficial for the global overview and correct interpretation of complex scenes.

8. Conclusions

The following capabilities for JRC's 3D modelling system – indoors and outdoors – were successfully tested during the experiments. It was possible to prove the system's:

(a) **Portability**: the system is highly portable. It can be easily packed and transported in either a car or a plane

(b) **Fast deployment**: once an emergency occurs the system can be installed and made operational in a few minutes

(c) **Accuracy**: the system can model a real environment – both indoors and/or outdoors – and provide a dimensionally accurate representation (typical accuracies are 3mm indoors and 2cm outdoors)

(d) **Speed**: the system only takes a few minutes to scan an environment and produce a good quality documentation

(e) **Ease of Processing**: all data processing can be done in a normal laptop computer, i.e., there is no need for sophisticated and expensive hardware

(f) **Interoperability**: JRC's *Reconstructor*® software has many import and export filters enabling the easy connection to other related applications, e.g., CAD programs.

From the 3D models – either archived or acquired – it is possible to launch a 3D scene change detection application and detect minor changes in the environment. The tool is highly effective in detecting minute changes, though a detected change might not be necessarily associated to a security alarm. Indeed, it is up to the system's operator (e.g., inspector, police officer, security guard) to interpret the scene and validate the change. The more information she or he has available the easier the interpretation of the event will be, i.e., the more accurate results will be.

One of the laser scanning systems showed the capability to detect smoke plumes and map its dimension and orientation. Though it was not possible to fully represent a whole plume, one can say, however, that the acquired 3D plume information can be used as a simplified "ground-truth" representation of the plume's behaviour – dimensions and orientation. This can be relevant in fine tuning some dispersion modelling parameters.

9. References

[1] V. Sequeira, A. Busboom, G. Boström, J.G.M. Gonçalves, "3D Technologies in Safeguards Applications", IAEA Symposium on International Safeguards: Addressing Verification Challenges, Vienna, Austria, October 16-20, 2006.

[2] V. Sequeira, G. Boström, M. Fiocco, D. Puig, J.G.M. Gonçalves, "Outdoor Verification System", in Proc. INMM 46th Annual Meeting, Phoenix, Arizona, USA, July 10-14, 2005.

[3] E. Agboraw, S. Johnson, C. Creusot, S. Poirier, H. Saukkonen, B.Chesnay, V. Sequeira, "IAEA Experience using the 3-Dimensional Laser Range Finder (3DLRF) for Design Information Verification (DIV) at the Rokkasho Reprocessing Plant", IAEA Symposium on International Safeguards: Addressing Verification Challenges, Vienna, Austria, October 16-20, 2006.

[4] L. Mihailescu, K. Vetter, W. Ruhter, D. Chivers, M. Dreicer, C. Coates, S. Smith, J. Hines, A. C. R. Caiado, V. Sequeira, M. Fiocco, and J.G.M. Gonçalves "Combined Measurements with Three-Dimensional Design Information Verification System and Gamma Ray Imaging", Proc. INMM 47th Annual Meeting, Nashville, Tennessee, USA, 16-19 July 2006.

[5] "Combating Nuclear Terrorism – Federal Efforts to Respond to Nuclear and Radiological Threats and to Protect Emergency Response Capabilities could be Strengthened", GAO – United States Government Accountability Office, Report GAO-06-1015, September 2006.

[6] Guillot L., Proscoura M. I., Marchand D. – "Utilisation du Système de Détection Gamma Aéroporté HELINUC en Situation Accidentelle ou Post-Accidentelle. Application à Tchernobyl", Radioprotection – Colloques, vol. 37, C1, 2002.

[7] Guillot, L. – "Aerial Gamma Spectrometry in case of Emergency: Example of Franco-German Exercise Helga", Proc. INSINUME 2004 Conference, Albena, 2004.

[8] Guillot, L., Bourgeois, Ch. – "The Use of the Airborne System Helinuc for Orphan Sources Search", Proc. of International Conference on the Safety and Security of Radioactive Sources, Bordeaux, 2005, IAEA-CN-134.

Use of Imaging for Nuclear Material Control and Accountability

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Abstract

The recent addition of imaging to the Nuclear Materials and Identification System (NMIS) using a small portable DT neutron generator with an embedded alpha detector to time and directionally tag neutrons from the DT reaction is discussed. The generator weighs ~35 lbs including power supplies ($5x10^7$ n/sec) and operates on 50 watts of power. Thus, the source can be easily moved to a variety of locations within an operational facility with minimum impact or it can be used at a fixed location for example to monitor receipts. Imaging with NMIS not only characterizes the detailed shape of a containerized object by transmission tomography but also determines the presence of fissile material by measuring the emitted radiation from induced fission. Imaging measurements with a time tagged Cf spontaneous fission source are also discussed. Previous work studies have shown that this type of imaging has a variety of applications other than nuclear material control and accountability (NMC&A). These include nonproliferation applications such as verification of configuration of nuclear weapons/components shipped or received, warhead authentication behind an information barrier, and traceability of weapons components both fissile and non fissile in dismantlement and counter terrorism. This paper focuses on the use of imaging for NMC&A. Some of the NMC&A applications discussed include: verifying inventory and receipts, making more accurate holdup measurements especially where thicknesses of materials affect gamma ray spectrometry, determining the shape of unknown configurations of fissile materials where the material type may be known but not the form, determining the oxidation of fissile metal in storage cans, fingerprinting the content of storage containers going into a storage facility, and determining unknown configurations for criticality safety.

Keywords: neutron, imaging, uranium metal, NMC&A, DT generator

1. Introduction

Tomographic and radiographic imaging capabilities have been added to the Nuclear Materials Identification System (NMIS) [1] at Oak Ridge National Laboratory (ORNL). The system uses transmission imaging with a time and directionally tagged portable DT generator [2 & 3] or a time tagged Cf spontaneous fission source [4]. Neutrons from the DT generator are more effective on penetrating of hydrogenous materials than Cf fission neutrons by a factor of ~5. However, Cf is adequate for objects with low attenuation and where measurement time is not a factor. The generator has an additional advantage in that it can be turned off when not in use. These imaging capabilities supplement the system's existing time-correlation (coincidence) measurements to quantify fissile material and enhance the system's ability to identify fissile material in containers. This capability has a variety of applications in NMC&A such as determination of the fissile mass holdup in pipes; especially in situations where heavy deposits produce self shielding that make gamma ray spectrometry determinations of hold up have large uncertainties. One such holdup measurement was satisfactorily performed in 1998 on the so called "hockey stick" deposit at the K-29 building of the former gaseous diffusion plant at Oak Ridge [5]. The present NMIS with imaging can satisfy all DOE requirements for confirmation of receipt of weapons components at the Y-12 plant and its continued development will lead to a system that accurately measures the fissile mass in Y-12 receipts. Other than identification and confirmation of weapons components there a variety of other applications such as: determining the form of legacy materials such as highly enriched uranium (HEU) in storage cans where the form is not known, fingerprinting the configurations of HEU going into long term storage facilities, determining the fissile mass in plant components that process oxide and other materials without cleaning out the components, comparing the images of fissile materials in shipping containers where the detailed shape is available from the shipper, determining the amount of oxidized metal in storage cans for fissile metal, determining unknown configurations of fissile materials for criticality safety, and identifying appropriate standards for other NMC&A measurements on cans where internal configuration is not known. The present status of this system is described in this paper and examples of

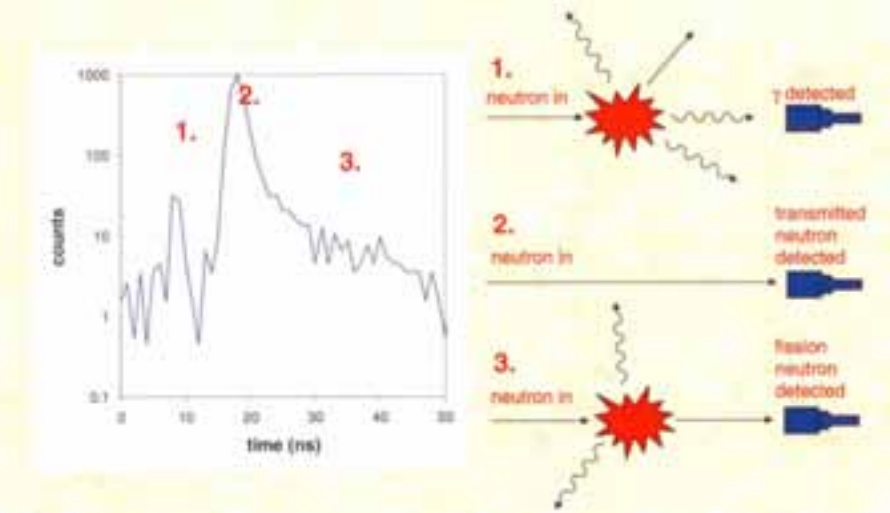


Figure 1: Typical time distribution of counts in a plastic scintillator after the DT reaction in a transmission measurement through a highly enriched uranium metal.

imaging of objects with both a DT generator and a Cf source are presented with some comparisons of results for both sources.

2. Methodology

Figure 1 shows a plot of the time distribution of counts after the DT reaction. Three regions are of interest. In region 1, the gamma rays from induced fission and inelastic neutron scattering are detected first because they travel to the detectors with the speed of light and arrive before the transmitted neutrons. In region 2, the 14.1 MeV transmitted neutrons arrive at the detector. Region 2 also contains forward elastically scattered neutrons that have little change in direction and energy for HEU. Finally, in region 3 the neutrons and gamma rays from induced fission in the HEU are detected and arrive with an amplitude and time dependence characteristic of the HEU mass and multiplication. Region 2 is used for the imaging measurements, while region 3 is used to distinguish HEU from depleted uranium (DU).

Imaging with a DT generator consists of measuring the detector counts as a function of time after the DT reaction with the sample between the source and the detectors I(t) and the count rate as a function of time without the sample present $I_0(t)$ using the relationship $I(t) = I_0(t)exp(-\mu x)$, where x is the sample thickness, and μ is the attenuation coefficient to obtain the value of μx, the attenuation. The values of I and I_0 are obtained by integration of the transmission peak shown in region 2 of Figure 1 with an empirical correction to remove some scattering. This imaging was performed as a function of height using a scanner that could rotate the eight small 1 × 1 × 6-in.-thick detectors 15° in the horizontal plane to simulate a larger number of detector positions.

The time distribution of counts for 4 plastic scintillators spaced ~100 cm in air from the source for a time tagged Cf spontaneous fission source is shown in Figure 2. The initial peak at the time of flight of gamma rays (~3 nanoseconds) from the source fission to the detector is the prompt gamma rays and is broadened by the time resolution of the detection systems for detecting spontaneous fission in the time tagged source and the event in the detector. The following peak (25-80 nanoseconds) is the neutron distribution whose time dispersion is determined by the energy distribution of prompt neutron emitted in spontaneous fission of Cf. Beyond 80 nsec, the distribution is mainly from neutron scattering from the floor. Separation of the gamma rays and neutrons allows both neutron and gamma ray imaging, with neutrons sensitive to light materials and gamma rays sensitive to heavy materials. For neutron imaging the area near the peak of the neutron distribution was used while for gamma rays the integral of the gamma peak was used.

The source of preference is the DT generator with embedded alpha detector because of the penetrating capability of 14 MeV neutrons, shorter measurement time, and ability to turn it off when not in use.

3. Results for DU metal casting with DT generator

A standard DU-metal annular Y-12 National Security Complex storage casting in a steel can was available for imaging measurements at ORNL. This casting has an outside diameter of 12.70 cm, an in-

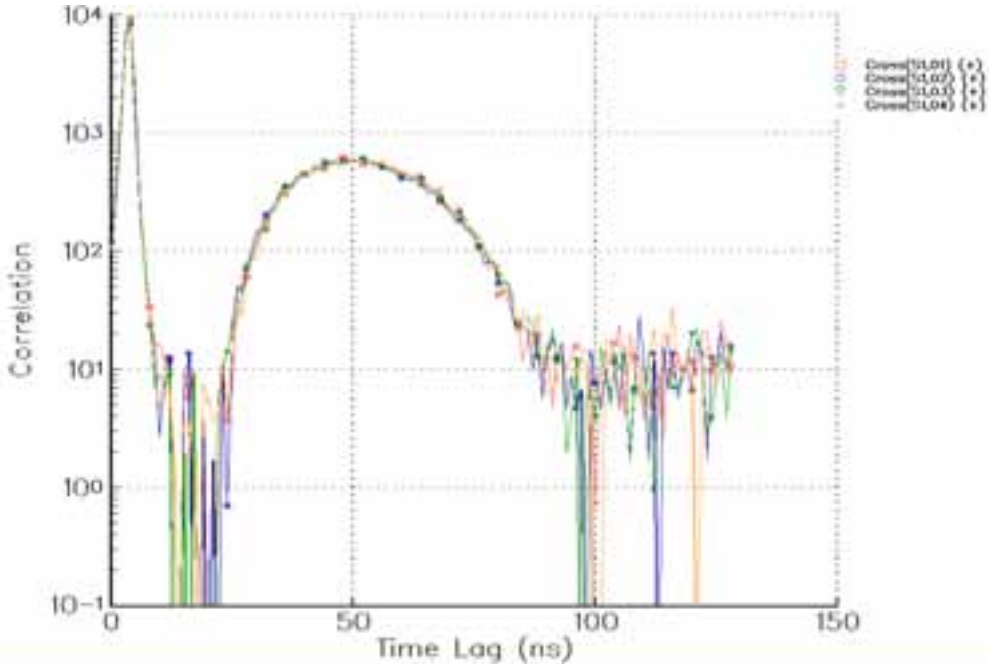


Figure 2: Typical time distribution of counts from a detector after spontaneous fission of Cf. The initial peak is from gammas and the subsequent peak is from the neutron distribution.

side diameter of 8.89 cm, and a height of approximately 14 cm, with a density of approximately 18.8 g/cm^3. The casting was inside a 0.05-cm-thick sealed steel can with an outside dimension of 15.2 cm and a height of 22.9 cm. A photograph of the imaging measurement for the casting on a rotational platform is shown in Figure 3, with the DT generator on the left and the eight 1 × 1 × 6-in.-thick plastic scintillation detectors on the right.

Initially, a vertical scan was performed over the full height of the casting, and the results are shown in Figure 4. A full vertical scan at a single projection angle provides an overall picture of the object to be imaged and allows the user to choose a particular height to image further. While the vertical scan shown in Figure 4 was obtained with high resolution at each level, these vertical scans can often be taken much quicker with fewer pixels and shorter exposures without affecting the ability to locate an area of interest. Vertical scans can be interpreted in the same fashion as x-ray images. The DU casting on top of the rotational platform can clearly be seen in the image. Next, two high-resolution scans were made at one height through the casting. These projections were captured at two rotational positions 90° apart. For both positions of the rotational platform, the 1-in.-square detectors were shifted 1/3-in. for successive measurements until the imaging arc was fully covered. Measuring with finer detail than the 1-in. detector face yields some gain in image resolution, depending in part on the image reconstruction method used.

Figure 5 shows the attenuation values for each of these positions in both projections (0° and 270° rotational positions). For a cylindrically symmetric casting, this 90° rotation will confirm symmetry and determine the position of the casting with respect to the center of rotation. For objects that are not cylindrically symmetrical about the axis of rotation, more projections are required to obtain the shape of the contents of the can. The NMIS analysis software includes a model-based fitting algorithm to determine

Figure 3: Source-uranium metal-casting-detector configuration for imaging.

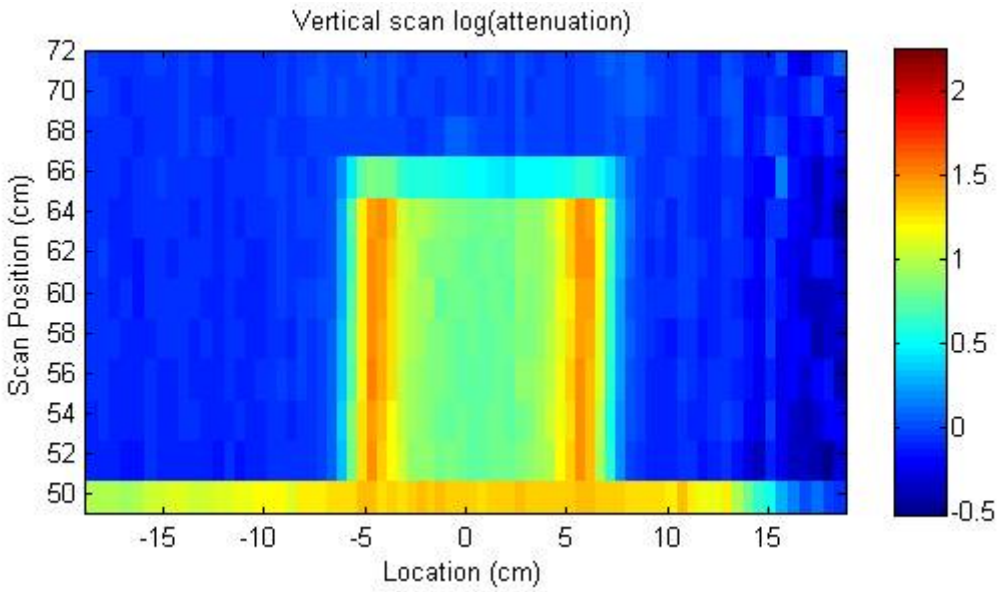


Figure 4: Full scan of DU casting: lag of attenuation versus lateral location and height above the floor.

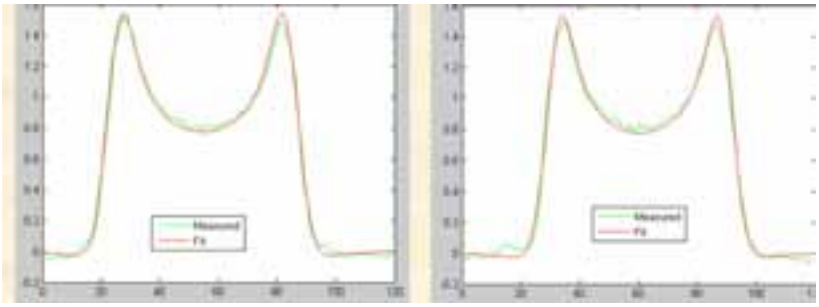


Figure 5: Attenuation projections at 0° and 90° rotational positions and the results of fitting the data.

Parameter	Initial guess	Final fit	Actual dimensions
Inner diameter (cm)	10.00	8.56	8.89
Outer diameter (cm)	11.00	12.93	12.81
Attenuation coefficient (2/cm or cm^{-1})	0.280	0.202	0.280
X offset (cm)	0.00	-0.418	Unknown
Y offset (cm)	0.00	0.570	Unknown

Table 1: Dimensions in centimeters from fit of casting data set containing two projections 90° apart.

geometric dimensions and attenuation coefficient values. This algorithm begins with the user entering an initial geometry of basic shapes from data such as that shown in Figure 4 and selecting key parameters (positions, sizes, and attenuation coefficients), allowed to vary during the fit. The fit uses an unconstrained nonlinear optimization algorithm in conjunction with a simulation tool based on ray tracing to find a fit of the measured projections to those generated by simulating the geometry. The results of fitting the attenuation projections at 0° and 90° rotational positions are shown in Figure 4, and the dimensions obtained are given in Table 1.

The fitted dimensions are close to the actual. These measurements were performed with 1- by 1-in. pixels (detectors). Reduced pixel size would improve the accuracy of the dimensions. The fitted attenuation coefficient (0.202 cm^{-1}) is lower than the actual (0.28 cm^{-1}) because neutrons scattered at small angles from uranium still arrive at the detector and at nearly the same time as the directly transmitted neutrons. With proper correction for scattering, the values of the attenuation coefficients can be fixed for known materials, and more accurate dimensions can be obtained. Based on the X offset in Table 1, the axis of the casting was approximately 0.42 cm

farther from the detector array than the axis of rotation. A second set of experiments was performed on the DU casting by placing shielding around the casting. Two different shielding configurations were measured: (1) a 1-in.-thick lead box surrounding the steel can and (2) a 1-in.-thick lead box plus 4-in.-thick lead plates and bricks added to the front and back as shown in Figure 6.

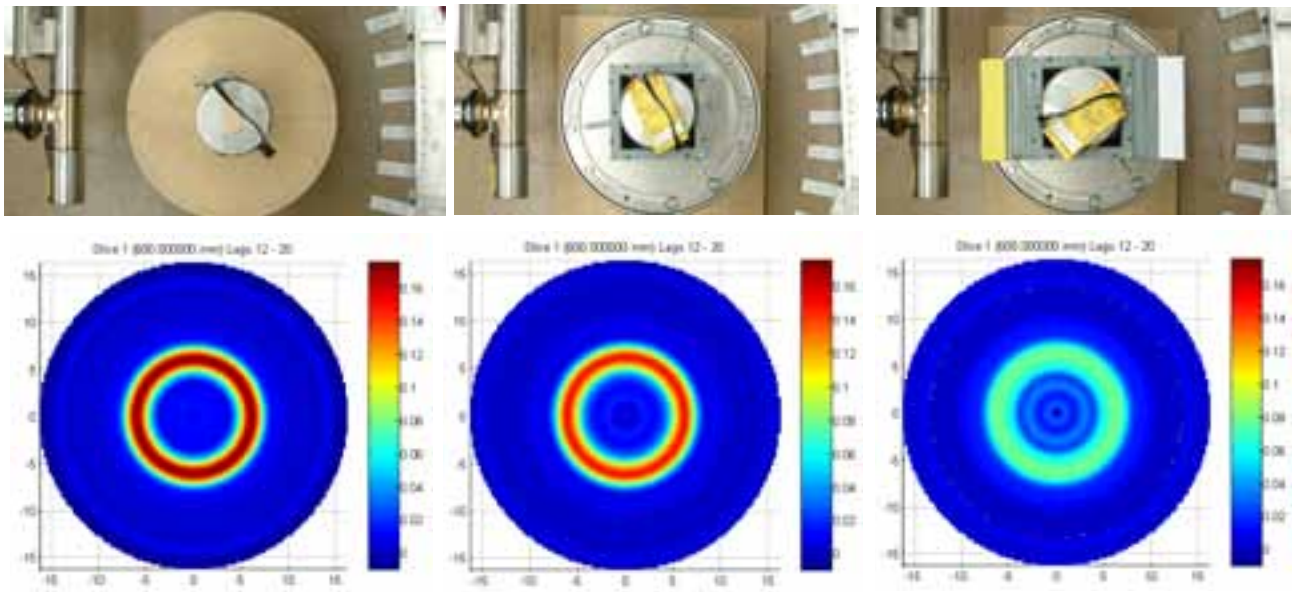
Figure 6: DU casting within a 1-in.-thick lead box surrounding and 4-in. thickness of lead plates in front and back.

In an effort to determine whether the internal components still provide a useful signature, a second set of projections was obtained for just the shielding without the DU casting and used to normalize the projections (I_0). The measurement times for the bare casting and the casting with 2 in of lead were 2 minutes and that for the casting with 10 in of lead was 10 minutes. The images for the different configurations of lead shielding are shown in Figure 7. Based on these results, the casting is still distinguishable with 5-in.-thick lead shielding front and back. Thus, the transmission was measured through 10 in. of lead and the casting. This result illustrates that if the shielding or container material is known and available, a measurement of transmission, I_o, with the shielding or container in place can normalize out the container and shielding effects.

4. Measurement of a 3D object with Cf

Imaging measurements were performed for a variety of objects that were not cylindrically symmetric. In this case the object needs to be rotated to measure the projection at various angles. One such object is shown in Figure 8. The bottom of this object consists of an 8-in-square lead with a 6.5-in.-diam. central hole that is 6.75 in high. Spaced 0.5 in above that is a 0.5-in-thick, 7-in-square piece of Plexiglas. On the Plexiglas is a 3.5-in-high, 6.5-in-OD, 5.5 -in –ID lead part whose outside diameter increases to 7 in. for the top 0.75 in. Inside the top piece of lead and resting on the Plexiglas is a 3.5-in.-diam., 2-in.-high polyethylene cylinder adjacent to the inside surface of the lead. These features of the object are visible in Figure 8. This was a contrived test object used for training to test the ability to image an unknown 3D object. The object was then inserted into a drum and located as in Figure 9 between the Cf source (on the left) and 16 plastic scintillators (on the right) on a radial arm that could be rotated. The object was raised off the bottom of the drum by a low mass metal ring and plate.

The initial measurements were a vertical scan of the object in the container with the arm supporting the detectors rotating laterally to simulate other radial detector positions. The results of these measurements are shown in Figures 10 and 11 where two



(a) Bare DU casting **(b) DU + 2-in. Pb** **(c) DU + 10-in. Pb**

Figure 7: Various shielded casting configurations and resulting images.

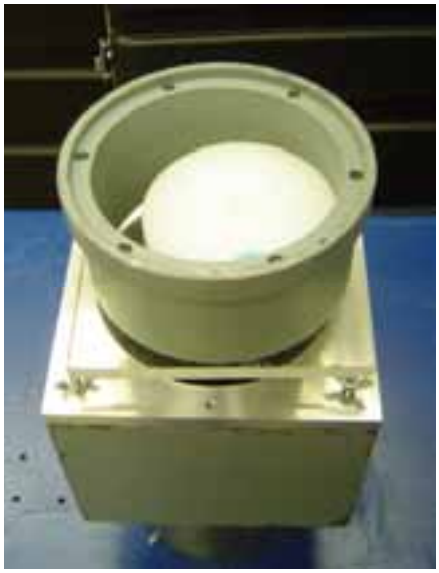


Figure 8: 3D object of lead, aluminum, polyethylene, and Plexiglas.

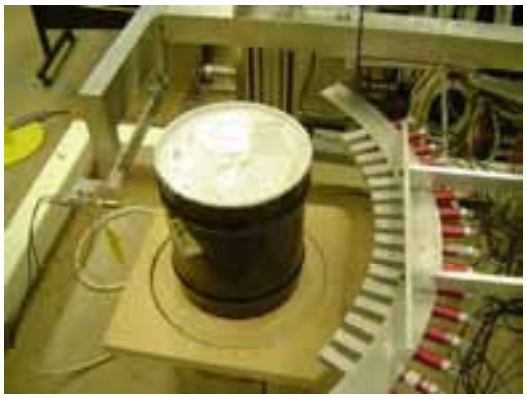


Figure 9: 3D test object in a container on the rotating table between the Cf source on the left and the detectors on the right.

dimensional plots of the attenuation length are presented. The polyethylene and the Plexiglas are clearly visible in the neutron scan whereas they are not in the gamma scan. This is not the case when imaging with a DT generator which is sensitive to both light and heavy materials [2].

Based on these vertical scans, a detailed image was measured at 2 vertical locations through the middle of the lower lead and through the middle of the polyethylene. Since these objects were not cylindrically symmetric, the transmission was measured at 18 different projection angles. The reconstructed images are shown in Figures 12 and 13. Clearly, the configurations of the test object at the 2 heights are determined. Future work in imaging testing on 3D objects will employ an automated object turntable. The use of a DT generator with pixilated alpha detector for this type of measurement will facilitate much better imaging through removal of scattering result in shorter measurement time.

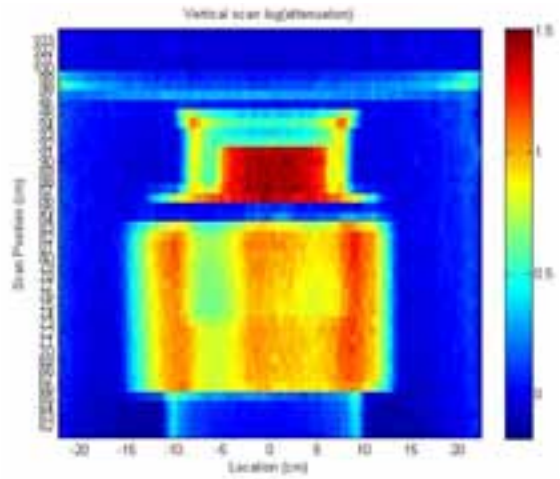


Figure 10: Neutron radiograph of the test object from the vertical scan.

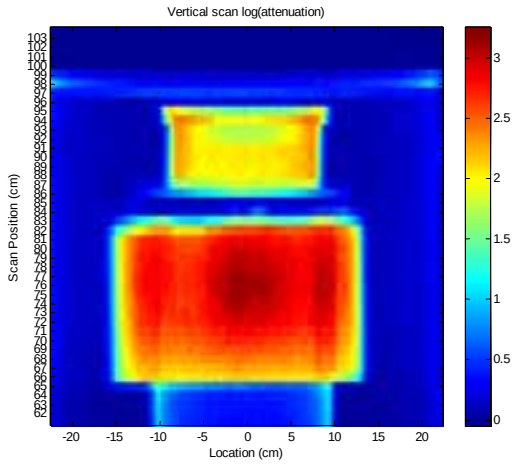


Figure 11: Gamma radiograph of the test object from the vertical scan.

5. Applications

The imaging capability of NMIS can enhance NMC&A:

- Determination of the configuration of items in containers, and it is ideal for fissile material transfers between facilities. This capability is accomplished by providing the detailed configuration of the fissile and non-fissile components of an item under scrutiny. The images can be compared to drawings provided by the shipper avoiding repackaging delays to verify the contents of a shipment that may not be immediately used. Induced fission emitted radiation can be evaluated to distinguish fissile from non fissile materials.
- Periodic inventory confirmation.
- Determination of the form of legacy materials such as HEU in storage cans where the form is not known.
- Determination of unknown configurations of fissile materials for criticality safety and identifica-

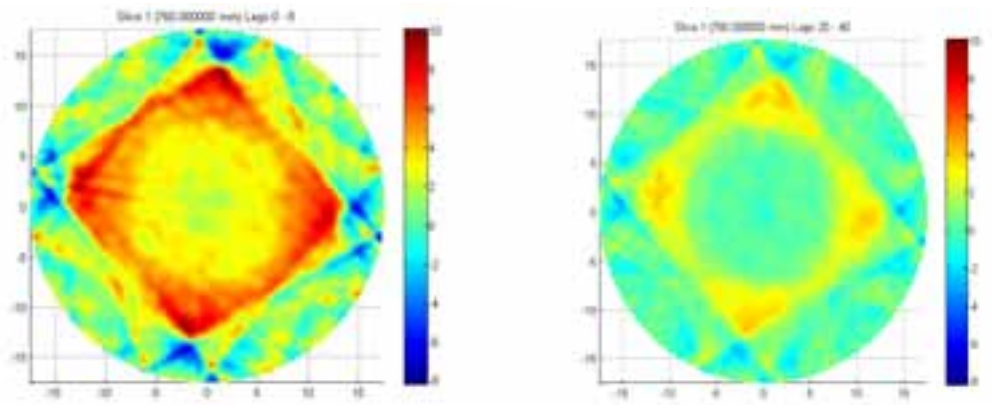


Figure 12: Gamma and neutron images of the test object through the lower lead.

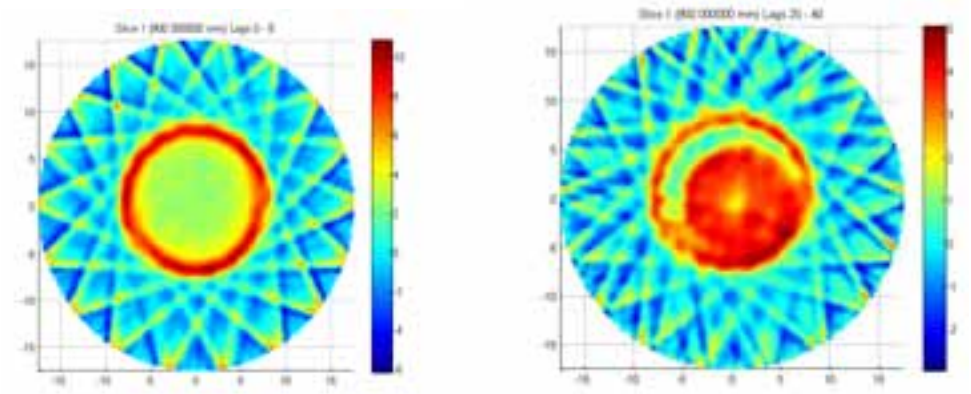


Figure 13: Gamma and neutron images through the test object at the height of the polyethylene.

tion or to determine appropriate standards for other NMC&A measurements.

- Determination of the amount of oxidation of fissile metal in cans.
- Providing more accurate hold up measurements in pipes and process applications where the material and process vessel shape are known and process vessels cannot be emptied. In some cases the density distribution in the container can be measured accurately so that the mass can be inferred to a few percent.

6. Conclusions

The imaging capability of NMIS using a small portable DT generator with an embedded alpha detector can enhance NMC&A for a wide variety of NMC&A applications addressing such problems as fissile transfers, fissile inventory, unknown fissile configurations, oxidation of fissile metal, and hold up in pipes and process vessels, especially when the object is many attenuation lengths thick. However, for low attenuation length objects, a time tagged Cf spontaneous fission source of prompt neutrons and gamma rays from fission may be more useful. This type of imaging has a variety of other uses such as verification of the configuration of nuclear weapons/components shipped or received, warhead authentication behind an information barrier, traceability of both fissile and non-fissile weapons components/parts during dismantlement, and counter terrorism.

7. References

[1] J. T. Mihalczo, J. A. Mullens, J. K. Mattingly, T. E. Valentine, "*Physical description of nuclear materials identification system (NMIS) signatures*," Nucl. Inst. and Meth. A 450 (2000): 531.

[2] J. A. Mullens, "*Addition of Tomographic Capabilities to NMIS,*" Y/LB-16,160, March 2003, http://www1.y12.doe.gov/search/library/documents/pdf/YLB-16160.pdf.

[3] *"API120 D-T Neutron Generator User Manual,"* Thermo Electron Corporation, 5074 List Drive, Colorado Springs, CO 80919, www.thermo.com/neutrongenerators.

[4] J.T. Mihalczo, "*The Use of ACalifornium-252 as a Randomly Pulsed Neutron Source for Prompt-Neutron Decay Measurements*," Nucl. Sci.& Engr., 53, 393-414 (1974)

[5] Wyatt, M.S., et al. 1998. "*Characterization of an Enriched Uranyl Fluoride Deposit in a Valve and Pipe Intersection Using Time-of-Flight Transmission Measurements with ^{252}Cf*," Proc. Inst. of Nuclear Materials Management Meeting, Naples, FL, July 26–30,1998.

Uranium Enrichment Assay with a $LaBr_3$(Ce) Scintillation Detector: A Promising Option for the 2nd Generation of COMPUCEA

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Abstract

A careful evaluation of uranium enrichment measurements with a 2" x 2" $LaBr_3$(Ce) scintillation detector has been carried out in order to test this new type of detector as a room-temperature detector option for future use in the in-field analyses made with COMPUCEA. Spectrum analysis has been made with an adapted version of the NaIGEM analysis code. Comparative performance data obtained from measurements with HPGe, NaI(Tl) and $LaBr_3$(Ce) detectors on low-enriched uranium reference samples are presented.

Keywords: uranium enrichment; gamma spectrometry; lanthanum bromide detector; NaIGEM

1. Introduction

The acronym COMPUCEA (COMbined Procedure for Uranium Concentration and Enrichment Assay) refers to a measurement procedure for the determination of the uranium elemental concentration and ^{235}U isotopic abundance ("enrichment") in relatively small samples of unirradiated uranium product materials. The applied measurement techniques include absorption edge spectrometry for the uranium concentration analysis, and passive high-resolution gamma spectrometry for the ^{235}U enrichment determination. The respective measurements are made on site in European Low-Enriched Uranium (LEU) fuel fabrication plants by analysts from the Institute for Transuranium elements (ITU), Karlsruhe, in support of joint Euratom/IAEA Safeguards inspections during the physical inventory verification (PIV). This measurement support from ITU with COMPUCEA during the PIV campaigns has been provided for more than a decade with the 1st generation of equipment, consisting of a radioisotope-based K-edge densitometer and a HPGe-based high-resolution gamma spectrometer [1, 2, 3].

Recently, ITU started to develop a so-called 2nd generation of equipment for COMPUCEA. One of the objectives for the redesign aimed at replacing the liquid nitrogen-cooled HPGe detectors by room-temperature detectors in order to simplify the equipment for the in-field measurements. This goal has been fully achieved, without loss in performance, for the uranium concentration measurement part of COMPUCEA, where the change from K-edge densitometry to L-edge densitometry allowed to replace the previous HPGe detector by an ultra high-resolution Si detector operated under modest Peltier cooling [4, 5]. As for the enrichment measurement part of COMPUCEA, searches for an adequate substitute for the high-resolution HPGe detector are still under way.

With the recently discovered Ce-doped lanthanum halide (LaCl3, $LaBr_3$) scintillators [6], a new type of scintillation detector is now becoming available, which is opening up good prospects for high-quality enrichment measurements with a room temperature detector. In this paper, we report on a performance study for enrichment measurements with a $LaBr_3$(Ce) scintillation detector, addressing the specific application to small-size uranium sample measurements as encountered in the in-field analyses made with COMPUCEA.

2. Enrichment assay in COMPUCEA

Before describing the application of the new $LaBr_3$(Ce) scintillation detector as a potential detector alternative, it might be helpful to briefly recapitulate the specific measurement conditions and procedures for the enrichment measurements with COMPUCEA. Three measurement conditions ultimately determine the currently applied method for the gamma-spectrometric enrichment measurement:

1. The physical inventory samples provided by the inspectors are of small quantity, typically a single uranium pellet or a few grams of oxide powder. This automatically rules out the classical type of enrichment measurement made in the "infinite thickness" geometry.
2. Since the measurements serve the purpose of accountancy verification, they are calling for the

highest possible measurement accuracy with small to negligible systematic measurement errors. It seemed questionable whether this requirement could be fully met by the alternative measurement option for enrichment measurements based on the intrinsic assay approach [7].

3. The parallel measurement with COMPUCEA for the determination of the uranium element content made by L-absorption edge spectrometry requires a dissolution of the solid uranium samples, because nondestructive measurement techniques allowing a direct determination of the uranium concentration from a solid uranium sample with high accuracy unfortunately do not exist up to date.

With the uranium samples prepared in liquid form, the enrichment measurement with COMPUCEA follows the procedure of counting the most prominent ^{235}U gamma ray at 186 keV from a well-defined volume of sample solution in a well-defined and calibrated counting geometry as shown in Fig. 1. The gamma counting is performed in a well detector to achieve the highest possible detection efficiency. This approach allows accurate enrichment measurements, provided (i) the sample volume is kept constant, (ii) the uranium content in the measurement sample is accurately known, (iii) the matrix of the measurement solution is well controlled, and (iv) the counting set up is calibrated against representative reference solutions.

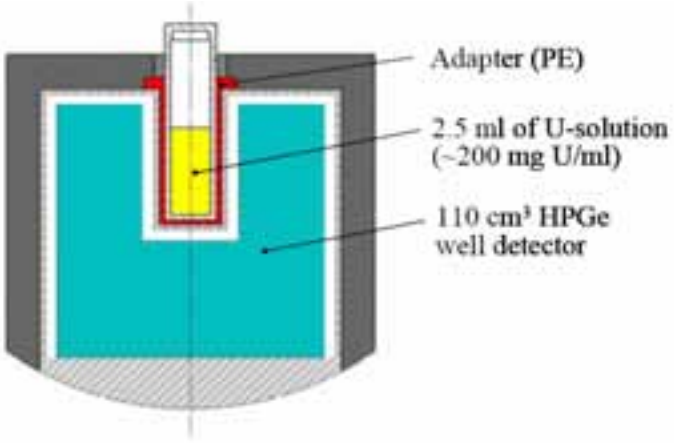

Figure 1: Present counting configuration for the $_{235}U$ enrichment measurement in COMPUCEA with a HPGe well detector.

An exact volume of 2.5 ml of uranium solution, precisely determined with an accuracy of 0.02% from a combined sample weighing and density measurement, is transferred into a cylindrical plastic vial with an inner diameter of 11 mm for counting in the well detector. The uranium concentration of the solution, and hence the amount of uranium in the measurement sample, is known from the parallel densitometry measurement of COMPUCEA with an accuracy better than 0.2%. The uranium concentration and the nitric acidity of the solution matrix are established within narrow limits – through strict procedures followed during sample dissolution and subsequent dilution – at nearly constant values of 200 mgU/ml and 3M HNO3, respectively. Small corrections for gamma self attenuation are applied for samples slightly deviating from these nominal values. These corrections also take into account the additional self attenuation effects caused by gadolinium, if present. Information on the Gd content will in the future be obtained from the L-edge densitometer operated in the XRF mode [4].

The type of detector used up to now during the in-field measurements is a 110 cm^3 HPGe well detector with a 16 mm diameter x 40 mm deep well in the detector cap. For 3 different units of this type of detector, the detection efficiency for 186 keV gammas in the counting configuration shown in Fig. 1 was determined to 26.4, 28.3 and 31.6%. This yields count rates between 57 and 68 cps per % enrichment for the typical amount of 0.5 g of uranium contained in the 2.5 ml of sample solution. This rate necessitates a counting time of about 20-25 minutes to reach a measurement precision of 0.2% for a sample enrichment of 3%.

For highest measurement accuracy and reliability, enrichment measurements made with the high-resolving HPGe detector undoubtedly offer the best choice. Practical drawbacks for the in-field measurements, however, are the need for liquid nitrogen for detector cooling, and the relatively long cooling times for the well-type detector. For the given HPGe well detector it takes about 24 hours before the detector has sufficiently cooled down to reach the nominal energy resolution. During the recent re-design of the COMPUCEA equipment towards a more compact and ready-to-use type of instrumentation we have therefore also evaluated detector alternatives, which do not require cooling.

The use of CdZnTe solid state detectors has been ruled out a priori because of their small volumes and corresponding low detection efficiency. Realistically, this leaves only the use of a suitably dimensioned scintillation detector as a viable detector option. In a first approach we have carried out a performance assessment from in-field measurements with a 3” x 3” NaI(Tl) well detector, which with its 16 mm well diameter offered the same counting geometry as the HPGe well detector. Despite the much poorer

energy resolution of the NaI detector, the results obtained showed acceptable performance [5].

It is a fortunate situation that, with the recent discovery of the new lanthanum halide scintillators, the gamma spectroscopy community is now provided with a promising new type of room temperature scintillation detector with distinctly improved performance compared to the well-known NaI(Tl) detector.

3. Features of $LaBr_3$(Ce) scintillation detectors

The new cerium-doped lanthanum chloride, LaCl3(Ce), and lanthanum bromide, $LaBr_3$(Ce), scintillation detectors, manufactured by Saint-Gobain Ceramics & Plastics Inc. and now commercially available under the registered trademarks BrilLanCe®350 and BrilLanCe®380, respectively [8], have the potential of significantly enhancing the performance and capabilities of scintillator-based gamma spectroscopic systems in many areas. The application to ^{235}U enrichment measurements for Safeguards purposes as discussed in this paper represents just one example.

The production of larger-sized lanthanum halide crystals has rapidly progressed during the last 2-3 years. Standard detector sizes up to 3" x 3" are now available, although detector prices are still relatively high (~200 € /cm^3). Existing performance data show about 20-30% better energy resolution for the lanthanum bromide than for the lanthanum chloride detectors. We therefore chose the former one for our studies. Some of the noteworthy practical advantages of the new $LaBr_3$(Ce) scintillator, compared to the traditional NaI(Tl) scintillation detector, are:

- Improved energy resolution;
- About 10 times faster light output decay, enabling high count rate applications;
- High temperature stability;
- Higher gamma stopping power and detection efficiency.

Table 1 lists comparative values for some pertinent physical properties of $LaBr_3$(Ce) and NaI(Tl) detectors. Comparative data for the energy resolution are given in Table 2. The quoted energy resolutions were measured for the actual 2" x 2" $LaBr_3$(Ce) detector used in the present study, and for a NaI(Tl) detector of same dimensions with a nominal energy resolution of 6.8% at 662 keV. The right-hand column in Table 2 gives the improvement factor for the energy resolution offered by the $LaBr_3$(Ce) detector.

Better energy resolution for the lanthanum bromide detector is obtained over the whole energy range covered, with the improvement factor increasing with increasing gamma energy. For the 186 keV gamma ray, the lanthanum bromide detector offers two times better energy resolution than the NaI detector.

Property	$LaBr_3$(Ce)	NaI(Tl)
Crystal density (g/cm3)	5.29	3.67
Thickness for 50% attenuation of 662 keV gammas (cm)	1.8	2.5
Light yield (Photons/keV)	63	38
1/e decay time (ns)	16	250
Temp. coefficient of light output (%/°C between 0° and 50°C)	<0.02	-0.3

Table 1: Comparison of scintillator detector properties (from [8]).

Energy (keV)	FWHM energy resolution (%)		Improvement factor for $LaBr_3$(Ce)
	$LaBr_3$(Ce)	NaI(Tl)	
29	13.58	20.10	1.48
59	9.80	14.65	1.50
88	7.55	11.88	1.57
186	4.87	9.71	1.99
662	2.68	6.50	2.43
1001	2.09	5.46	2.61
2614	1.34	3.55	2.65

Table 2: Measured energy resolutions for 2" x 2" aBr3(Ce) and NaI(Tl) detectors.

The spectral appearance of gamma spectra from low-enriched uranium is displayed in Fig. 2. The figure shows spectra for a typical COMPUCEA sample (0.5 g of uranium with an enrichment of 4.4% in 2.5 ml of nitric acid) taken with 3 types of detectors: the 110 cm^3 HPGe well detector so far routinely used in COMPUCEA, a 3"x3" Na(Tl) well detector, and a 2"x2" $LaBr_3$(Ce) detector. The measurement example produced with the lanthanum bromide scintillation detector shows a uranium gamma spectrum, in which the main ^{235}U gamma rays (143, 163, 186, 205 keV) are reasonably resolved.

A peculiarity of the lanthanum-based scintillators is the presence of an inherent background originating from the electron capture and β-decay of the long-lived minor La isotope ^{138}La (t1/2 = 1.05x10^{11}y) to ^{138}Ba (66.4%) and ^{138}Ce (33.6%, see schematic decay scheme in Fig. 3). The Ba X-ray peak visible in the $LaBr_3$ spectra in Figs. 2 and 3 results from the EC decay of ^{138}La to ^{138}Ba (the single X-ray peak near 37.4 keV in fact represents the accumulated

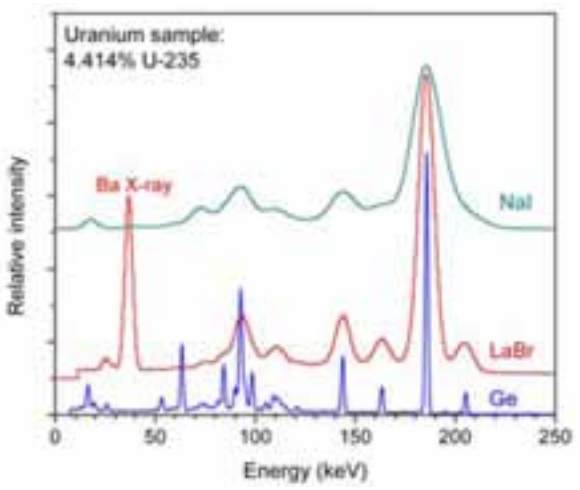

Figure 2: Comparison of HPGe, LaBr3(Ce) and NaI(Tl) gamma spectra from a low-enriched uranium sample.

energy given off by the X-rays in the EC process). The β-decay to ^{138}Ce, in turn, manifests itself in a beta continuum background with an endpoint energy of 255 keV as shown in Fig. 3. This continuous beta background occurring in the energy range of the ^{235}U gamma rays normally turns out to be negligible for enrichment measurements on bulk uranium samples, but it may notably contribute (about 20-30%) to the Compton and Bremsstrahlung background from the high-energy ^{234m}Pa gamma rays in enrichment measurements on small quantities of uranium (1 g or less as in the case of the COMPUCEA measurements). This is illustrated in Fig. 3, which shows the magnitude of the internal beta continuum background in relation to the gamma spectrum produced by a 5 ml uranium solution sample with 1 g of natural uranium (kept at 1 cm distance from the surface of a 2" x 2" $LaBr_3$ detector). Fortunately, even in the case of the low-activity COMPUCEA samples, the added detector background component in the energy range up to 250 keV is properly accounted for in the spectrum evaluation and has proved to have a negligible effect on the performance of the enrichment assay (Note: An elevated detector background due to radiations from the decay of ^{138}La also occurs in higher energy regions, outside of the region of interest for enrichment measurements).

It should be noted that for the efficient detection of 186 keV gamma rays lanthanum bromide crystals do not need to be thicker than about 2-2.5 cm. A 2 cm thick $LaBr_3$ crystal doped with 5 wt% Ce stops 95% of 186 keV photons. A standard crystal thickness of 1" is therefore more than adequate for enrichment measurements. The reduced detector volume would also reduce the internal detector background.

4. Application to enrichment measurements on COMPUCEA samples

Sample counting in a well counter as routinely practiced so far in the COMPUCEA measurements with the HPGe well detector not only offers high detection efficiency, it also represents a counting configuration being least sensitive to measurement variabilities such as sample positioning or small dimensional variations of the sample containers holding the uranium solutions. Since lanthanum bromide detectors are not yet available in the form of well detectors, a different counting geometry as shown in Fig. 4 had to be chosen for the test measurements with the $LaBr_3$(Ce) detector.

The sample-detector configuration represents a compromise between closest possible sample-to-detector distance for maximum detection efficiency, and desirable larger sample distance for increased immunity to variations in sample and container dimensions. To partly compensate for the somewhat

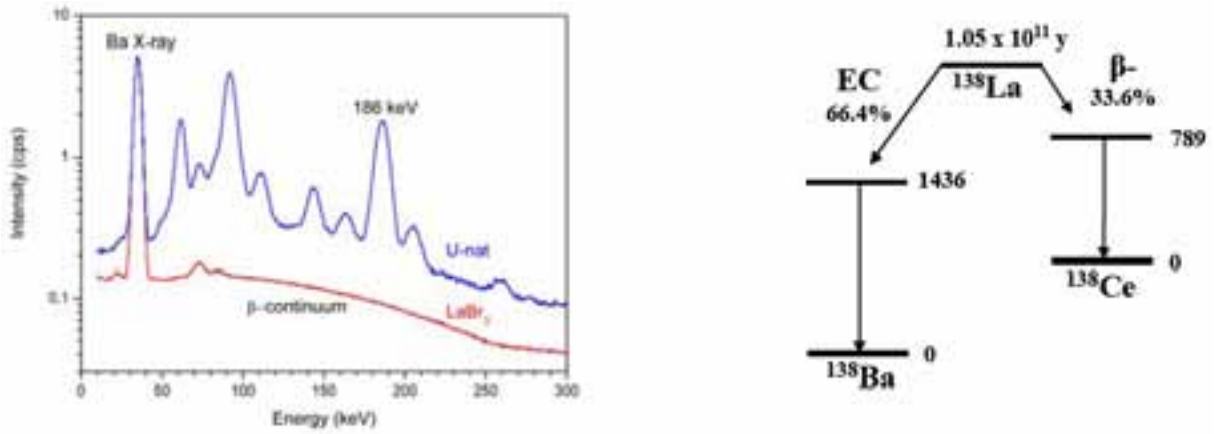

Figure 3: Magnitude of internal background from a 2" x 2" LaBr3 detector in comparison to the gamma spectrum from a 1 g U-nat sample.

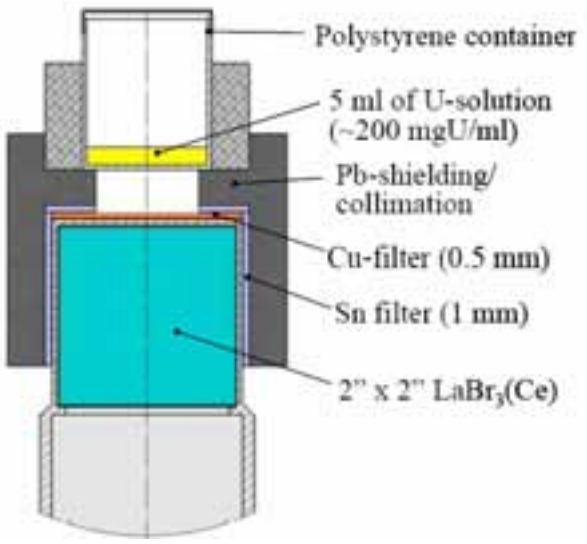

Figure 4: Counting configuration used for the measurements with the LaBr3 detector.

lower detection efficiency, the volume of the uranium solution was increased from 2.5 ml to 5 ml. For the polystyrene sample container with an inner diameter of 36 mm this yields a 5 mm thick solution layer. The sample was viewed by the detector through a 10 mm high, 30 mm diameter Pb collimator and a 0.5 mm thick Cu filter.

Table 3 compares the count rates for the 186 keV gammas obtained with the configuration shown in Fig. 4 with the corresponding rates measured in the standard 110 cm^3 Ge well detector of COMPUCEA, and with an alternatively tested 3" x 3" NaI well detector. It is obvious that the less efficient counting geometry of Fig. 4 provides lower peak count rates than the well-type configurations. For the same amount of uranium the counting efficiency decreases by about a factor of 4. A counting configuration with significantly enhanced detection efficiency, however, could be realized with available $LaBr_3$ detectors in the form of a sandwich-type configuration, incorporating, for example, two 2" x 0.75" $LaBr_3$ detectors as sketched in Fig. 5. Such a configuration would provide a relatively robust and efficient counting geometry, yielding 186 keV count rates of about 150 cps per % enrichment from a 5 ml sample as indicated in the right-hand column of Table 3. Graphs showing the expected measurement precision from this 186 keV count rate in dependence on counting time and enrichment are given in Fig. 6.

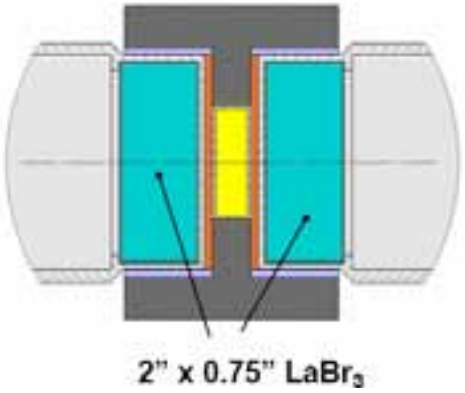

Figure 5: Proposed sandwich detector configuration for enhanced detection efficiency. Sample volume 5 ml with 1g of U.

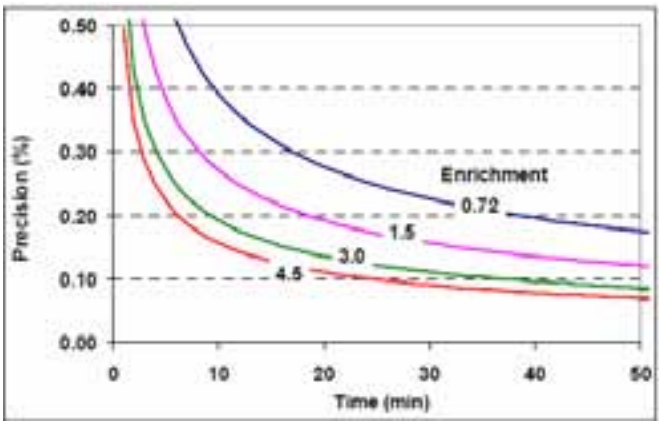

Figure 6: Expected counting precision for the configuration of Fig.5.

5. Spectrum analysis

The method for spectrum analysis applied to the $LaBr_3$ spectra was adopted from that used in the NaIGEM code for ^{235}U enrichment measurements with NaI detectors [7]. In this method, response profiles are computed for each of the components contributing to the 130 – 290 keV region of the spectrum that is analyzed. These responses are then fit to the observed data by the method of least-squares. Two of the components, of course, are spectrum

Detector/ Configuration	110 cm^3 HPGe well (Fig. 1)	3" x 3" NaI well (Fig. 1)	2" x 2" $LaBr_3$ planar (Fig. 4)	2" x 0.75" $LaBr_3$ sandwich (Fig. 5)
Sample vol. (ml)	2.5	2.5	5	5
Amount U (g)	0.5	0.5	1	1
Filter	1 mm PE	1 mm Ti	0.5 mm Cu	0.5 mm Cu
186 keV (cps) per % enrichment	60, 65, 72 (rates from 3 units)	109	36	~ 150 (expected)

Table 3: Comparison of 186 keV peak count rates from different measurement configurations.

profiles for ^{235}U and ^{238}U. However, other profiles are generated to account for small shifts in the spectrum gain and changes in the detector resolution that may have occurred.

The peak structure in the 130 – 290 keV region is primarily due to ^{235}U gamma rays. The principal gamma ray peaks of ^{235}U are: 143.76, 163.37, 182.57, 185.72, 194.94, 202.1, and 205.33 keV. Although gamma rays from ^{238}U and its daughters are very weak in this region, their Compton scattering and Bremsstrahlung radiations do contribute significantly to the background continuum. In the case of the $LaBr_3$ detector the internal background produced by the detector material itself contributes to the background.

The principal shape of the gamma ray peaks for both the $LaBr_3$ and NaI detectors is described by Gaussian functions. The ^{235}U response profile is generated using known gamma-ray branching intensities that have been modified to account for attenuation by filters and other materials and by the relative detection efficiency. The equation describing the "background" continuum contains two components. The first accounts for the "step" function associated with the different background levels in front of and after the peaks. The other is a polynomial equation used to describe the shape of the Compton plus Bremsstrahlung continuum associated with the decay of ^{238}U and its daughters, and with the internal background continuum of the $LaBr_3$ detector. The method of analysis is iterative. Convergence must first be reached to account for changes in gain and resolution before a final calculation is made to determine the intensity of the ^{235}U response. An example showing the final fit of the principal ^{235}U response from the $LaBr_3$ detector is given in Fig. 7. The fitting graph in Fig. 8 shows another fitting option in which the 186 keV peak is isolated from the rest of the ^{235}U response. The $LaBr_3$ spectral responses displayed in the figures, if compared with the corresponding response from a NaI detector shown for comparison in Fig. 9, best illustrate the kind of spectral improvement gained with a lanthanum bromide detector.

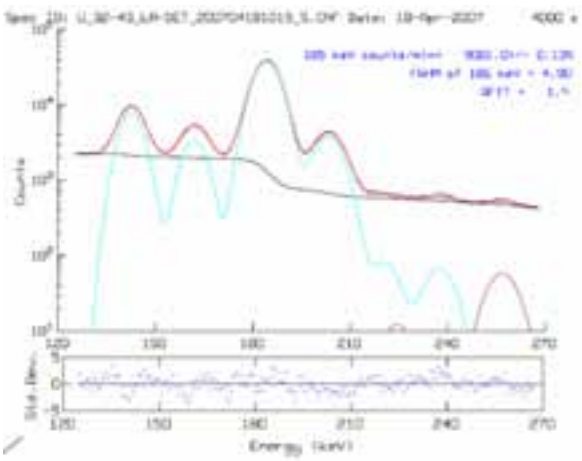

Figure 7: LaBr3 graph fitting the principal ^{235}U response.

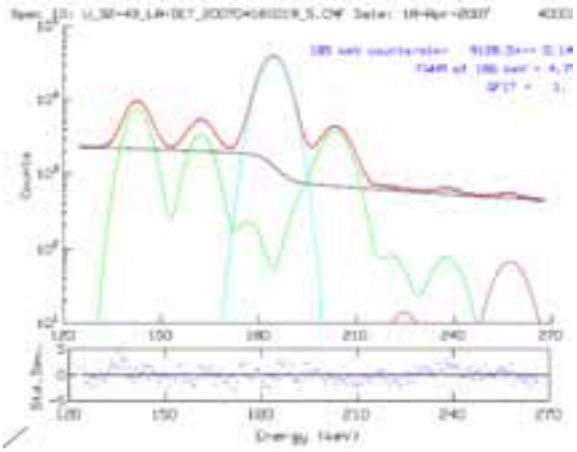

Figure 8: Another fitting version for LaBr3 spectra where the 186 peak is isolated from the rest of the ^{235}U response.

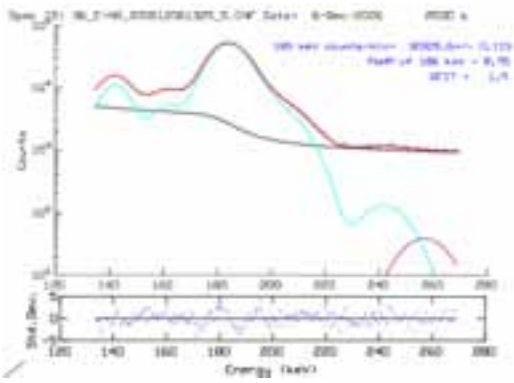

Figure 9: Fitting graph for a NaI spectrum.

6. Performance

In a perfect enrichment measurement, the registered 186 keV peak count rates are expected to be strictly proportional to the ^{235}U isotopic abundance. This feature, if realized, not only simplifies calibration, but also helps to reduce and/or to avoid systematic measurement errors. Achieved strict proportionality primarily proves the ability of the applied method for spectrum analysis to extract unbiased net peak counts from the measured gamma spectra, irrespective of given spectral features like, for example, the degree of energy resolution or changing peak-to-background ratios.

In order to verify the assumed proportionality, we have carried out high-precision measurements on a set of carefully characterised uranium reference solutions with 6 different grades of ^{235}U enrichment (0.3158, 0.7202, 1.3802, 2.1039, 3.2469 and 4.3138

wt% ^{235}U). Comparative measurements were made with 3 different units of the standard COMPUCEA HPGe well detector, with a 3" x 3" NaI well detector, and with the 2" x 2" $LaBr_3$ detector. From each of the 6 reference solutions 2.5 ml aliquots were transferred into the cylindrical sample vials used for the counting in the well detectors (Ge and NaI), and 5 ml aliquots were loaded into the polystyrene sample containers for the counting with the $LaBr_3$ detector in the configuration shown in Fig. 4. The combined uncertainties for the uranium concentration (from gravimetry), for the ^{235}U isotope abundance (from mass spectrometry), and from sample weighing yielded a total uncertainty of 0.12% (1s) for the amount of ^{235}U contained in the samples with enriched uranium. For the samples with depleted (0.3158%) and natural uranium the accuracy of the ^{235}U content was limited to 0.30% and 0.15%, respectively, by the respective uncertainties of the mass spectrometry measurements for the ^{235}U abundance.

For all gamma measurements, the statistical counting error for the 186 keV gammas was kept below 0.1%. Since the reference solutions did not have exactly the same uranium concentration (ranging between 193 and 201 mgU/ml), small corrections for gamma self attenuation had to be applied to the measured 186 keV count rates. The differential change of the count rate as a function of uranium concentration around a reference concentration of 200 mgU/ml has been determined from measurements made on samples of same enrichment but different uranium concentration. The observed differential changes are listed in the 1st row of Table 4. Another correction made to the measured 186 keV peak rates accounted for small contributions of gamma rays from ^{234}Pa and ^{234m}Pa to the 186 keV line from ^{235}U. The respective corrections amount to 0.38% and 0.16% for the depleted and natural uranium samples, and to less than 0.1% for the enriched uranium materials.

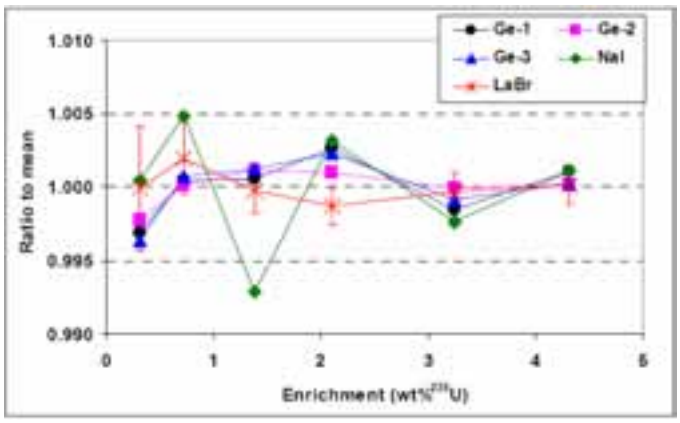

Figure 10: Normalized 186 keV count rate as a function of $_{235}$U enrichment.

The derived values for the corrected 186 keV count rates per mg ^{235}U, relative to the mean value of the results for the 6 different enrichments, are plotted in Fig. 10. For the sake of clarity, error bars (1s) are only given for the data set obtained with the $LaBr_3$ detector. The same uncertainties also apply to the other data sets (in numbers: 0.32% for the depleted uranium sample, 0.18% for U-nat sample, and 0.13% for the enriched uranium samples). Numbers for the weighted mean values and the weighted standard deviations of the 6 measurement results per detector are given in Table 4.

From a visual inspection of Fig. 10, and from the derived values for the weighted standard deviations, we conclude: (i) the desired proportionality between 186 keV peak count rate and enrichment indeed exists, and (ii) the performance of the $LaBr_3$ detector favourably compares with the performance of the HPGe detectors.

Quantity	Ge-1	Ge-2	Ge-3	NaI(Tl)	$LaBr_3$(Ce)
Self attenuation correction (% per change of 1 gU/l)	0.063	0.068	0.063	0.055	0.038
Weighted mean (186 keV counts/s per mg U-235)	12.057	13.085	14.451	22.830	3.575
Weighted standard deviation (%)	0.124	0.0759	0.125	0.435	0.092

Table 4: Pertinent measurement data from linearity test.

Another aspect investigated separately concerns the question of measurement reproducibility for the counting configuration chosen for the $LaBr_3$ detector (Fig. 4). We had noted that the disposable polystyrene sample containers used in this measurement showed some variations in the bottom thickness of the order of up to 0.1 mm, which at the given sample–to-detector distance of 11 mm will notably affect the effective solid angle. For this reason only sample containers showing a uniform bottom thickness within ± 0.01 mm were selected for the enrichment measurements. The results of a reproducibility test, consisting of measurements on 7 different sample containers holding the same uranium solution, is shown in Fig. 11. Taking into account the average statistical counting error of 0.06% per individual measurement, the results suggest a measurement reproducibility of about 0.12%.

7. Outlook

We have demonstrated from our investigations that gamma spectrometry with a lanthanum bromide scintillation detector, if used in conjunction with the modified NaIGEM code for spectrum analysis, allows high-accuracy enrichment uranium measure-

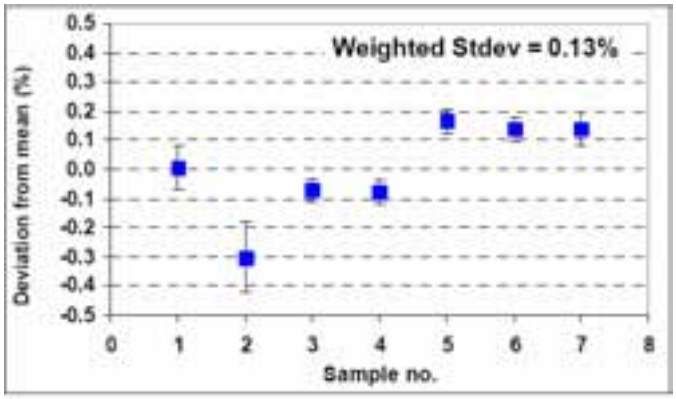

Figure 11: Results of reproducibility measurements on 7 samples of same enrichment.

ments under the conditions of the COMPUCEA analyses. We are therefore planning to apply this type of detector in the 2nd generation of the COMPUCEA equipment, meeting then our initial design goal of having an instrumentation operating only with room-temperature detectors. Also with a $LaBr_3$ detector it appears realistic to keep the systematic measurement uncertainty for enrichment measurements at the level of 0.2% as set by the international target values for the present COMPUCEA measurements with a high-resolution HPGe detector [10].

References

[1] P. Matussek, I. Michel-Piper, H. Ottmar, "A Compact Densitometer for Uranium", Proc. 13th ESARDA Symp., Avignon, France, 14-16 May, 1991, 329.

[2] P. Matussek, I. Michel-Piper, H. G. Wagner, B. Brandalise, O. Cromboom, "First In-Field Experience with COMPUCEA – A Portable Assay System for the Combined Determination of the Uranium Concentration and 235U Abundance in Uranium Solutions by NDA Methods", Proc. 15th ESARDA Symp., Rome, Italy, 11-13 May, 1993, 415.

[3] P. Matussek, Ph. Laurent, W. Janssens, F. MacLean, U. Blohm-Hieber, "Routine Use and Recent Developments of the COMPUCEA Instrument", Proc. 19th ESARDA Symp., Montpellier, France, 13-15 May, 1997, 313.

[4] H. Ottmar, H. Eberle, S. Abousahl, N. Albert, H. Schorlé, "Design of COMPUCEA 2nd generation for simplified in-field uranium inventory verification", Proc. 27th Annual ESARDA Symp., London, UK, 10-12 May 2005.

[5] S. Abousahl, N. Albert, P. Amador, H. Eberle, H. Ottmar, H. Schorlé, "Performance and validation of COMPUCEA 2nd generation for uranium measurements in physical inventory verification", Proc. Intern. Safeguards Symposium on Addressing Verification Challenges, International Atomic Energy Agency, Vienna, Austria, October 16-20, 2006.

[6] E. V. D. van Loef, P. Dorenbos, C. W. E. van Eijk, H. U. Gudel, K. W. Kraemer, Applied Physics Letters 77 (2000) 1467-1469.

[7] R. Gunnink, "MGAU: A brief review of the method and some results", Proc. International MGA Users' Workshop, 19-20 Oct. 1994, IRMM, EC-JRC, Geel, Belgium, EUR 16163 EN (1995) 179.

[8] http://www.detectors.saint-gobain.com.

[9] R. Gunnink, R. Arlt, R. Berndt, "New Ge and NaI Analysis Methods for Measuring 235U Enrichments", Proc. 19th Annual ESARDA Symp., Montpellier, France, 13-15 May, 1997, 431.

[10] H. Aigner et al. "International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Materials", ESARDA Bulletin 31, April 2002, 39.

Fluorine as a Safeguards Tool for Age Dating of Uranium Oxyfluoride Particles?

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Abstract

When uranium hexafluoride (UF_6) is released into the environment, it reacts with the atmospheric moisture forming uranium oxyfluoride particles and hydrogen fluoride (HF). Fluorine-containing compounds such as HF are recognized as signatures for enrichment activities. In this study, the connection between the fluorine in the particles and the age of the particles has been investigated. Establishing this link is not evident, as uranium oxyfluoride particles are highly hygroscopic and little is known about their reaction mechanisms and stability. There are various environmental factors such as time, humidity and light or heat exposure that could affect the amount of fluorine. Even after the particles are collected, the fluorine could decrease further due to sample preparation or storage conditions.

The influence of ultraviolet light exposure combined with the time elapsed after preparation has been investigated on uranium oxyfluoride particles prepared at the Institute for Reference Materials and Measurements (EC-JRC-IRMM) using an aerosol deposition chamber. The presence of fluorine in particles was confirmed by energy-dispersive X-ray analysis (EDX). Semi-quantitative information was obtained by evaluating specific peak height ratios obtained by secondary ion mass spectrometry (SIMS). With these measurements, possible correlations existing between the amount of fluorine and the age of a particle could be investigated, taking into account the environmental conditions to which the uranium oxyfluoride particles were exposed.

Keywords: safeguards; uranium hexafluoride; uranium oxyfluoride particles; SEM-EDX; SIMS

1. Introduction

Environmental sampling, as introduced in 1996 as part of routine INFCIRC-153 safeguards agreements and strengthened since the late 1990's by the Additional Protocol (INFCIRC-540) of the International Atomic Energy Agency (IAEA), has proven to be an important safeguards tool in the verification of the absence of undeclared nuclear activities [1, 2]. By wiping surfaces in or around nuclear facilities, using small pieces of cotton cloth called swipes, uranium-bearing particles are collected among millions of other dust particles. The analysis of these uranium particles can reveal key information on a site's current and past activities. Although the emphasis is currently on the analysis of the uranium isotopes, a lot of information can be deduced from the particle's morphology, surface structure, crystallinity or elemental composition [3].

Uranium-bearing particles from swipe samples taken at uranium enrichment facilities often contain fluorine. Although these particles may result from deposits that are formed when adsorbed water and uranium hexafluoride (UF_6) interact, many of them result from the gaseous release of small amounts UF_6. Initially, uranium oxyfluoride (UO_2F_2) particles are formed when UF_6 reacts with atmospheric moisture. However, there are several variables that affect the particle composition and the amount of fluorine present. These include exposure to humidity, heat and light, both during and after particle formation, in addition to the time elapsed since particle formation.

In this study, we investigated whether the analysis of fluorine in UO_2F_2 particles could be used to supplement information on the uranium isotopic composition. In particular, if a correlation could be found between the amount of fluorine and the age of the particle, the measurement of fluorine in particles would not only provide information on the likely source of the particles, i.e. enrichment activities, but also on their history.

The particles formed from uranium hexafluoride are highly hygroscopic, and little is known about their long-term stability. Before being collected on swipes they may have been exposed to high temperatures, a high humidity or sunlight. All of these factors could have altered their morphology and composition and, more specifically, their fluorine content. Any study of the fluorine content of particles originating from UF_6 must therefore take into account the environmental parameters that influence the amount of fluorine. In this work, the effects of ultraviolet light and storage time on the fluorine content of particles

prepared under controlled conditions were examined.

2. Materials and Methods

UO_2F_2 particles were prepared using an aerosol deposition chamber developed at the Institute for Reference Materials and Measurements (EC-JRC-IRMM) (Fig.1). When released into the aerosol deposition chamber, the UF_6 reacts with the atmospheric moisture to form hydrogen fluoride (HF) and UO_2F_2 particles [4]:

$$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF$$

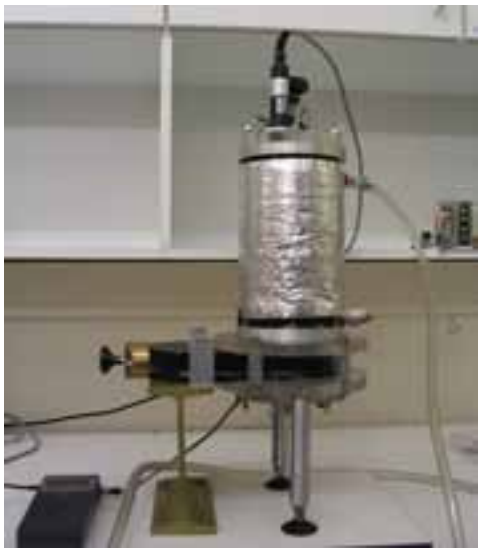

Figure 1: Aerosol deposition chamber designed at the IRMM for the preparation of UO_2F_2 particles from the controlled hydrolysis of UF_6.

The UO_2F_2 particles were collected on graphite planchets of 25 mm diameter (Schunk, Germany) at the base of the deposition chamber. It has been demonstrated that the humidity of the air inside the deposition chamber is a determining factor for the particle morphology [4-6]. A dry atmosphere, of less than 15 % relative humidity, results in submicron particles that agglomerate to larger structures of up to 100 micrometers. In contrast, when the relative humidity of the air exceeds 60 %, the particles become more spherical and the degree of agglomeration is lower. Using a high humidity to limit agglomeration, particles of diameters between a few hundred nanometers and 2.5 micrometers were collected, with individual particles separated by a few micrometers (Fig. 2). Such particles were well-suited for direct analysis.

As part of the original preparation procedure, the particles were heated for at least 6 hours in an open furnace at 350 °C to remove excess water and other volatile elements. This heat treatment inevitably removes most of the fluorine in the particles. As a consequence, any existing information between the amount of fluorine and the age of a particle is lost. The heat treatment was therefore omitted from the preparation procedure and freshly prepared particles were measured directly after preparation by SEM-EDX.

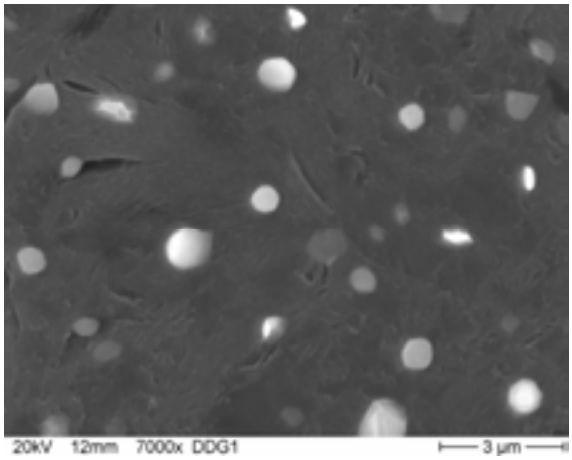

Figure 2: Scanning electron image in secondary electron mode showing uranium particles (white dots) produced in high humidity conditions (> 60 %) on a graphite planchet.

A FEI Quanta 200 3D scanning electron microscope equipped with an Oxford Si(Li) energy-dispersive X-ray detector (SEM-EDX) was used to locate the uranium particles and to determine their elemental composition. The electron acceleration voltage was set to 10 kV.

SIMS ion-microprobe measurements were carried out to assess the effect of ultraviolet light and storage time on the relative amount of fluorine on a set of 14 particle samples. For every sample, SIMS ion-microprobe measurements were applied to between 4 and 7 particles. All but one sample contained UO_2F_2 particles: one sample contained UF_4 particles stored in a laboratory environment for more than 2 years. This UF_4 sample was used to test the repeatability of SIMS UF_4 spectra and their quantitative distinctiveness from UO_2F_2. The storage time for the UO_2F_2 particle samples varied between 2 weeks and 29 months. One sample was exposed to ultraviolet light for 3 weeks.

At QinetiQ in Malvern (UK), a Cameca IMS 4f SIMS spectrometer was used with 8.5 keV O_2^+ primary ion bombardment at a current of 2 nA in a focused spot (estimated diameter 10 µm). The data were obtained by cycling the masses 238 (U), 239 (UH), 254 (UO), 257 (UF), 270 (UO_2), 273 (UOF) and 276 (UF_2) 10 times each to reveal any ratio variations with sput-

tering. The mean values of these ion intensity measurements were evaluated relative to the intensity at mass 238 (U).

3. Results

The signature of freshly prepared particles was recorded using EDX, to serve as a reference level. Spectra of these freshly prepared particles clearly showed the fluorine K_α line at 0.677 keV, in addition to the U M_α (3.17 keV) and M_β (3.34 keV) X-ray lines (Fig. 3). The fluorine K_α line was still detected in the EDX spectrum after 3 weeks of ultraviolet light exposure. In contrast, particles subject to a heat treatment of 350 °C for 6 hours, as originally foreseen in the particle preparation procedure, did not show the characteristic fluorine K_α line (Fig. 4). Carbon and oxygen were detected in all EDX spectra. The carbon peak was attributed to the graphite planchet substrate, whereas the oxygen peak was partially attributed to the particles, partially to surface contamination and residual gases in the specimen's chamber.

SIMS ion-microprobe measurements were carried out, providing greater sensitivity than EDX in measuring fluorine, in a range of samples subjected to various exposure and storage conditions. The 254 (UO) signals were inherently strong for all samples due to the oxygen primary ion beam that was used. In general, the $^{254}(UO)/^{238}U$ ion ratio measurements were fairly reproducible, with a variation of a factor 3 between samples. The $^{270}(UO_2)/^{238}U$ ratio generally tracked the $^{254}(UO)/^{238}U$ ratio.

In contrast to the uranium oxide ions, the fluorine-containing uranium ions did show significant differences between the sample types, the storage times and the degree of exposure to ultraviolet light [7]. For samples stored in the laboratory for between 11 and 16 months, the $^{257}(UF)/^{238}U$ ratio was about 10 times lower than for samples that were only 2 months old. Similar decreases were observed for the $^{273}(UOF)/^{238}U$ and $^{276}(UF_2)/^{238}U$ ratios, which generally tracked the $^{257}(UF)/^{238}U$ ratio, although the variations in the $^{276}(UF_2)/^{238}U$ ratio were much more pronounced.

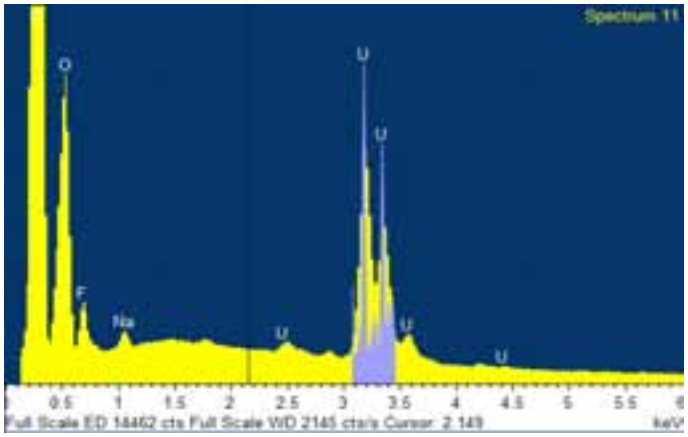

Figure 3: Energy-dispersive X-ray spectrum of a uranium-bearing particle prepared by the aerosol deposition chamber showing the U M_α (3.17 keV) and M_β (3.34 keV) X-ray lines, in addition to the fluorine K_α line at 0.677 keV. The other peaks in the spectrum were attributed to oxygen, carbon and sodium.

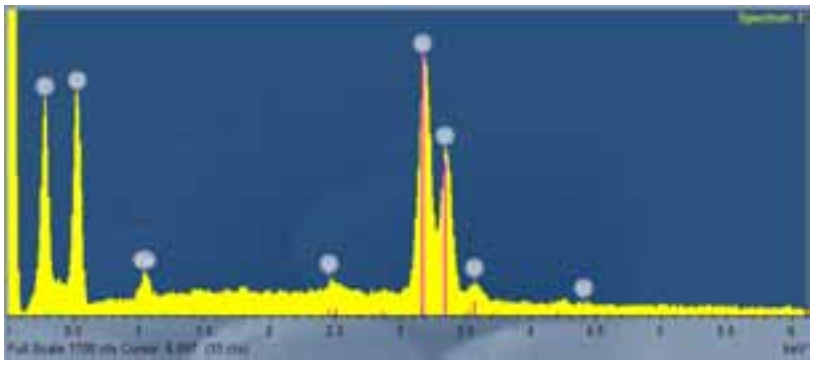

Figure 4: Energy-dispersive X-ray spectrum of a uranium-bearing particle prepared by the aerosol deposition chamber and heat-treated at 350 °C for 6 hours showing the U M_α (3.17 keV) and M_β (3.34 keV) X-ray lines, in addition to carbon, oxygen and sodium. The fluorine K_α line at 0.677 keV line could no longer be detected in this spectrum.

To assess the reduction in the level of fluorine due to exposure to ultraviolet light, particles on a graphite planchet were exposed to ultraviolet light for 3 weeks. Although the sample was only 2 months old at the time of measurement, the $^{257}(UF)/^{238}U$ ratio had fallen to the level observed for particles that were stored for almost 1 year. This demonstrated that exposure to ultraviolet light significantly accelerated the particle ageing process.

Although the SIMS measurements showed distinct differences between samples with different storage or exposure conditions, large particle-to-particle variations within the same sample were also observed. These were attributed either to variations in the particle morphology or to SIMS measurement effects related to the ionisation and detection efficiencies. In some cases, a uniform film of uranium was detected instead of particles. The fact that the UO_2F_2 particles are highly hygroscopic could explain this observation.

5. Conclusion and outlook

The aerosol deposition chamber produces micrometer-sized uranium oxyfluoride particles from the controlled hydrolysis of UF_6. These were used to study the effect of storage time and ultraviolet light exposure on the relative amount of fluorine in particles. A link between the relative amount of fluorine and the age of a particle was established, although large particle-to-particle variations within the same sample did occur. Planned transmission electron microscopy (TEM) and Raman measurements will give us more insights into the particle ageing processes. The results of a further study will be reported elsewhere [7].

The observation that the relative amount of fluorine in single particles is an indicator of the time since deposition, and possibly of the environmental influences, could be important in interpreting the data obtained from analysis of particles collected during safeguards inspections.

6. Acknowledgements

The authors acknowledge A. J. Pidduck, M. R. Houlton and J. D. Mace of QinetiQ Malvern, UK for the SIMS measurements and the valuable discussions undertaken within the UK Support Programme to the IAEA.

Special thanks to S. Van den Berghe and A. Leenaers of the Belgian Nuclear Research Centre (SCK•CEN) for the SEM-EDX measurements. The authors would also like to thank O. Marie and F. Pointurier of the Commissariat à l'Energie Atomique (CEA) Bruyères-le-Châtel, France for their support. Many thanks to P. Van Espen, M. Moens and W. Dorriné for the SIMS and SEM measurements at Antwerp University.

7. References

[1] Donohue, D.L.; *Strengthening IAEA safeguards through environmental sampling and analysis*; Journal of Alloys and Compounds; Elsevier; 1998; p 11-18.

[2] Hedberg, P.M.L.; Ingeneri, K.; Watanabe, M.; Kuno, Y.; *Isotopic measurements of U particles by secondary ion mass spectrometry (SIMS)*; Proceedings of the Institute of Nuclear Materials Management Annual Meeting, Phoenix, AZ USA; 2005.

[3] Stebelkov, V.; *Informativeness of morphology analysis of uranium microparticles from industrial dust at nuclear plants*; Proceedings of the GLOBAL 2005 conference, Tsukuba, Japan, 2005.

[4] Pickrell, P.W.; *Characterization of the solid, airborne materials created by the interaction of UF_6 with atmospheric moisture in a contained volume*; Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tennessee, 1982, p 1-88.

[5] Kips, R.; Leenaers, A.; Tamborini, G.; Betti, M.; Van den Berghe, S.; Wellum, R.; Taylor, P.D.P.; *Characterization of uranium particles produced by hydrolysis of UF_6 using SEM and SIMS*; Microscopy and Microanalysis; Cambridge University Press; in publication; 2007.

[6] Bostick, W.D.; McCulla, W.H.; Pickrell, P.W.; *Sampling, characterization and remote sensing of aerosols formed in the atmospheric hydrolysis of uranium hexafluoride*; Journal of Environmental Science and Health A20; Taylor and Francis; 1985; p 369-393.

[7] Kips, R.; Pidduck, A.J.; Houlton, M.R.; Leenaers, A.; Mace, J.D.; Marie, O.; Pointurier, F.; Stefaniak, E.A.; Taylor, P.D.P.; Van den Berghe, S.; Van Espen, P.; Van Grieken, R.; Wellum, R.; *The analysis of fluorine in uranium oxyfluoride particles as an indicator of particle age*; Submitted to Spectrochimica Acta Part B: Atomic Spectroscopy

Nuclear Forensic Methods in Safeguards

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Abstract

The introduction of strengthened safeguards, the implementation of the additional protocol (INFCIRC 540) and the nuclear material intercepted from illicit trafficking led to a more investigative character of analytical measurements. The more specific questions will be asked with respect to a given sample, the more investigative analytical methodologies will be required and the more thorough, interpretative and comparative evaluation of results needs to be done. Specific applications, often in combination with only minute amounts of sample call for methods of high sensitivity, low detection limits, high selectivity and high accuracy. Consequently, the new sample types triggered the transfer of analytical techniques from the environmental area, materials science and geological or cosmological area to the safeguards community. The selection of the method or combination of methods is done according to the sample and information required. Data interpretation is calling for reference information, comparison samples and thorough understanding of the processes taking place throughout the nuclear fuel cycle. Environmental analysis and nuclear forensic science have experienced during last ten years significant developments in the mentioned area which safeguards can now profit from.

Keywords: strengthened safeguards, nuclear forensics

1. Introduction

Measurements of nuclear material were the backbone of the verification measures in the early days when the safeguards agreements, INFCIRC 153 and the Euratom regulation 3227/76, were implemented. Consequently, measurement methods were put in place, which provided information on the uranium, plutonium or thorium content, as well as U and Pu isotopic compositions in a given material. These measurements served the verification of declared amounts of nuclear material. Apart from verification of the nuclear material accountancy, the information inherent to the nuclear material was never exploited.

When the International Atomic Energy Agency (IAEA) started introducing strengthened safeguards and the additional protocol was implemented, the mandate of the IAEA expanded from the verification of correctness of a state's declaration to comprise also the completeness of such declarations. The detection of undeclared nuclear activities or materials requires establishing a comprehensive picture of a state's nuclear activities and checking the consistency of the declarations against other evidence. In consequence, a tremendous need for information at different levels arises in order to enable the evaluation required.

All types of information sources can be drawn upon: e.g. satellite imagery, design information verification, on-site inspections and sample taking (comprising nuclear material samples and environmental samples). In the present paper, we discuss the challenges the strengthened safeguards approach brings along, as well as we describe how methodologies that were initially developed for nuclear forensic use could be exploited in safeguards.

2. Challenges

2.1. Information

Verification of the absence of undeclared nuclear material or activities is very complex task. The answer needs to be composed of a variety of indicators, which allow drawing conclusions on the completeness of state's declaration. The nuclear material and environmental samples taken, provide a useful source of information on the processes applied. Let us recall in this context two main prerequisites:

1. The production and processing of nuclear material leaves (inevitably) traces in the environment. Highly sensitive measurement techniques as applied in the IAEA's Environmental Sampling programme make use of this fact. Depending on the cleanliness of the process and on the quality of the installations, the amount of detectable traces can be rather small. Many years of experience gained in environmental sampling and, in particular in the analysis of single particles has demon-

strated the power of this methodology. The main limitations of particle analysis are caused by the tiny amounts (few pico grams or even less) of material available in micrometer-sized particles. Moreover, the measurement of minor isotopes in individual particles suffers from poor precision (due to counting statistics) and from molecular interferences.

2. Every production process leaves characteristic patterns in the material. These measurable parameters vary as a function of starting material, process parameters, reagents used, storage conditions or vessel materials. The complexity of the data and the interrelations between individual parameters require a careful step-by-step approach from measurement to data interpretation.

The information obtained through the analysis of nuclear material may be divided into two categories: **endogenic** data, i.e. data that is self explaining (e.g. the $^{235}U/^{238}U$ ratio pointing at the enrichment of the material and the intended use), and **exogenic** data, i.e. data that can only be understood with the help of reference data (e.g. comparison against data from known material or from model calculations). The latter type of information is certainly more difficult to understand and requires more resources before a conclusion can be drawn. Chemical impurities, isotopic composition of the nuclear material, isotopic composition of accompanying elements and microstructure are data which are accessible through measurements and which allow to build information. The information measurements and the respective data interpretation provide are expected to prove (or disprove) the absence of undeclared nuclear activities. The conclusion to be reached at the end of this evaluation process is based on “four C’s”:

- **Consistency** of information
- **Coherence** between samples or materials
- **Conformity** of findings with declared processes
- **Comparison** of data

In contrast to traditional safeguards, such an evaluation is not based on quantities of material, but rather on certain qualities of material such as impurities, age, stable isotopes and microstructure.

2.2. Measurement

The challenge in performing measurements of investigative character is twofold: first, a wide spectrum of parameters needs to be measured; and secondly, those parameters providing the most significant information need to be identified. The instrumental techniques applied for this purpose are well established, e.g. mass spectrometry, electron microscopy, anion chromatography. However, the analytical methods need to be adapted to the specific requirements of investigative safeguards analysis. For developing such methods, one can benefit from experiences made in other fields of science, e.g. in nuclear forensics, isotope geology or material science.

2.3. Data evaluation

In order to properly evaluate the measurement data, the availability of reference information is required, in particular for exogenic data. To some extent the safeguards community can draw upon experience and use the data available in the geochemical community. Variations in the isotopic composition of the chemical elements have been studied in other contexts and in some cases cadastral registers of isotopic data are available (e.g. $n(^{18}O)/n(^{16}O)$ ratio in rainwater or lead isotopes in natural lead). Information related specifically to nuclear material is, however, less widely available, e.g. data on metallic impurities in nuclear fuels are often subject to commercial confidence. In order to make best use of the additional information obtained through the methods, a comprehensive set of reference data or of reference samples (i.e. samples obtained from known sources and produced though known processes from known starting materials) needs to be established. In addition, a multidisciplinary team of analysts is required as the information arises from diverse scientific areas covering chemistry, physics and material science.

3. Characteristic parameters

3.1. Isotopic patterns of U and Pu

For long time the safeguards community has made use of the isotopic composition of nuclear material. Increased attention to the minor abundant isotopes in uranium (^{234}U and ^{236}U) was paid only after the introduction of strengthened safeguards, when the need arose to establish capabilities for distinguishing between samples of (apparently) the same enrichment. The isotope abundances of ^{234}U and ^{236}U may help to verify coherence between different samples and consistency with declared operations. The presence of small amounts of ^{236}U will indicate a contamination with recycled uranium and hence point at reprocessing activities. However, also in natural uranium variations in ^{236}U as well as in ^{234}U abundances have been recorded [1]. At ^{236}U abundance levels close to natural abundance (i.e. $n(^{236}U)/$

Isotope	Isotopic Composition [Mass%]		
	Q1.1	Q1.2	Q1.3
^{234}U	0.0159 (8)	0.0158 (8)	0.0158 (8)
^{235}U	0.3480 (70)	0.3501 (70)	0.3406 (68)
^{236}U	0.1383 (41)	0.1396 (42)	0.1361 (41)
^{238}U	99.497 (99)	99.494 (99)	99.507 (99)
^{238}Pu	1.316 (26)	1.315 (26)	1.321 (26)
^{239}Pu	59.66 (60)	59.61 (60)	59.87 (60)
^{240}Pu	28.19 (42)	28.25 (42)	28.06 (42)
^{241}Pu	5.30 (10)	5.29 (10)	5.32 (10)
^{242}Pu	5.51 (11)	5.52 (11)	5.42 (11)

Table 1: Isotopic composition of uranium and plutonium in a seized sample containing radioactive liquor. Measurement uncertainty UC (k=1) is given in brackets and refers to the last two digits of the isotope abundance.

$n(^{238}U) < 10^{-9}$) more sophisticated instrumentation, like Accelerator Mass Spectrometry is required. This technique is available only in few specialized laboratories. In addition, different enrichment processes may result in slight differences in the ^{234}U abundance.

The isotopic composition of uranium and plutonium also allow drawing conclusions on the reactor type in which the material has been irradiated. Table 1 shows the results of isotope abundance measurements (three sub-samples) on a sample seized in the context of a criminal investigation. Comparing the measured values to burn-up calculations, it has to be noted that uranium and plutonium are not originating from the same reactor type: plutonium shows an isotopic composition close to an LWR reactor, while the uranium isotopic composition points at natural uranium fuelled research reactor.

The isotope correlation technique was used in safeguards in 1970's for two reasons: to verify the consistency of the isotopic analyses performed at the reprocessing plants, and to deduce the amount of specific isotopes by measuring other isotopes and using established correlations. Lately it was also adapted to nuclear forensics. In particular the isotopic composition of plutonium is a useful indicator of the reactor type in which the nuclear material was produced. The neutron capture cross-section of the individual plutonium isotopes vary as a function of neutron energy. In consequence, the build up of plutonium isotopes is different in reactors with different neutron energy spectrum. In addition, the initial enrichment of ^{235}U is various in different reactors. These two parameters are reflected in the isotopic composition of plutonium. Knowing the plutonium isotopic composition, we can draw conclusions on the reactor type, where the Pu is coming from (Fig.1).

3.2. Age determination

Age determination of nuclear materials makes use of the radioactive decay of these elements. Assuming a complete separation of the daughter products during the production process (e.g. during chemical purification of the material), we can determine the "age" (i.e. the time that has elapsed between the last chemical treatment of the material and today) of the material by quantifying the amounts of parent and daughter nuclides. Age determination of plutonium is classically being performed by gamma spectrometry using the $^{241}Pu/^{241}Am$ parent/daughter ratio. However, in a few cases it has been noticed that the Am separation has not been complete, thus the age from this parent/daughter may give a wrong answer. The use of the uranium daughters of ^{238}Pu, ^{239}Pu and ^{240}Pu offers a consistency check [3], as these three parent/daughter relations should result

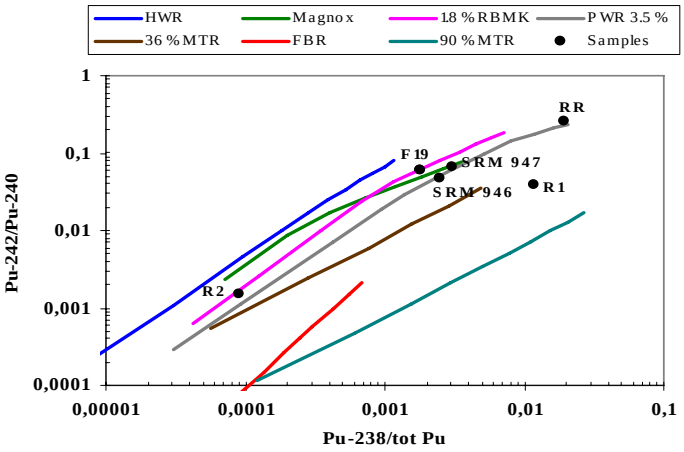


Figure 1: Pu isotope correlation for different types of reactors [2]. SRM 946 and SRM 947 (both are NBS certified Pu reference materials) originate apparently from pressurized water reactors, as well as sample RR used in a round robin exercise. F19 and R2 denote seized materials, which can be attributed to an RBMK reactor.

in the same age – provided the separation of uranium was complete during processing of the material.

Residual amounts of uranium isotopes will lead to biased results in the Pu age determination. The degree of the bias is dependent on the Pu composition (weapons or reactor Pu) as well as on the parent/daughter relation. Fig. 2 shows the relative biases for the worst and best cases of Pu materials. The bias is a function of the age of the material (the older the material, the more U is produced and the less any residual uranium will affect the result) and of the amount of residual uranium after the last chemical separation of the plutonium (the more residual uranium is left in the plutonium sample, the higher the bias will be). As is seen from the model calculations, the parent daughter ratio can be very sensitive to residual amounts of uranium and thus lead to significant biases in the age determination (Fig. 2b). The data in the model calculations were obtained by combining burn-up calculations, decay calculations and isotope mixture calculations.

Another interesting aspect in the age determination, especially in the safeguards context, is the question of the age of particles. Age determination of plutonium particles has been demonstrated earlier [5]. Age determination of uranium particles proves to be much more challenging, due to the very long half-lives of the uranium isotopes ^{234}U and ^{235}U. Even if the ^{234}U is the lower abundant isotope in U materials, due to the 1000-fold shorter half-life the parent daughter ratio ^{234}U/^{230}Th is more favourable ratio for the age determination of the uranium than is the ^{235}U/^{231}Pa ratio.

The particles of interest in swipe samples from enrichment plants are typically only one micrometer in diameter. Based on this assumption, we can calculate the detection limit for the age determination as a function of the age of the particles and the ^{235}U enrichment. Assuming further a detection efficiency of 0.5% (i.e. for detection of 10 ions we need 2000 atoms) in the secondary ion mass spectrometer, we see from Fig. 3 that age determination can only be successfully performed for particles of highly enriched uranium.

3.3. Metallic impurities

Metallic impurities are present in nuclear material samples at varying concentration levels. In starting materials (e.g. ore concentrate) the impurities may have the character of accompanying elements and are present in relatively high concentrations. In intermediate products (e.g. yellow cake) the concentration of most of the chemical impurities has been drastically reduced. After this, towards the final

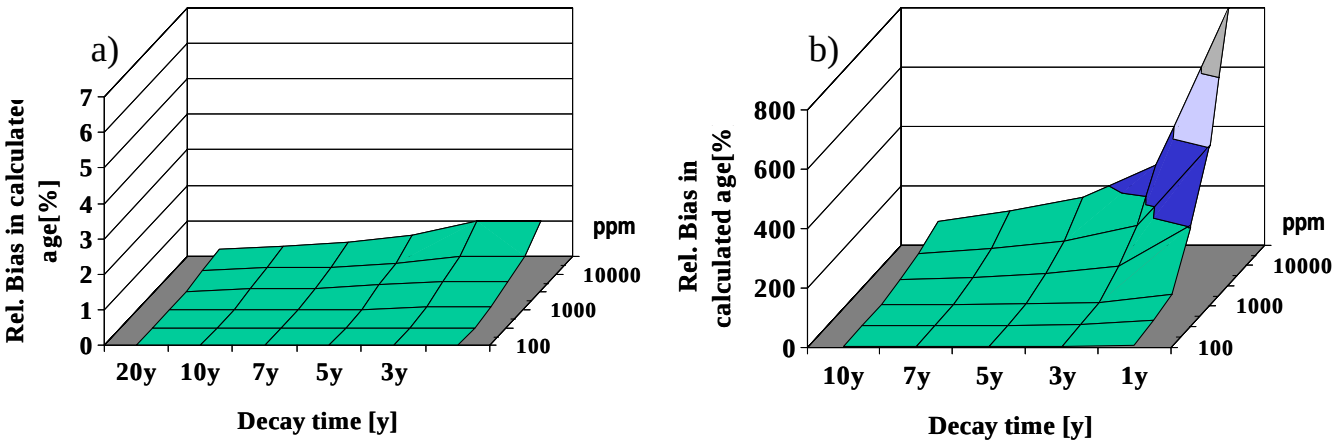


Figure 2: Relative bias in the age of reactor grade Pu (a) and weapons grade Pu (b) using the ^{238}Pu/^{234}U parent daughter ratio as a function of the age and of the amount of residual uranium [4].

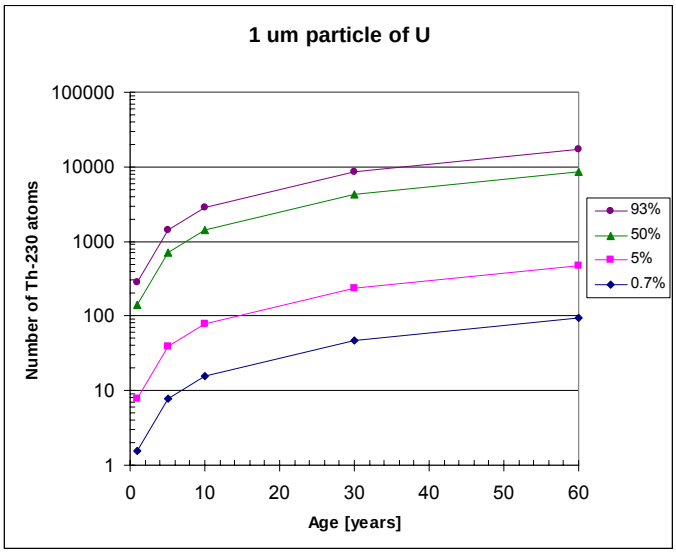


Figure 3: The decay of ^{234}U produces ^{230}Th. The number of ^{230}Th atoms contained in a uranium oxide particle (with an assumed number of 10^{10} atoms of uranium) depends on the age of the particle and the initial enrichment.

product, further decrease of impurities is minute if any. Figure 4 shows metallic impurities in natural uranium compounds of different origins. Five samples from the same origin can be clearly recognized through their identical pattern of metallic impurities.

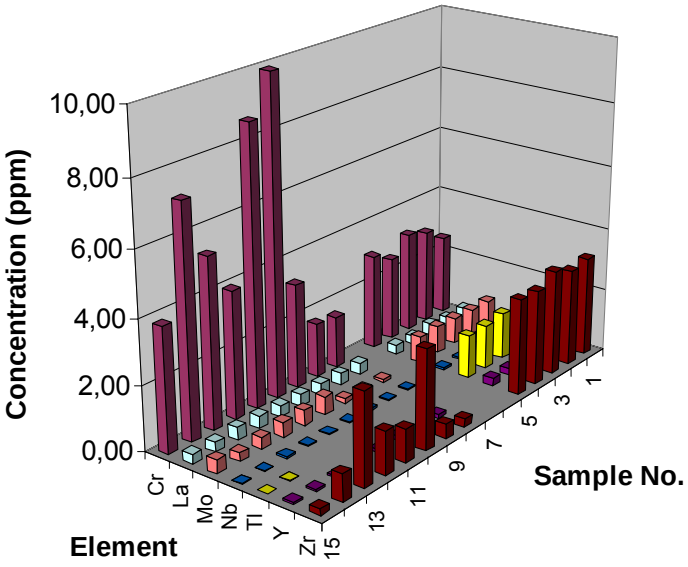


Figure 4: Selected metallic impurities in intermediate natural uranium products. Samples 1 to 5 are apparently of the same origin.

Although metallic impurities can be used for identifying coherences between samples or batches of material, the systematics behind the impurity patterns are not well understood. This is because the metallic impurities may be carried into the material at different stages of the process. The concentration of some impurities may for instance vary as a function of exposure time to the container material or the storage tank, as they are leached from the surface of the walls. In sample analysis the concentration of such elements appears to be fluctuating randomly. One should, in general, avoid the use of common elements as indicators. Another solution to this dilemma could be, instead of looking at the absolute concentrations of impurities to look at ratios of chemical elements. While the absolute concentration of the impurities may change, the ratio of certain elements will vary only within narrow limits. This applies in particular for elements of similar chemical behaviour, e.g. the rare earth elements [6].

3.4. Stable isotopes

In the field of food science and geochemistry, analysis of stable isotopes (e.g. ^{1}H, ^{2}H, ^{12}C, ^{13}C, ^{16}O, ^{18}O) have been successfully applied for a few decades. The principle of the use of stable isotopes is very straightforward: The stable isotope compositions of elements, which are part of a substance, are a function of the origin and history of that substance. That is, two substances which are chemically the same may have different stable isotope compositions if either their origin and/or history differ. This methodology was also introduced recently to nuclear forensics.

The application of oxygen isotope ratio measurements for geolocation purposes has been demonstrated several years ago [7]. A correlation between the geographic location of the production site of uranium oxide samples and the variation in the $n(^{18}O)/n(^{16}O)$ could be established. Moreover, it could be shown that the method is also applicable to individual particles, i.e. the oxygen isotope ratios established by “bulk” measurements using thermal ionisation mass spectrometry (TIMS) could be reproduced on individual particles using secondary ion mass spectrometry (SIMS) [8]. This type of information does obviously not identify a specific plant, yet it provides a parameter for attributing the material to a region. This can be utilised for instance to distinguish between imported and domestic materials.

Another parameter that has been widely used in geochemistry and in environmental sciences is the isotopic composition of lead. Lead isotopes may be primordial (natural lead) or they may be produced through the decay of uranium isotopes. The small variations in the isotopic composition of natural lead have been used to locate the origin of some fuel additives (mainly consisting of tetra-ethyl lead). The adaptation of this methodology for nuclear safe-

guards and nuclear forensics purposes has been studied [9]. It could be shown that the lead isotopic composition of yellow cake provides useful information to distinguish between natural uranium materials of different origins. As lead is omnipresent in our environment, special care has to be taken when performing the chemical separation of the lead from the uranium samples in order not to introduce any natural lead from dust particles or chemical reagents and thus bias the results.

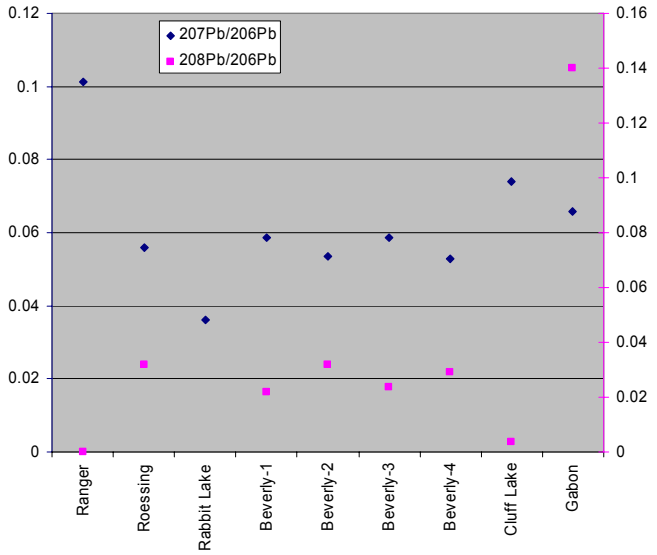


Figure 5: Lead isotope ratios observed in yellow cake samples from different mines [9]. Combined uncertainties, U_c (with k=2) on the ratios $\boldsymbol{n}(^{207}Pb)/\boldsymbol{n}(^{206}Pb)$ and $\boldsymbol{n}(^{208}Pb)/\boldsymbol{n}(^{206}Pb)$ are between 0.000 2 and 0.000 08, thus too small to be visualized on the above graph.

Lead is often used as shielding material for nuclear samples. This may introduce bias in the results as natural lead from the shielding cross-contaminates the lead contained in the sample. There are two possibilities for dealing with this problem: first, one :can correct for all contributions from natural lead using the ^{204}Pb as pilot isotope. ^{204}Pb is not contained in radiogenic lead, and may therefore serve as indicator for the amount of natural lead present in a sample. The second option requires the availability of a reference sample from a suspected origin. In this case isotope mixture calculations can be performed, assuming a binary mixture between natural lead and the lead contained in the reference sample. An example is given in Table 2, where a seized uranium ore sample had been wrapped in a lead foil. The isotope mixture calculation showed that the measured isotopic composition can be fully explained by a binary blend of natural lead and the lead (as measured before) in uranium ore from Joachimsthal (Czech Republic).

Sample	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb
Find-25	1.20	33.27	19.32	46.20
Nat. Pb	1.4	24.1	22.1	52.4
Joachimsthal	0.96	45.12	16.56	37.36
Mixture 56/44	1.21	33.36	19.67	45.78

Table 2: Lead isotope abundances (mole-%) of a seized uranium ore sample (Find-25) and of natural lead. The lower line shows the results of a blending calculation, assuming a mixture of 56% natural lead and 44% lead from uranium ore from Joachimsthal mine.

3.5. Anionic impurities

Aqueous processing of nuclear material is encountered at a number of stages in the nuclear fuel cycle. In these processes mineral acids are frequently used. They leave anionic impurities (e.g. Cl^-, F^-, SO_4^{2-}, NO_3^-) in the material behind, together with those anions that were initially present in the starting material. We have studied such anionic impurities in yellow cake samples from different origins. Depending on the type of ore from which the uranium was extracted and the type of process applied as well as the associated chemical reagents used,

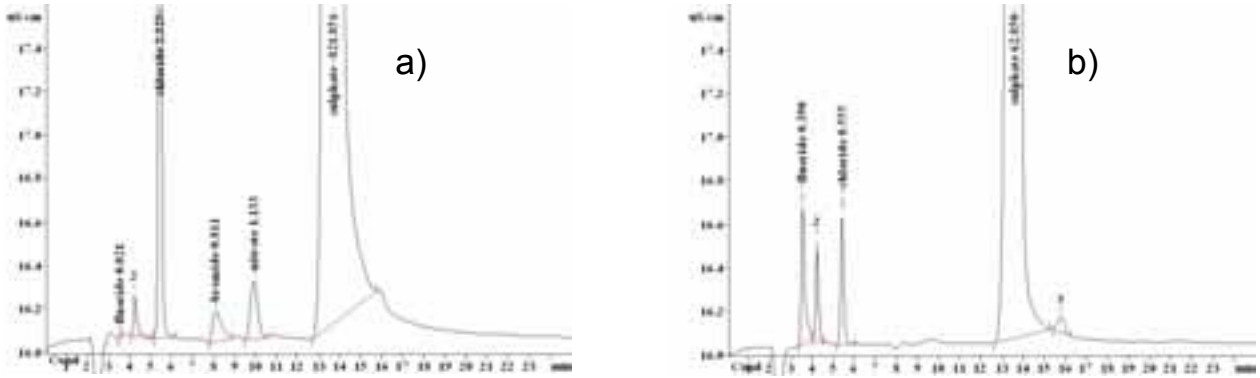


Figure 6: Anionic impurities in yellow cake samples from a German mine (a) and from a Gabonese mine (b) by ion chromatography.

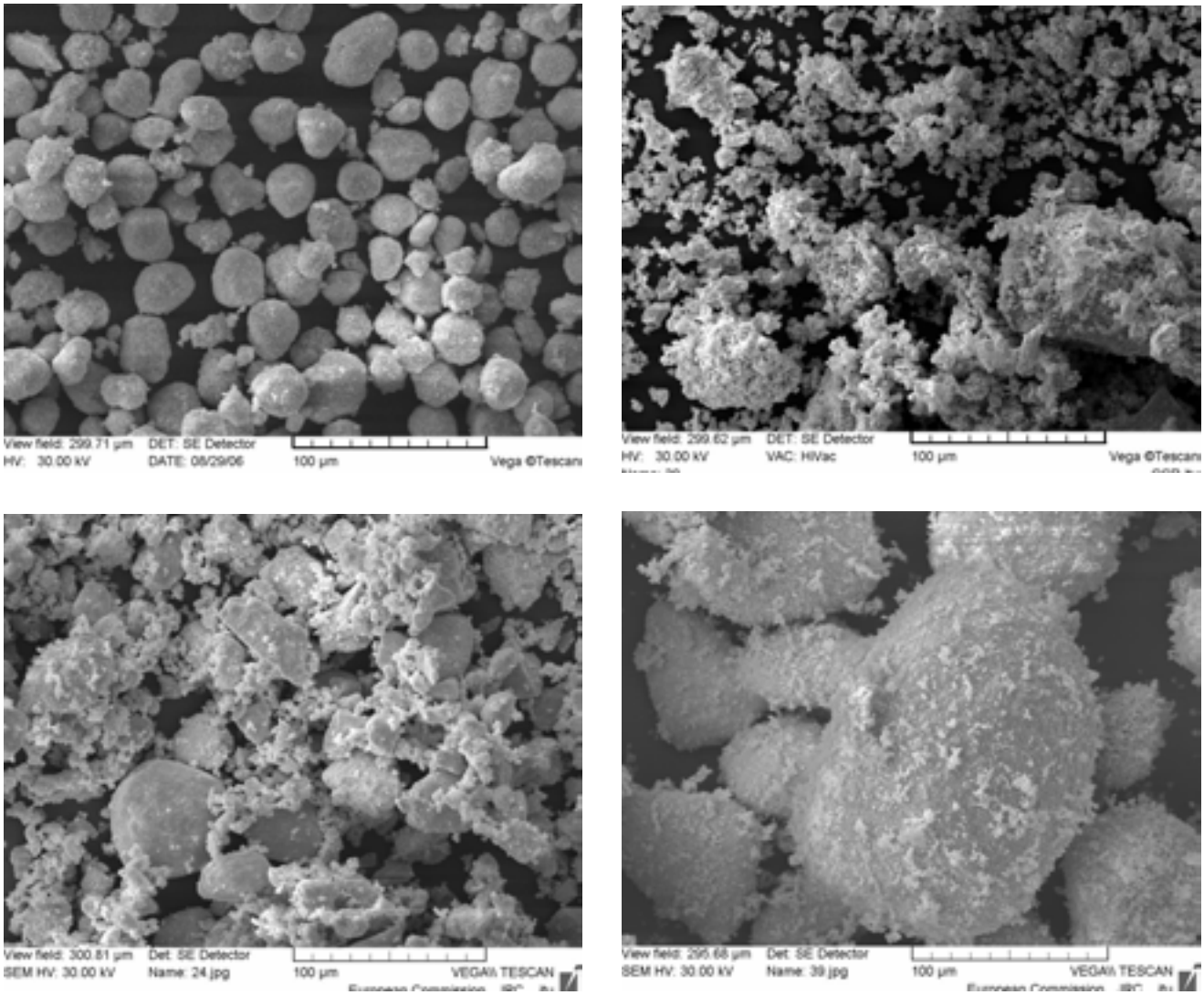


Figure 7: Comparison of microstructure in four UF_4 samples.

the isotopic patterns generated in the yellow cake are significantly different. These patterns provide additional information for distinguishing materials from different origins or – if appropriate reference data is available – for relating a given material to a specific facility. For data evaluation, the pattern of anionic species is more informative than the actual concentration values. Figure 6 shows examples of chromatograms obtained from yellow cake samples from Germany and Gabon [10].

3.5. Microstructure

Very little use has been made of microstructural information of nuclear materials in safeguards. This can be understood by the nature of the information, i.e. such information is essentially of qualitative character. Still the particle and grain size distributions and the surface structure of the particles are material characteristics that reflect the production process of the material. These data allow the direct comparison of samples enabling conclusions on coherence between samples. Fig. 7 shows a comparison of four UF_4 samples. The particles are shaped and sized very differently, thus they can be clearly distinguished from each others, indicating different origins of the four samples in question.

4. Conclusions

The challenges associated with strengthened safeguards call for more investigative analytical methods. The verification of treaty compliance according to comprehensive safeguards agreements and the additional protocol are associated with a tremendous need for information. Part of the information required for the evaluation of the completeness of a state's declaration is inherent to the nuclear material. Advanced and investigative measurement methods, such as applied in nuclear forensics, need to be introduced in nuclear safeguards. Consequently, we will see a convergence of nuclear forensic and of classical safeguards analysis.

Acknowledgement

The support of the Australian Safeguards and Non-Proliferation Office (ASNO) and the International Atomic Energy Agency (IAEA) for providing samples of natural uranium is highly appreciated.

References

[1] S. Richter, A. Alonso, W. de Bolle, R. Wellum, P.D.P. Taylor, Int. J. Mass Spectrom. 193 (1999) 9-14

[2] M. Wallenius, P. Peerani, L. Koch, J. Radioanal. Nucl. Chem. 246 (2000) 317-321

[3] M. Wallenius, K. Mayer, Fresenius' J. Anal. Chem. 366 (2000) 234-238

[4] K. Mayer, A. Morgenstern, M. Wallenius, G. Tamborini, C. Apostolidis, 42nd Annual Meeting of the INMM, 15-19 July 2001, Indian Wells (CA), USA

[5] M. Wallenius, G. Tamborini, L. Koch, Radiochim. Acta 89 (2001) 55-58

[6] M. Wallenius, K. Mayer, G. Tamborini, A. Nicholl, International Conference on Advances in Destructive and Non-Destructive Analysis for Environmental Monitoring and Nuclear Forensics, 21-23 October 2002, Karlsruhe, Germany

[7] L. Pajo, K. Mayer, L. Koch, Finnish-Russian Radiochemistry Symposium, 7-8 November 2000, Helsinki, Finland

[8] L. Pajo, G. Tamborini, G. Rasmussen, K. Mayer, L. Koch, Spectrochim. Acta Part B 56 (2001) 541-549

[9] J. Svedkauskaite-LeGore, K. Mayer, S. Millet, A. Nicholl, G. Rasmussen, D. Baltrunas, Radiochim. Acta 95 (2007) 601-605

[10] V. Badaut, M. Wallenius, K. Mayer, Addressing Verification Challenges – International Safeguards Symposium, 16-20 October 2006, Vienna, Austria

Working Groups activities

ESARDA Working Groups activities in 2007
Summary report

Abstract

Working groups are the 'backbone' of ESARDA. As last year, the Editorial Committee was pleased to receive and compile in a single report a summary of the ESARDA working group activities and achievements for the year 2007 presented hereafter.

Please note the activities of the NMACAF group are not included here as these were already published in Issue No 37 of the Bulletin; while the 2007 report by the Editorial Committee working group is the Editorial opening the present Issue.

1. *Activities of the ESARDA Destructive Analysis Working Group*
2. *Activities of the Non Destructive Analysis Working Group*
3. *The ESARDA Working Group on Containment and Surveillance*
4. *Activities of the ESARDA Working Group on Integrated Safeguards*
5. *The ESARDA Working Group on Verification Technologies and Methodologies*
6. *ESARDA Working Group Training and Knowledge Management*

1. Activities of the ESARDA Destructive Analysis Working Group

Chairman's Report by Klaus Mayer

The ESARDA Working Group on Standards and Techniques for Destructive Analysis (DA) addresses the challenges related to the measurement of samples of nuclear material. The methods applied in DA normally involve a change in the nature of sample induced by the measurement process (i.e. a change of chemical or physical appearance). The spectrum of activities of the working group reflects a wide variety of measurement techniques, of sample types and also of applications. At present the group consists of 48 members and special members from some 20 organizations. Recently, the working group experienced a shift in emphasis of its activities. In addition to the traditional methods for nuclear material measurements, aiming at determining the amounts of nuclear material for accountancy and verification purposes, the group addresses increasingly measurement challenges related to environmental sampling and to verification of completeness of declaration, also keeping abreast of the developments in the nuclear forensic area.

The review of inter-laboratory programmes (EQRAIN, SME, REIMEP and NUSIMEP) is a regular activity of the working group. During its 2007 meeting, the group noted with satisfaction that most laboratories achieve good performance in the element assay of nuclear material when participating in inter-laboratory comparisons. These inter-laboratory comparisons cover traditional materials and techniques, like titration or thermal ionisation mass spectrometry applied to uranyl nitrate solution, but extend also to isotope ratio measurements in uranium particles (produced by controlled hydrolysis of UF_6) using secondary ion mass spectrometry. The latter is organized by IRMM (NUSIMEP-6) and samples are being distributed in the first semester 2008. CETAMA offers inter-laboratory comparisons on uranium and plutonium (EQRAIN U and EQRAIN Pu) but also on multi elemental trace analysis (EQRAIN Traces). The group also initiated an evaluation of the need for revision of the International Target Values for Measurement Uncertainties (ITV). To this end, the performance data observed in the inter-laboratory exercises shall be examined and a working paper will be discussed at the 2008 meeting in Luxemburg.

With regard to training and education in the specific area of destructive analysis, the group finalized a "Technical Sheet" on mass spectrometry, describing a variety of mass spectrometric techniques and applications. Three more technical sheets on Titration, on reference materials and on Quality Control are under preparation. Furthermore, training material on destructive analysis and on nuclear forensics was drafted in order to support the nuclear safeguards and non-proliferation training course which

the ESARDA WG on Training and Knowledge Management offers at the JRC Ispra site. The group contributed also to this year's training course with two lectures on nuclear forensics and on destructive analysis.

Measurement problems arising from new challenges in safeguards and in related areas are addressed. Therefore, the WGDA promotes the use of new parameters for obtaining supplementary information on nuclear material for forensic and for safeguards purposes. In April 2008 IRMM (Institute for Reference Materials and Measurements in Geel, Belgium) hosted a workshop on "Minor isotopes in uranium" addressing the challenges and benefits associated with the measurement of minor abundant isotopes in uranium (i.e. ^{234}U and ^{236}U). Increasing use is also made of chemical impurities in nuclear materials. This topic will be addressed in a dedicated workshop.

2. Activities of the NDA Working Group

Chairman's Report by Paolo Peerani

An overview of the recent past and present activities of the ESARDA NDA Working Group has been presented at the 2006 IAEA safeguard Conference in Vienna and the paper is posted on the ESARDA website and available at the following link:

http://esarda2.jrc.it/internal_activities/WG-NDA/index.html

We will give here a summary of the main activities carried out in 2007 and some perspectives on the planned activities for 2008.

During 2007 the WG launched the 3rd and 4th phase of the *Multiplicity Benchmark*. The first two phases were completed in 2006 and the final report published in the special issue of the ESARDA Bulletin, number 34 (September 2006). With respect to the previous phases, the new benchmark focuses on experimental data instead of synthetic pulse train generated by Monte Carlo simulations. The measurement campaign has been done on the last week of January in the PERLA lab in a joint team including JRC, IRSN and LANL, acquiring in particular the 6 reference measurements that have been used for the exercise, using an AWCC and repeating the acquisitions with a conventional multiplicity shift register and a list mode system. The 3rd phase is full simulation of the exercise, which consists in reproducing by Monte Carlo calculation and data treatment the experimental results, and the 4th phase is only data treatment of the list mode data acquired by IRSN. The measurement campaign also allowed an inter-comparison between different digital acquisition systems developed or used by JRC, IRSN, LANL and later in a following test by IKI. The results were published in a joint paper presented at the 29th ESARDA Symposium (Aix-en-Provence, May 22nd-24th 2007). The pulse trains and the technical specifications of the benchmark were distributed to the participants in February and delivery of results was expected in autumn. Indeed not all the participants provided their answers before the end of the year, so the collection of results extended to the beginning of 2008. The preliminary report of the benchmark will be presented at the 30th ESARDA Annual meeting (Luxembourg, May 27th-29th 2008).

Another project that advanced significantly in 2007 was the development of a document on "*Good Practice Guide for the use of modelling codes in Non Destructive Assay*". The collection of contributes from different authors has been mostly completed and already assembled in a structured document. Most of the chapters have been already reviewed and accepted. Few minor sections are missing and planned to be drafted at the beginning of 2008. The goal is to have the first official complete version ready at the annual meeting in order to have it externally reviewed and finally endorsed by IAEA, who was the promoter and main requester of this document.

The NDA section of the *Syllabus of the ESARDA course on safeguards and non-proliferation* has been completed, reviewed and endorsed by the NDA-WG and finally transmitted to the TKM-WG.

A new initiative with potential to open a lot of future activities in the field of gamma spectrometry was born during a topical meeting organised by the NDA-WG together with the US support programme to the Agency in Aix. During this meeting the organisation of an international *"Working Group on Gamma Spectrometry Techniques for U/Pu Isotopic"* was proposed following a request of support from the Agency concerning the sustainability and standardisation of gamma evaluation codes. Whereas the new working group will organise himself independently, it will be intimately interconnected with the ESARDA NDA-WG, from which it will profit of the networking capabilities and of the e-infrastructure (i.e. the website). The Terms of Reference of the new working group have been agreed and are published on this number of the ESARDA Bulletin. The first activity of the new working group will be an INMM/ESARDA workshop to be held in Oak Ridge in November 2008, see link: http://www.inmm.org/events/gamma/index.cfm.

3. The ESARDA Working Group on Containment and Surveillance

Chairman's Report by Bernd Richter

In 2007, the working group had 19 members from R&D establishments, safeguards equipment manufacturers, safeguards inspectorates, plant operators, regulatory agencies, and ministries. They represented the ESARDA organisations: European Commission, Finnish and Swedish nuclear regulatory authorities, French Institute for Radiation Protection, Safety and Security, and AREVA, German Nuclear Fuel Cycle Association and Jülich Research Centre, United Kingdom Safeguards Organisation and Sellafield Safeguards Department. Members from outside ESARDA represented the International Atomic Energy Agency, Argentine-Brazilian safeguards authority ABACC, Canadian Nuclear Safety Commission CNSC, and US Sandia National Laboratories.

The working group addressed the following issues: performance & assurance of containment and surveillance (C/S) instrumentation, wireless in-plant data transmission, IAEA's Next Generation Surveillance System, safeguards for geological repositories, laser scanning for design information verification at underground repositories, JRC Ispra safeguards review station concept, containment verification methods, guidelines for developing sealing and containment verification systems, remote monitoring centre and activities in progress at the IAEA.

Recurrent activities are: information exchange and discussions on R & D within the working group, support of the ESARDA working group on Training and Knowledge Management, and support of the ESARDA Editorial Committee.

Achievements of the working group are publications in the ESARDA Bulletin and in conference proceedings (2006 IAEA Symposium, 2007 ESARDA Symposium), and contributions to the ESARDA web site, such as the compendium on C/S instrumentation and, most recently, technical sheets on: radiation monitoring techniques for monitoring the movement of discharged fuel, JRC ultrasonic seals.

The working group's ongoing major project is to develop a methodology for determining the performance and assurance of the C/S instrumentation. It was agreed to do a first trial application on a dry storage facility for spent fuel paying attention to issues like instrument reliability and suitability. It is anticipated that the method will also help to draw up guidelines for developing sealing and containment verification systems. By March 2008, the WG will receive example design information of the dry storage facility, a requirements profile, specifications of seal candidates and DCM14 surveillance system, and a checklist for determining the C/S performance. Before May 28, 2008, WG member are invited to perform the trial application to determine the performance of C/S instrumentation, so that the results can be discussed at the next WG meeting.

Another issue is wireless in-plant data transmission. The first part of a paper was published in ESARDA Bulletin 36, providing specific arguments for considering the use of wireless communications as a complement to fixed cable installations. From a safeguards authority's point of view, wireless in-plant data transmission is attractive for the ease of installation and the ability to respond to the changing requirements as the inspection approach evolves. However, for wireless technologies to be considered as a viable complement to cables, a number of concerns have to be accounted for. There are specific requirements arising from the plant operators' boundary conditions which have to be addressed before wireless techniques are deployable in nuclear facilities. The paper presents an overview of state-of-the-art wireless technologies and makes a projection on capabilities that are likely to be reached in the near future. The second part is being prepared for publication in ESARDA Bulletin 38. This paper will be dedicated to RF technologies in a safeguards concept, to information security considerations, and to the integration of wireless technologies into existing and new facilities.

Spent fuel management: Sweden and Finland cooperate in identifying the C/S functions required to maintain the continuity-of-knowledge after final verification of the nuclear material in an encapsulation plant. The working group endorsed the following: Any undeclared retrieval of spent fuel from the repository has to be detected. C/S may include surveillance, sealing, dual C/S, finger printing, tracking after encapsulation, portal monitoring. Encapsulation may take place under surveillance. Surface scanning of the cask may be foreseen. Performance testing may be needed for the portal monitoring. Remote monitoring may be foreseen to reduce inspection effort. Reliability testing may be required (for repository conditions).

Design information verification (DIV) in an underground nuclear repository: the Finnish safeguards authority in making certain assumptions regarding facility misuse and definition of repository, has developed a concept for exploration, construction and implementation of a repository for final disposal of nuclear waste at Olkiluoto (ONKALO underground laboratory). Laser scanning for DIV during repository construction could play a major role in a safe-

guards approach. JRC Ispra has developed a laser scanning technique for DIV and presented promising results of a first scanning campaign at the ONKALO site. If agreeable in a safeguards approach, the plant operator will provide design information in the form of paper documentation to the IAEA, and the IAEA will verify the declaration by laser scanning.

Safeguards review station concept: JRC Ispra presented a concept for enhancing the GARS-based review of surveillance images (GARS = General Advanced Review Software). To this end, change detection, Markov Models, image retrieval, and decision tree analysis were introduced and combined. Keywords in the subsequent discussion were confirmation of declaration, detection of undeclared operations, shape and size of relevant objects, search by content, time consistency checking, possibility to switch to shorter picture taking interval, cross-triggering of image and radiation data. The WG found the development very promising.

Identification and containment verification: JRC Ispra demonstrated laser techniques for identification and containment verification of Pu cans and UF6 cylinders.

Input was provided to the ESARDA working group on Training and Knowledge Management for the Ispra Course on Safeguards (a chapter on C/S within the textbook on safeguards for students).

The working group will issue further technical sheets on: mail box systems, design information verification, optical surveillance techniques, and cap-and-wire seals.

The working will further discuss the following topics: guidelines on sealing and identification systems, containment verification methods and techniques, geological repositories, remote system control, data review, trial application on performance of C/S devices, interface between safeguards and security.

4. Activities of the ESARDA Working Group on Integrated Safeguards

Chairman's Report by Arnold Rezniczek

The ESARDA Working Group on Integrated Safeguards was created in 2000 with the objective to provide the Safeguards Community with expert advice on methodologies and approaches to integrate INFCIRC/193 and INFCIRC/540 measures and to present a forum for the exchange of information, views and experiences in that regard. Its members represent inspectorates, national authorities, operators and research centres active in the field of safeguards.

In the first years, the working group concentrated on issues of the implementation of the Additional Protocol (AP), which is a prerequisite for Integrated Safeguards (IS). Later, the emphasis passed over to the discussion of different IS concepts and elements. In2007, the working group had 2 meetings, one in March 2007 in Almelo and one in October 2007 in Ispra. In these meetings we discussed issues like the mailbox concept for enrichment plants, the SNRI concept for fuel fabrication plants, experiences with environmental sampling, the implementation of IETS principles in power reactors (IETS means Implementing Euratom Treaty Safeguards), differences between INFCIRC/193 and INFCIRC/153-type agreements and offering voluntary access to locations to the IAEA according to article 8 of the AP. The central task to provide a forum for exchange of information always remains an essential function of the working group filling a considerable part of our meeting's time.

Although all of the 27 EU Member States have an AP to their safeguards agreement in force, the status of implementation of IS by the IAEA is different in the EU. The arrangements for and the implementation of IS seem to be an elaborate process taking more time than thought in the beginning. As of December 2007, only Member States that joined the Community in 2004 or later had IS implemented because the IAEA had started this process already before their accession to the Community's Safeguards Agreement (INFCIRC/193). In the beginning of 2008, some of the 'old' EU Member States with minor nuclear activities got IS; for the States with major nuclear activities this process is still under way.

This diversity induces active discussions and exchange of information. Both IAEA and the Community's safeguards services are regularly represented in our meetings and provide the group with information on the actual status of discussions and developments in their organisations. The group in turn communicates opinions, conceptions and peculiarities of operators and national authorities concerned and thus delivers a valuable feedback to the inspectorates to assess approaches under consideration. We are expecting further progress being made in the current year towards a timely implementation of IS in EU Member States with more complex nuclear activities.

5. The ESARDA Working Group on Verification Technologies and Methodologies

Chairman's Report by Gotthard Stein

The mission of the ESARDA Working Group on VTM is to provide the safeguards community with expert

advice on modern verification technologies and methodologies and to act as a forum for the exchange of relevant information in this area. Individual working group members volunteer to prepare discussion and working papers, subgroups are established, conferences and meetings with special topics are performed, and an important goal is to publish the results of major activities.

A wide range of verification issues in nuclear and non-nuclear treaties, research and development of innovative verification technologies, synergies between verification regimes or the implementation of advanced verification technologies and approaches constitutes the workload of the VTM group and determines the agenda of the meetings. The following list serves as an example for the VTM activities in the last years:

- Chemical Weapons Convention Verification Regime
- Verification under the Kyoto Protocol
- Nuclear Forensics
- Non-proliferation and EU Security Network
- Dual Use and Export Control Issues
- Role of Information Analysis for International Verification
- Satellite Imagery and International Security
- Wide Area Monitoring
- Environmental Monitoring
- Potential of Laser Measurements for Safeguards

During the year 2007 the VTM group had presentations and discussions in the following fields:

- Novel Technologies for IAEA Safeguards
- Laser Enrichment Measurement
- Satellite Imagery in Cooperation with GMOSS and LIDAS
- Satellite and Seismic Monitoring
- Proliferation Resistance
- Discussion of ISTC Issues
- Integration of Information for Non-proliferation Studies

Due to these broad issues and various activities group members have expertise and background from laboratories, universities, international organisations, national authorities and ministries, industry, research centres and institutions. The complexity of the VTM issues made it necessary to create subgroups:

- Subgroup on Environmental Monitoring (Chair: Martin Kalinowski)
- Subgroup on Satellite Imagery (Chair: Bhupendra Jasani)
- Subgroup on Novel Technologies and Approaches for IAEA Safeguards (Chair to be decided)

In the frame of the VTM group two publications have been issued:

- Verifying Treaty Compliance: Limiting Weapons of Mass Destruction and Monitoring Kyoto Protocol Provisions, by (eds.) Rudolf Avenhaus, Nicholas Kyriakopoulos, Michel Richard and Gotthard Stein, Springer, July 2006.
- International Safeguards and Satellite Imagery: Key Features of the Nuclear Fuel Cycle and Computer-based Analysis, by (eds.) Bhupendra Jasani, Irmgard Niemeyer, Sven Nussbaum, Bernd Richter and Gotthard Stein, Springer, August 2008.

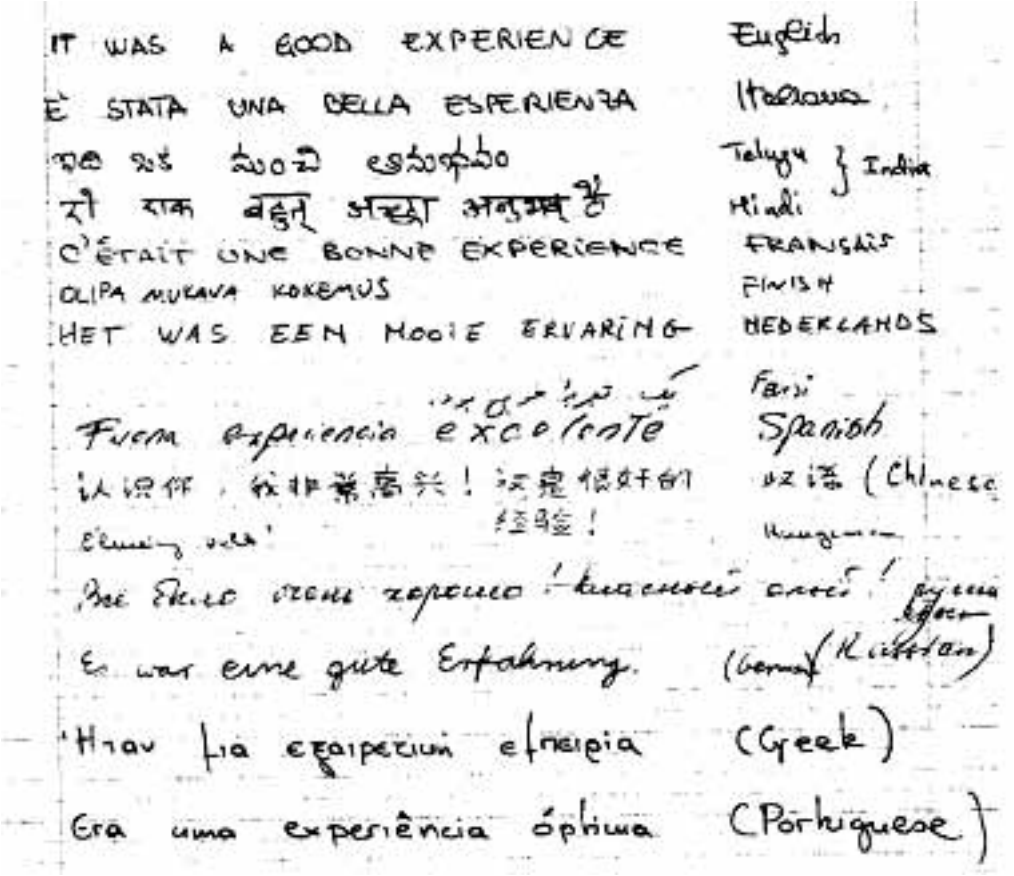
A further book publication is planned for the complex field of proliferation resistance.

6. ESARDA Working Group Training and Knowledge Management

Chairwoman's Report by
Greet Janssens-Maenhout

The renaissance of nuclear technology urges the parallel development of the necessary human resources potential. Technological, economical, legal and political considerations all flow in the debate on future reactor systems and fuel cycle facilities. The decreased number of students going for engineering these last years, more particularly in the nuclear area, led to the current lack of managers and experts necessary for operating a complex technical system like a NPP. Engineers who master also juridical aspects or vice versa juridical experts who master also engineering are scarce. University faculties teach their field of competence, either technical or juridical with courses traditionally in their university faculty. To meet these challenges and streamline the educational resources, new synergies with interuniversity collaboration have to be imagined, as is demonstrated by the European Nuclear higher Education Network, setup in 2002. A next step is interfaculty collaboration, which was explored between the faculties of Physics and Political Sciences at the University of Florence in 2007. In addition to this education, the World Nuclear University established multidisciplinary training for young professionals in summer schools.

The ESARDA WG TKM supported these initiatives by providing the annual one-week course "Nuclear Safeguards and Non Proliferation", which is hosted by the Joint Research Centre in Ispra. The course was taught in the last 4 years to almost 200 students and is meanwhile recognized by the academic world. The aim is to integrate this as compulsory course in the standard curriculum for a Master of Science degree in Nuclear Engineering. The interest



Spontaneous feedback of the students on the third ESARDA course, Ispra, 5-9 March 2007

for the course is steadily increasing (with a peak of several thousand visitors of the course website per month) and the demand for participation is approaching a numerus clausus.

The feedback of the students on the course expressed a very high satisfaction, which is also illustrated by the spontaneous students' paper of thanks. In response to the feedback analysis, the course equilibrium between technical and juridical matters is improved by taking on board juridical experts in the pool of lecturers. ESARDA is grateful for the support of the different organizations (IAEA, EURATOM, University of Uppsala, University of Hamburg, University of Liège, AREVA, BNG, SKI, IRSN, JRC), sending and sponsoring the high quality lecturers.

In the EU no many similar multidisciplinary education initiatives on safeguards, non-proliferation and security exist and a strengthening by cooperation is fostered. The JRC is looking for supporting the International School of Nuclear Law by the OECD organized at the University of Montpellier, the new master on nuclear material management and control of the fuel cycle in the non-proliferation regime at the University of Florence, and the education and research by the Friedrich von Weiszäcker Centre for Atoms and Peace at the University of Hamburg. This is completely in line with the IAEA Director General, who was promoting during the Conference in Florence of October 2008 a twofold education in nuclear technology and in nuclear law.

Working Group on Gamma Spectrometry Techniques for U/Pu Isotopics

Terms of Reference

1. Background

In November 2005 an International Workshop on "Gamma Evaluation Codes for Plutonium and Uranium Isotope Abundance Measurements by High-Resolution Gamma Spectrometry: Current Status and Future Challenges" was held in Karlsruhe, Germany. This workshop was attended by many of the primary experts in gamma spectrometry techniques including many of the initial code developers. One of the main issues discussed during that meeting was addressing the concerns of the inspectorate authorities (IAEA, Euratom and ABACC) about the standardisation and sustainability of gamma evaluation codes. Clear guidelines were identified for future challenges (including technical developments). As a follow-up the Agency issued a roadmap for future developments of gamma codes that culminated in the submission to several MSSP's (US, EC and F) of a request of support in this field.

In parallel a different initiative was carried on under the framework of the US-DoE/ABACC bilateral safeguards cooperation agreement with the establishment of an Isotopic Measurements Working Group (focusing only on uranium enrichment measurements) gathering experts from US, Brazilian and Argentinean laboratories with the goal to "*test and evaluate existing uranium isotopic and enrichment software through an interactive and cooperative process among users and developers and make recommendations for standardized software that meets regional and international quality assurance (QA) requirements for the safeguards community*". The work under Action Sheet 14 (AS 14) will be successfully completed concerning the initial scopes.

Therefore it is considered by those involved in the development, control and use of these gamma evaluation codes that we should endeavour to continue the positive experience of AS14, enlarging the participation to the international community, broadening the scope in order to include plutonium isotopic measurements in order to establish a working group (WG) that would respond the needs expressed by the Agency.

Since AS14 already includes many of the American subject matter experts, it was quite straightforward to look at ESARDA, and in particular to the NDA working group as the catalyser for the institution of this dedicated WG. The proposal to launch the institution of an international **Working Group on Gamma Spectrometry Techniques for U/Pu Isotopics** was decided in a meeting organised in Aix-en-Provence on the 21st of May 2007.

2. Scope

The scope of the WG is to provide a forum for exchange of information, technical developments, validation and testing of gamma spectroscopy techniques used to determine the isotopic composition of uranium and plutonium samples in nuclear safeguards applications. Specifically, but not exclusively, it will address issues related to gamma evaluation codes, such as their applicability, capability and limitations, standardization, sustainability and version control.

3. Composition

In principle any stakeholder involved in gamma spectrometry applications, either as technique developer or as an end-user for safeguards applications, can join the WG. Typical membership will include: national and international research laboratories, nuclear plant operators, inspection authorities, code developers and instrument manufacturers.

It is recognized that instrument manufacturers have extensive experience with respect to integration of such codes into comprehensive measurement systems, In addition, it is recognized that commercial entities currently provide support and sustainability for codes that they currently market. Therefore, participation of detector manufacturers is encouraged but strictly monitored to address any conflict of interest because the scope of the WG is purely scientific and not for profit,. Representatives of the commercial companies will be permitted to:

- attend and actively participate in the meetings,
- provide information and experience feedback on their products,

- have access to the results and documents produced by WG,
- participate in the development of international standards with the consensus of other subject matter experts,
- participate in drafting recommendations for technical specifications and requirements with the consensus of non-commercial subject matter experts,

but will be excluded from participating in comparative evaluation of the techniques to eliminate the potential for a conflict of interest.

4. Management

The WG will have no financial budget. Each participating organization will contribute to the WG bearing its own costs for manpower and missions.

The deliverables (reports and other documents) of the WG will be available free of charge to all the participants.

The WG will be formally hosted within the ESARDA-NDA working group in order to profit from the existing network and e-infrastructure.

The WG will be coordinated by the NDA-WG chairman and a co-chairman elected by the participants, whose main role will be the organization and management of the meetings, distribution of the official documents produced by the WG, establishing links with international institutions and administrative bodies for project funding such as the U.S. DoE, the IAEA, various MSSP's and other non-government organizations.

5. Activities

The activities to be performed will be planned and agreed by the WG in the regular meetings. These activities can cover, but are not limited to, any of following topics:

- set-up and maintenance of a web forum for discussions and information repository of gamma spectroscopy related issues,
- organisation of experimental round robins and evaluation benchmarks,
- selection of a commonly agreed platform containing a set of gamma reference spectra to be used for test and evaluation of gamma codes,
- maintenance, upgrade and availability of the testing platform,
- analysis and validation of hardware and software techniques,
- issue guidelines for QA, best practice guides, recommendations on the (application-dependant) use of hardware setups and/or evaluation codes,
- propose/sponsor production of reference material for calibration purposes,
- recommend standardisation of nuclear data,
- perform a version control of the main codes, recommending the last fully-validated version and advising WG members of the issue of new releases by checking the validation process,
- collect users' needs and convene them to the developers,
- assure that codes are maintained and updated according the rules expressed by the regulators,
- monitor advances in gamma spectrometry, such as new detectors, and
- notify members of planned workshops and encourage attendace.

The Joint Research Centre, which is providing the secretariat of ESARDA and hosting the website of the Association, will provide the e-infrastructure for all the WG activities within the ESARDA website.

The WG should meet indicatively once a year (alternating Europe and America) possibly in conjunction with major safeguards events (INMM or ESARDA conferences) in order to minimise the travelling inconveniences of the WG members. Advance notification of such meetings will be posted on the website and distributed to other nuclear non-proliferation and nuclear safeguards organizations to ensure the information is disseminated to all personnel who want to attend.

Wireless Communications for Monitoring Nuclear Material Processes PART II: Wireless In-plant Data Transmission

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Abstract

The wireless transmission of data from sensors, monitoring both static and dynamic safeguards processes, is highly appealing for the simple fact that there are no wires. In a nuclear safeguards regime, this has the implied benefits of low-cost installations, versatile configurations, and the elimination of conduits to inspect. However, with the implied solutions of wireless, we are presented with a new set of problems for system implementation and operation management, in particular (i) Radio Frequency (RF) interference and (ii) security in information transmission. These problems are addressable. This paper looks at the clear benefits of wireless technologies and the cautions regarding the possible pitfalls of poorly applied technology, discusses the integration of radio frequency in existing and new facilities, provides high-level considerations for information security, and reviews prospects for the future.

Keywords: Wireless Communications, Interference, Security in Communications, Containment and Surveillance

1. Introduction

From a safeguards authority's point of view, wireless in-plant data transmission is attractive for the ease of installation and the ability to respond to the changing requirements as the inspection approach evolves. However, for wireless technologies to be considered as a viable complement to cables, a number of concerns have to be accounted for.

Plant operators may need to impose limitations on the transmission standards proposed to be used in facilities to avoid interference with their safety equipment, including physical security systems. The safeguards authority needs to be guaranteed that C/S equipment and data transmission processes will not be affected by the other existing RF plant equipment. As such, prior to any RF system implementation, the environment where it will be utilized must be characterized (e.g., by an RF survey), controlled after installation, and periodically re-assessed. Although preventing interference implies an additional cost to installation and maintenance, it is fair to say that interference is a quite well understood problem: procedures and best practices exist to deal with it as it is a shared issue to several industrial sectors using RF devices.

For confidentiality reasons, the safeguards data being transmitted must not be available for analysis by a third party. In addition, the safeguards authority requires data to be authenticated as close to the point of acquisition as possible.

As a general trend, it is noted that the data security provided by RF communication systems is continuously improving due to the requirements of applications in various industrial sectors. In certain cases, stringent data security requirements imposed by safeguards may be addressed by extreme protocols embedded either at system level or at the lowest level of data acquisition.

In this context, the ESARDA Working Group on Containment and Surveillance addressed the issue of wireless in-plant data transmission for safeguards purposes and supported the publication of a paper in two parts.

Part I [1] provided specific arguments for considering the use of wireless communications as a complement to fixed cable installations. In addition, it points to specific requirements arising from the plant operators' boundary conditions to be addressed for wireless technologies to be deployable in facilities. Furthermore, it introduces basic concepts underlying wireless communication, including methods for transmission, issues on power consumption, frequency, range, and considerations on interference and noise resilience. Finally, part I presented an overview of state-of-the-art wireless technologies and makes a projection on capabilities that are likely to be reached in the near future.

Hereafter, Part II of the paper covers the principal issues regarding *information security* and why and how processes and protocols can be implemented to significantly mitigate security and information confidence concerns. Another area of discussion is

considerations for the *integration of wireless technologies into existing nuclear facilities* taking into account the *RF interference* problem, and ways to *integrate support for these technologies into the design phase of new facilities*.

2. Information security considerations

Wireless Local Areas Networks (LANs) face the same security challenges as their wired counterparts and more. Because the medium for wireless is air, wireless LANs have the added issue of securing data that travels the airwaves.

Traditional wired networks use cables to transfer information, which are protected by the buildings that enclose them. To access a wired network, a hacker must bypass the physical security of the building or breach the firewall.

On the other hand, wireless networks use the air, which is an uncontrolled medium. Wireless LAN signals can travel through the walls, ceiling and windows up to thousands of feet. Additionally, airwaves are a shared medium that allows anyone in the proximity to 'sniff' the traffic.

The details of the design of equipment to address the security needs for safeguards applications is beyond the scope of this paper and this section only provides a summary. For a detailed discussion, the reader can refer to [2].

Data must be assured to be *authentic* if it is to be used for safeguards purposes. It must be known that the data originated from the intended source was not changed in transit, and it is not a repeat or delayed copy of previous data.

For example, if the purpose of the system is to track items, the devices attached to the monitored items must be secure from duplication or impersonation attacks and must be securely attached to the monitored item so that any attempt to transfer the monitoring device to another item will be detected. RF identification (RFID) systems that were not designed specifically for safeguards applications do not provide these assurances. There is a trend, however, on more secure protocols [3] to mitigate the set of possible attacks [4] and enable new applications such as the electronic passport. Further considerations are presented in Table 1.

Data encryption protects it from disclosure to anyone who is not authorized to receive the information. In some cases, the facility operator or the host state will require the data to be encrypted if there is a chance the data might be intercepted by parties outside the facility. Therefore, data that is transmitted over wireless connections will require encryption before transmission, in case an adversary is able to intercept the wireless signals. Similar concerns exist for regular State-of-Health messages. These messages, often with the same information content, should be unrecognizable as such, for example, by always appearing different in structure to a potential eavesdropper.

Some data must also be protected from disclosure to the operator or to the state's authority. For example, data that will be used to verify the operator's declaration of activities should not be disclosed to the operator before he makes his declaration. Otherwise, he could modify his disclosure to match the verification data.

Data must also be *cryptographically authenticated* before an adversary exposes it to possible alteration or substitution. Although encrypting the data during transmission will give this protection, the data will again be vulnerable once it is decrypted at the receiving station. A better approach is to add either a Message Authentication Code (MAC) or a Digital Signature (DS) to the data before it is transmitted from the monitoring device.

These information security considerations are especially important when working with wireless communications. Since an adversary can introduce falsified data without the need to physically access the network cables, attacks can be mounted more easily, so users are well advised to use strong authentication. Similarly, because an eavesdropper can listen in to the message traffic from anywhere the wireless signal can be detected, encrypting sensitive data will be required in many systems that would not normally require encryption if wired communications were used.

In what follows we present a review of data protection options in the safeguards context.

2.1 Utilizing symmetric key data protection techniques

In *symmetric cryptography* [5], the same key is used at both ends of the process. That is, the key used to apply the authentication information to the data is again used to verify it, and the key used to encrypt the data is again used to decrypt it. These algorithms are compact and fast, making them well-suited for use in small sensor platforms.

Data is protected from disclosure by a strong encryption algorithm, such as the *Advanced Encryption Standard* (AES, [6]). Some parts of the data packets, such as the routing information, might be left unencrypted for operational reasons with only

the sensitive information protected from disclosure. However, care must be taken to ensure that adversaries cannot draw conclusions that would give them insights into the sensitive information from these unencrypted portions of the transmissions. For example, safeguards authorities might be triggering a camera from a radiation detector. If the triggers are the only messages passed from the detector to the camera, adversaries could perform a set of experiments to determine the sensitivity of the detector, which could lead to their designing a shield to divert material without detection.

Data authentication is usually accomplished using a keyed hash function or encryption algorithm to add a MAC to the data at the sensor. The MAC is calculated using the data and a secret key, that must be held secure. When the authenticity of the data must be verified, the same calculation is performed using the same key, and the result is compared to the MAC that was applied at the sensor. This works well when only one party will use the authentication information, but it is very difficult to implement securely in a data-sharing scheme, since the secret key must be known to verify the authenticity of the data. Any party that can verify the data can also substitute bogus data using this key.

Symmetric key cryptography also requires the movement of secret keys between multiple locations and the use of these keys by a large number of users, greatly increasing the risk of disclosing the secret key to unauthorized users.

2.2 Utilizing public key infrastructure for joint use information

Public key cryptography [7] uses a pair of keys: one for signing or encrypting the data and another for verifying or decrypting the data. Knowledge of one half of the key pair does not allow the direct calculation of the other key. Although this calculation is theoretically possible, it is considered a 'hard problem'. Public key cryptography can be used for both encryption and authentication and greatly simplifies key management for most systems, especially when multiple parties use the data.

Unfortunately, the calculations for public key cryptography require much more computing power than is required for symmetric key algorithms of equal strength. Until recently, this meant that small systems, such as seals that run for years using relatively small batteries, could not use public key cryptography. However, advances in both microprocessor technology and cryptography make the use of public key cryptography possible in these systems, especially if Elliptic Curve Cryptography (ECC) [8] is used. The ECC algorithm has an appealing energy load distribution for its crypto processes. Of the total energy required, one-third is consumed in signing the information and two-thirds is consumed in the verification of the signature. For low-power RF sensor applications, the signing process is performed in the secure sensor housing where energy needs to be conserved, and the verification process is performed on a host computer where energy is abundant.

2.3 Message protocol considerations

Along with authentication and encryption usage to protect message content and integrity, there are many *protocol-based mechanisms* that can be used to thwart additional insidious attacks. Protocol is the area where many commercially useful wireless systems can present vulnerabilities when utilized for safeguards applications. Low data rate systems, such as those that may be used to collect sensor data for safeguards applications, have the luxury of adding protocol features to increase security. The addition of these protocol features may present high bandwidth systems, such as IEEE 802.11 (WiFi) [9], with too much overhead and too many constraints to be practical for these types of applications.

A subset of the protocol features that have been found to greatly increase the security of low data rate systems includes the following attributes:

- **Time variants:** Every message contains a piece of information that changes with time, such as time of day. This distinctive part of each message significantly reduces the possibility of repeated or counterfeit messages. Time variants address the possibility of an adversary capturing a message to play it back sometime in the future to elicit a particular response from the RF device and then utilizing this knowledge to circumvent the system's purpose.
- **Counters:** There are two counters that are generally included in every message—a message counter that creates a total count of every message that has been transmitted and an up-counter. The purpose of the message counter is to determine if any messages have been missed prior to the latest message. If the count difference (prior message to current message) is small (e.g., two), then the missing message is simply requested for re-transmission from the source device to complete the message set. This circumstance should be very infrequent; however, if this occurs frequently, it can be an indication that some characteristic in the RF environment has changed and merits investigation. If the dif-

ference between the counts is relatively large, it could indicate that an adversary is attempting to cause Denial of Service (DOS). The up-counter is a large counter (typically 4 bytes) with an ever-increasing value that may simply contain integer seconds as a value. The current value is compared with the previous value from the same verified source to ensure that the current value is larger. This feature is actually another time variant the adversary would need to know to successfully counterfeit a message.

- **Fixed message field format:** Assembling a message for transmission requires compiling fields of data in an array that is then streamed out across the airwaves. If the data fields are assembled in the same order for a given message type, it significantly reduces the possibility of authentication collision. This approach makes it highly improbable that the adversary could generate an intelligible message that would match the fixed message field format with an authentic signature. Additionally, the message must accomplish the adversary's intended purpose to represent a viable attack.
- **Minimal command set:** To mitigate the possibility of an adversary invoking a command that may alter the prescribed operation of a sensor module, over-the-air feature programming is not allowed. Feature programming is typically done prior to deployment over a special hardware connection. This connection is electrically disabled after programming. Internally, the software establishes constraints on buffer sizes and provides buffer size-monitoring processes that mitigate the possibility for buffer overrun attacks. Buffer overrun attacks attempt to externally exploit available commands and cause pre-determined changes in memory, which provide a beneficial outcome for the adversary.

3. Projections of future wireless communications capabilities

The future of low data rate RF communications will bring forth technologies that will be more integrated, more secure, and will require much less power.

As an example, RF transceivers demonstrate a continuing trend toward the development of more highly integrated RF transceivers utilizing Systems-on-a-Chip (SoC) [10]. Today, it is reasonable to expect to design an RF device that utilizes a transceiver integrated circuit with a microprocessor providing *direct* functions such as encryption, error detection/recovery, and collision avoidance. This is just one example of the advances seen in RF technology aimed at guaranteeing a consistent implementation of all these functions, an absolute requirement for future wireless communication that must conform to communication standards.

Table 1 lists some potential near-term projections for future low data rate, low-power RF communications: Ultra-wideband [11], Secure Sensor Platform [12], RuBee [13], ZigBee [14], and SoC [10].

Figure 1 shows a RuBee tadio tag.

Specific advances in RF transmission technologies will not be the only influence in the overall performance gains for wireless communications in the near future. These gains will be attributed to other associated technology areas as well. A recent example is the cell phone industry. Cell phones have evolved from large, cumbersome items that served only one purpose, to small, convenient items that have many uses (camera, internet, etc.). This evolution of the cell phone can be attributed to market forces.

Figure 1: A RuBee Radio Tag (4 bit CPU, 1kB sRam, and lithium battery with expected life of 5 years).

There is a significant difference between the evolution of cell phones and the technologies discussed here for safeguards applications. Currently, there are estimated to be 2.7 billion cell phone subscribers in the world, representing a huge market. Safeguards technology as discussed in this article is highly specialized and will not likely have a significant market force driving its development. In many cases, however, *safeguards applications will be able to leverage off some of the consumer and commercial market developments as well as developments driven by the large sector of industrial control and automation* [15]. For example, energy storage densities and the efficiencies of extracting the stored energy are driven greatly by the commercial market. These gains are being leveraged by designers for safeguards applications and will have significant impacts on future safeguards technologies. Longer device life and/or the luxury of simply consuming more energy for more capability (e.g., security processes) will be a designer's choice. It could be possible that energy for ultra low-power RF devices

Technology	Application	Transmission method	Comment
Ultra-wideband (UWB)	High data rate short range communications. Possible application in PAN (personal area networks. Short range radar and positioning tracking.	Information spread over a very wide (>500Mhz or 25% of the center frequency) bandwidth in the 3.1–10.6 GHz band. Pulse position or time modulation.	Virtually immune to typical multi-path problems. Short range limits possible applications.
Secure Sensor Platform (SSP) System	Low maintenance devices used where very high confidence data collection is required. Interface supports a variety of sensors and seals. Public key based authentication.	FM narrow-band, ultra-low power utilizing infrequent burst communications.	Commercially available since December 2007. Can last several years on a single battery.
RuBee	Asset tracking utilizing passive RFID and low bandwidth communication. Low speed inexpensive technology.	Long Wave (LW) magnetic signals to transfer information below 450 kHz and optimally at 132 kHz.	Final stages of IEEE development.
ZigBee	Currently suitable for simple control networks. May find application in sensor node applications in the near future, especially when energy storage densities increase.	Direct sequence spread spectrum (DSSS) with short data packets. 2.4GHz ISM frequency band.	Requires powered, full-function units to support mesh networking and frequent communication activity requires more power to maintain connectivity.
SoC	Most RF transceiver sets will utilize deeply embedded processors supporting new modulation techniques and security features.	This approach will eventually find its way into virtually all transmission techniques and protocols.	This is an ongoing trend and will become pervasive in the RF industry.

Table 1: Near-term RF communication technologies.

may be able to be parasitically extracted from the environment and the device could operate indefinitely.

Of the presently available radio hardware, the most interesting developments will be in the area of simple, low-cost and low-power SoCs that incorporate the radio, RF packet handling hardware, and a microprocessor for security processes.

Furthermore, the incorporation of protocol software in SoCs, such as ZigBee (IEEE 802.15.4) [14], to simplify application development based upon a standard architecture is likely to be quite useful for safeguards instrumentation for data collection. Being able to leverage off the large hardware development effort that has gone into these SoCs seems beneficial. Depending on the specific requirements and whether some performance can be sacrificed, it may also be beneficial to leverage off the development effort that has gone into a standard such as ZigBee. Traditionally, communications protocols between RF devices have been implemented through custom network solutions.

4. RF modulation: technical considerations for the future

Issues that need to be considered with respect to choosing an RF modulation type for a low-power sensor application are directly related to *modulation spectral occupancy*,[1] *Bit Error Rate* (BER),[2] the *complexity and power consumption of the radio hardware*, and the modulation's *robustness against interference* [1].

Modulation technologies [16] are addressing these issues through the evolution of techniques for low-power RF applications. In the past, On-Off Keyed and Amplitude Shift Keyed (OOK/ASK) were simple modulation techniques utilized in sensor applications, but now many single chip radios designs have migrated to Frequency Modulation (FM). This is because FM offers significantly better interference rejection. There are several FM modulation types and

[1] Spectral occupancy or Eb/No (E bee over en zero) is the measure of signal to noise ratio for a digital communication system. It is measured at the input to the receiver and is used as the basic measure of the signal strength.

[2] BER is the number of bits per unit time that are incorrect.

associated derivations. For example, Minimum Shift Keying (MSK) is a natural evolution of FM, which constrains the transmit spectrum and decreases the spectral occupancy relative to traditional FM techniques. Binary Phase and Quadrature Phase Shift Keying (BPSK/QPSK) take this same approach a bit further and represent an even lower spectral occupancy. However, not many single chip radios currently support these modulation techniques, due to the added complexity and power consumption associated with the demodulation requirements.

Burst spread spectrum receivers are more complex and require more power than any of the aforementioned receivers, but offers more robust interference rejection. This is because the transmission is spread over a broad frequency spectrum. The IEEE 802.15.4 standard (the hardware underlying ZigBee) specifies Direct Sequence Spread Spectrum (DSSS) [17] modulation for communication. The market pull of this standard has driven the commercial development of new SoCs that implement DSSS modulation and demodulation. A major drawback for DSSS is that the receiver draws about 50% more power than a similar narrow band FM transceiver. If the RF communication system can afford this power penalty, the IEEE 802.15.4 may be a good solution that provides the advantages of DSSS.

Ultra-Vide Band (UWB) [11] is another modulation approach that is on the RF horizon and could be a candidate for safeguards application. UWB is a low-energy, short-range, high band-width RF modulation technique that spreads the transmission energy over a very wide band width. UWB has all of the capabilities that more traditional wireless technologies have and, in some instances, may be even better. UWB still must obey the laws of physics and therefore will exhibit the same or similar received spectral occupancy as the previously discussed modulation techniques, but with reduced effective range.

At this point in development, UWB is much more immature than the previously discussed traditional wireless technologies. Most of the development effort for UWB is going into very short-range, high-speed data interfaces targeted to replace wired Universal Serial Bus version 2 (USB2) and the High-Definition Multimedia Interface (HDMI) for home entertainment. The systems currently being developed are not a good fit for safeguards application, and may not offer very much leveraging technology in this area. Presently, the synchronization requirement in UWB receivers takes more time and consumes more power than a traditional radio and is considerably more complex to implement. While this is not problematic in continuous data transmission, it is a significant problem in low-power burst communications. If UWB moves in the direction of lower bandwidth and longer-range burst communications, it might be able to be considered as a replacement for traditional radios in these types of applications. Recently, the United States Federal Communications Commission (FCC)[3] gave permission for UWB transmissions with limits on transmitter power and radiated spectrum. The first realizable UWB application has become a short-range, high-data rate cable replacement. Many UWB developers have progressed from the simple, short, low duty-factor monocycle pulses to a more traditional modulation format called Orthogonal Frequency-Division Multiplexing (OFDM). This may be the best way to implement UWB-based systems while falling within the FCC regulations. Currently this path of evolution does not represent a good fit with safeguards instrumentation applications, but this could change in the future.

5. Integration of RF technologies into existing facilities

There are two fundamental problems with establishing reliable RF communications in an existing facility—*interference* and *path loss*.

Interference sources can be any device that generates Electro-Magnetic Radiation (EMR). This includes equipment with electric motors and welders at one end of the frequency spectrum, to radar and x-ray machines at the other end. All of this equipment can emit broad spectrum RF noise, which causes transmission errors.

Path loss is caused by many factors, but the largest contributors to loss are distance relative to radiated power and obstructions between the transmission and reception antennas. This consideration is what called the *Fresnel zone* [18]. Figure 2 depicts a Fresnel zone under conditions of visual Line-Of-Sight (LOS) without any obstructions between the transmitter and receiving antenna and Non-LOS (NLOS) where there are visual obstructions. Obstructions entering the Fresnel zone of radiation have direct impact on the resulting achievable transmission distance.

Without obstructions, radiated power essentially dictates the reliable relative transmission distance.[4]

[3] The Federal Communications Commission (FCC) is a United States of America government agency.

[4] Transmission distance is also frequency- and equipment-dependant; however, for simplicity of discussion, these aspects of wireless communication will not be discussed.

It must be stated that not all obstructions induce loss. To the contrary, if an obstruction is reflective and in the right location, the phase differences between the primary and reflected waves (multi-path) can be supportive and provide signal gain. However, this is too complicated to predict and should not be depended upon as a system feature. It is best to accept that the influence of a visual obstruction on an NLOS link may be anything from negligible to complete suppression. DSSS-based systems address the NLOS situation by transmitting over individual multiple frequencies, which are reflected and absorbed differently across the band; therefore, many of the RF nulls (RF shadows and subtractive wave phases) are overcome fairly well. Narrow band transmissions require other remedies to address the NLOS issues, such as passive repeaters (highly technical) and active repeaters.

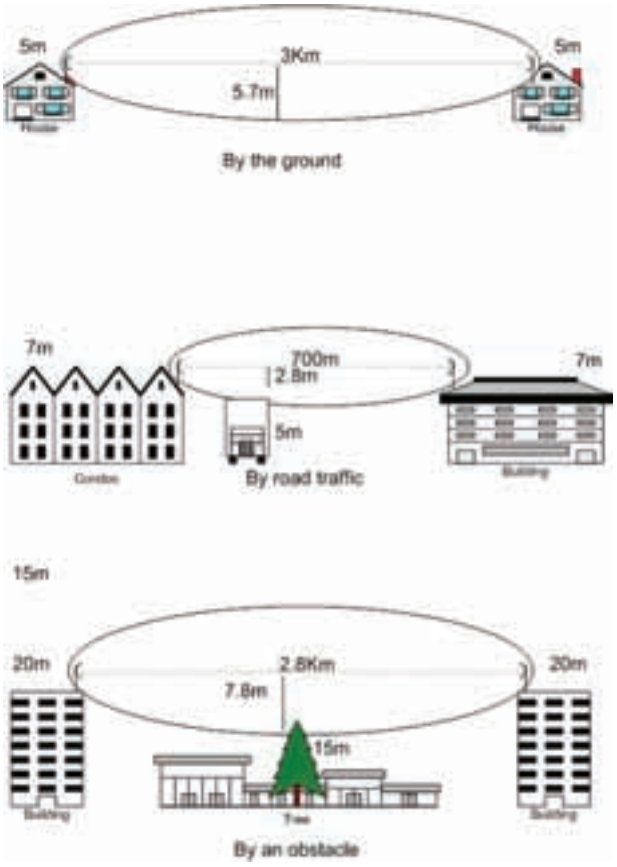

Figure 2: Examples of how the Fresnel zone can be disrupted.

One of the first steps required in an RF system design and deployment would be to characterize the environment. This process should be accomplished through an *RF survey conducted by a reputable team specializing in RF system evaluation*, and will establish the basis of scope for viable applications. This is done by identifying facility characteristics that impact wireless communications. Some of these characteristics can be taken advantage of in the system design and others will require remedies addressing NLOS shortcomings.

6. Designing new facilities for the integration of RF technologies

The best approach to designing a facility to be RF supportive can be summarized as the following:

1. establish the operating environment
2. control the environment
3. monitor the environment.

Generally, the best of all environments for deploying an RF-based communication system would be a facility that emulates an empty *Faraday cage* [19] (Figure 3). This would provide two of the most valuable attributes of an ideal RF operational situation—controlled electromagnetic radiation and no absorbing/reflecting obstructions. Obviously, this facility cannot exist and have any practical purpose.

Basically, a Faraday cage is a metallic enclosure connected to electrical ground that prevents the entry or escape of an electromagnetic field (EMF). In the real world of the RF application space, it is not practical to design a facility to be a Faraday cage. However, to the first order, most nuclear facilities come close. This is because typical construction consists of *concrete surfaces with rebar as reinforcement*. All of the rebar is connected together and usually connected to earth ground. This can establish an RF quiet environment, which is quite suitable for supporting a wide range of RF communications. Figure 3 depicts a simple representation of the Faraday cage concept, which shows how reinforced concrete surfaces can approximate this concept.

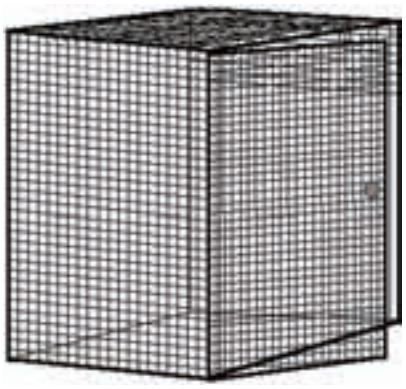

Figure 3: Simple representation of a Faraday Cage.

Other considerations that can be exploited during the design of a new facility are aspects such as the location of power outlets and Ethernet connections. These can be strategically located to support jump-off points from wired to wireless communications and active repeaters. Active repeaters are quite suc-

cessful in changing NLOS [20] situations into LOS. To provide near-uniform RF coverage in expansive volumes filled with RF instrumented items, such as active seals or radiation monitors, distributed antenna systems could be integrated into the wall or ceiling designs. Consideration for utilizing non-conductive (good insulator) partitions for subdividing space can allow virtual LOS communication.

Once the RF environment is established, it is significantly crucial to control the environment. In other words, it is important that unauthorized RF communications do not take place within the defined environment. This does not imply that other RF communications cannot occur within the area; however, the communications must be compatible with each other, typically operating on different frequency bands.

Finally, the best pre-emptive effort to maintain reliable interference-free operation is to periodically monitor the RF emissions. This can be done by utilizing a real-time spectrum analyzer. Today, spectrum analyzers are readily available, handheld, and relatively low cost. Most manufacturers provide software packages that have the analysis tools needed to identify unexpected or unauthorized emission sources, by frequency and power, that could cause communications problems.

7. Summary and conclusions

It is clear that the use of wireless communication to transmit data derived from Safeguards instrumentation has a number of advantages. The evolving nature in the way nuclear facilities are used means that it is quite often necessary to modify the existing infrastructure. Working in a controlled area to install new cabling and equipment is often difficult, takes a significant time to realise and is certainly costly. The benefit of wireless data transfer in terms of cost effectiveness is plain to see, provided that a number of technical challenges can be demonstrated as being overcome.

The first real challenge is in the security issues related to data. It is important that the issues related to a free air transmission of data are addressed and that only those persons authorized to exploit the data can do so. This requirement is primordial as the images coming from cameras for example may have physical security considerations associated with them. The Operator and National Authorities must be actively involved in the design stage of the system to ensure that national security interests are not compromised.

The second challenge relates to the effect on the installed Operator safety or plant control systems, often there are strict requirements related to the use of any emitting device being installed within the facility. It is vital to take into account the possibility that a device could affect the Safety Critical systems. The importance of this concern should not be under estimated, as it is very difficult to model the interference of a device on other systems. The last thing the Operator wants to discover is that a fault has been created during a real emergency. It is interesting to note that the use of mobile phones in aircraft is being reconsidered and they share the common concern noted above.

As a first approach the development of technical solutions which use wireless transmission of data within the facility is the most attractive. The data could be collected at a consolidator node where it is then transmitted via a Local Area Network (LAN) to a central recording unit. This recording unit could then be used to transmit data to the Inspectors office or Headquarters, via Virtual Private Network (VPN). Using this approach it would be possible to utilize the Operator installed and maintained infrastructure as part of the Safeguards infrastructure.

Another consideration is on the choice between custom designed and built verses Commercial-off-the-shelf (COTS) equipment. It should be noted that Inspectorate owned systems generally have an operational life of between 5 and 10 years, depending on the application. In the ‘real’ world technology can change every 2 to 5 years such that if we rely on COTS we may very quickly have a problem with obsolescence. To train staff, (both technicians and inspectors), replace instrumentation and obtain the necessary authorizations for use over such a short timescale are probably beyond the capabilities of the Safeguards inspectorates at this moment in time.

The use of wireless communications by Safeguards authorities has many attractions; there is also however a number of significant challenges which need to be overcome. It is certain that this type of technology will become more mainstream in the future and therefore easier to implement in nuclear facilities. The final solution however needs to be developed in full transparency with the key actors e.g. Inspectorates, National Authorities and Operators.

8. References

[1] F. Braina, J.G.M. Gonçalves, M. Heppleston, B. Schoeneman, K. Tolk, C. Versino (2007), *Wireless Communications for Monitoring Nuclear Material Processes – PART I: Context and Technologies*, ESARDA Bulletin No. 36, 32-41

[2] K. Tolk, M. Aparo, C. Liguori, A. Capel (2006), *Design of Safeguards Equipment for Authentication*, Symposium on Interna-

tional Safeguards – Addressing Verification Challenges, October 16-20.

[3] T. Karygiannis, B. Eydt, G. Barber, L. Bunn, T. Phillips (2006), *Guidance for securing radio frequency identification (RFID) system,* Recommendations of the National institute of standards and Technology (NIST), Special Publication 800-98.

[4] Special issue on RFID of 'Multi-System & internet security cookbook', n°33, septembre 2007, pp. 28-67.

[5] Symmetric cryptography, http://en.wikipedia.org/wiki/Symmetric_cryptography

[6] Advanced Encryption Standard, http://en.wikipedia.org/wiki/Advanced_Encryption_Standard

[7] Public key cryptography, http://en.wikipedia.org/wiki/Public_key_cryptography

[8] Elliptic curve cryptography, http://en.wikipedia.org/wiki/Elliptic_Curve_Cryptography

[9] IEEE 802.11, WiFi, http://en.wikipedia.org/wiki/IEEE_802.11

[10] SoC, http://en.wikipedia.org/wiki/System-on-a-chip

[11] Ultra-wideband, http://en.wikipedia.org/wiki/Ultra-wideband

[12] B. Schoeneman, S. Blankenau (2005), *Secure Sensor Platform (SSP) for Materials' Sealing and Monitoring Applications*, Proc. of the 46th Annual Meeting Proc. of the Institute of Nuclear Materials Management.

[13] RuBee, http://en.wikipedia.org/wiki/RuBee

[14] ZigBee, http://en.wikipedia.org/wiki/Zigbee

[15] RUNES project, http://www.ist-runes.org/

[16] Digital modulation, http://en.wikipedia.org/wiki/Digital_modulation#Digital_modulation_methods

[17] Direct Sequence Spread Spectrum, http://en.wikipedia.org/wiki/Direct_sequence_spread_spectrum

[18] Fresnel zone, http://en.wikipedia.org/wiki/Fresnel_zone

[19] Faraday cage, http://www.physics.gla.ac.uk/~kskeldon/PubSci/exhibits/E3/

[20] Non line of sight propagation, http://en.wikipedia.org/wiki/Non-line-of-sight_propagation

Technical sheets

Nuclear Mass Spectrometry

1. Objective of the technique

Mass spectrometry is the most commonly used Destructive Analysis (DA) technique in nuclear safeguards for measuring the isotopic composition and isotopic amount content (concentration) of uranium, plutonium and other actinides in a sample.

2. Presentation of the technique

2.1. Principle of measurement / Definition of the physical principle

For an analysis using a mass spectrometer a given sample is first

- introduced into the ion source (by various techniques, e.g. gas flow, nebuliser, sample magazine) and
- ionized (by various techniques, e.g. hot metal filament, plasma, ion beam),

The ions of the sample are then

- accelerated (usually by high voltage),
- separated by mass (by various techniques, e.g. using magnetic and or electric field, or time-of flight system), and
- detected (by various techniques, e.g. Faraday cups or Secondary Electron Multiplier).

Depending on the type of instrument and on the application, the detected ion current intensities obtained from an analysed sample can be used to calculate the elemental or isotopic ratios of the sample from which the elemental or relative isotope abundances (isotopic composition) are derived.

As an example, in Fig.1 the mass spectrum of a natural uranium sample is shown. By ramping the magnetic field strength, the mass range from m=233-239 a.m.u (atomic mass unit) is scanned through a Fara-

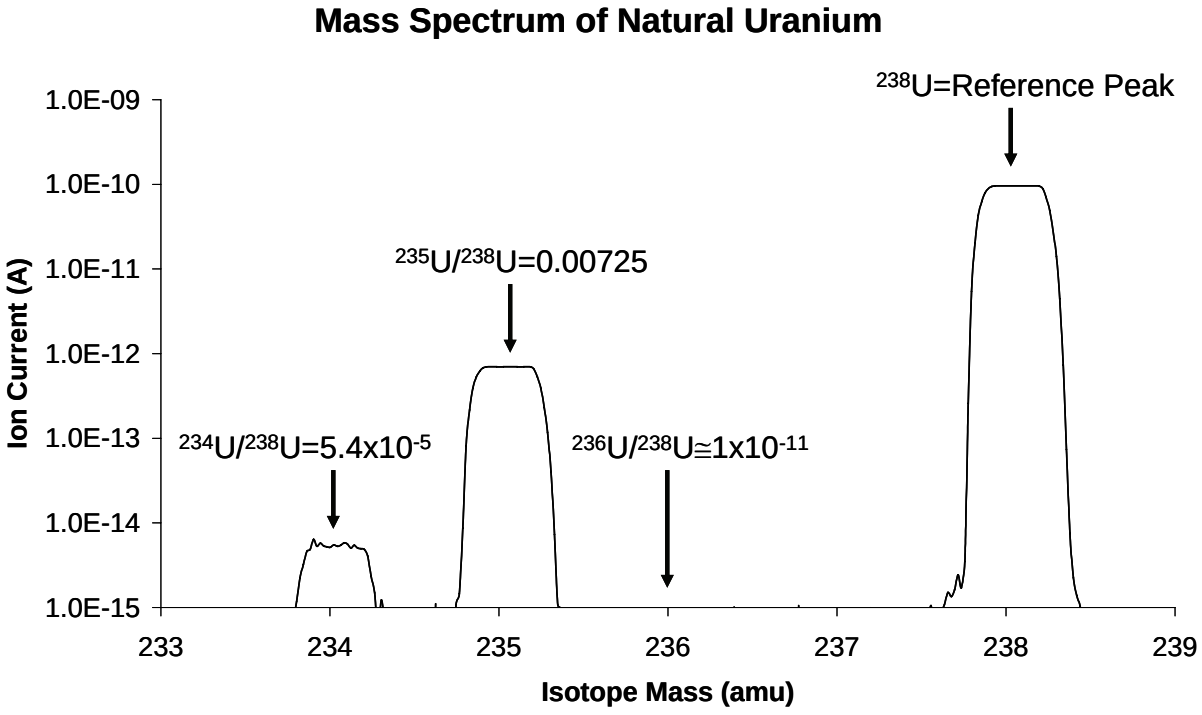

Figure 1: Mass Spectrum of Natural Uranium.

day cup detector. The isotopic abundances are proportional to the measured ion currents displayed on the graph. The ion currents as measured for a sample of natural uranium are displayed in logarithmic scale, in order to cover the large dynamic range of the uranium isotopic abundances.

Many nuclear material samples contain different elements with isotopes of similar mass, for example ^{238}U and ^{238}Pu. In such cases, either the mass spectrometer must have sufficient mass resolution to be able to distinguish between these isotopes, or the sample must be treated chemically (e.g. by ion exchange chromatography) to separate the different elements prior to the mass spectrometric measurement.

A second complication in the application of mass spectrometry is the potential presence of mass fractionation effects. These arise because different isotopes of the same element do not undergo the processes such as evaporation, ionisation and detection at the same rate, for given ion source conditions. Depending upon the mass spectrometer used and the procedures applied, mass fractionation effects may need to be corrected for through the measurement of certified standards and application of a correction factor derived from these standards to all results obtained.

The element amount (concentration) within the sample may be calculated through a process known as isotope dilution mass spectrometry (IDMS, see below). This involves the addition of a known amount of one particular isotope ("spike") of the element of interest. By mass spectrometric determination of the isotopic composition of a sample before and after the addition, the change in isotopic composition can be used to calculate the concentration of the element within the original sample.

2.2. Types of mass spectrometers

In order to cover various analytical needs about half a dozen of variants of mass spectrometry have been developed using different combinations of ionisation method, mass separation and detection. This has crystallized in a variety of instrument types which are useful particularly for modern nuclear safeguards:

- AMS accelerator mass spectrometry [1]
  is applied in particular, when large isotope abundance ratios are to be measured, for example ^{236}U/^{238}U in natural uranium (Figure 1).
- ICPMS: inductively coupled plasma mass spectrometry [2], is applied for measuring elements with high ionisation potential and as multi-element analysis method, both for quantitative elemental analysis and isotope ratio analysis. The 3 most important subtypes are:
  - ICP-QMS: quadrupole-ICPMS
    the most common type of ICP-MS in routine elemental quantification. Applied for isotope ratio measurements when no high precision (better 0.1 % RSD) is required. Sometimes equipped with a reaction cell to resolve spectral interferences.
  - ICP-SFMS sector field ICP-MS (also referred to as HR-ICPMS (high resolution ICPMS))
    for ultra low level elemental quantification and isotope ratio measurements. A magnetic sector field ICPMS enables isotope ratio measurement with a precision better than ICP-QMS. The high resolution capabilities allow the resolution of spectral interferences up to a mass resolution of m/Δm of 12.000).
  - MC-ICPMS: multi-collector-ICPMS
    allows simultaneous detection of several isotopes, thus enabling high precision measurements. Modern instruments are equipped with higher mass resolution capabilities (m/Δm of about 4.000) for interference separation (see ICP-SFMS).
- GSMS: gas source mass spectrometry [3] is applied for highly accurate isotope ratio measurements of gaseous samples, in particular for uranium hexafluoride UF_6, an important compound in the enrichment process.
- RIMS resonance ionisation mass spectrometry [4]
  allows the selective ionisation of predefined species within a mixture of elements. RIMS makes use of lasers of defined wavelength for the selective ionisation.
- SIMS: secondary ion mass spectrometry [5]
  allows isotope ratio measurements of individual particles and has found widespread application in particle analysis for safeguards.
- TIMS: thermal ionization mass spectrometry [6] is the most widely applied analytical technique for analysing samples of nuclear material.

A more comprehensive publication about isotope mass spectrometry is given in [7].

2.3. Analytical tasks in nuclear mass spectrometry and associated types of mass spectrometers

- Elemental abundances: these are most frequently determined using TIMS or ICPMS in combination with IDMS (Isotope dilution mass spectrom-

Sample Type	Measurand	Instrument Type
U in solution	Isotopic composition	AMS, ICPMS, GSMS, RIMS, TIMS
U in solution	Isotopic comp., in particular $^{235}U/^{238}U$ ("major ratio")	ICPMS, TIMS[1]
U in solution	Isotopic comp., in particular $^{234}U/^{238}U$ and $^{236}U/^{238}U$ ("minor ratios")	TIMS, ICPMS, AMS[2] *(only $^{236}U/^{238}U$), RIMS3*
U in UF_6 gas	Isotopic comp., in particular $^{235}U/^{238}U$ ("major ratio")	GSMS[4]
U or U-oxide particles	Isotopic composition	SIMS (directly), TIMS (if loaded on filament) ICPMS (using LASER ablation)
Pu in solution	Isotopic composition	ICPMS, TIMS
Pu or Pu-oxide particles	Isotopic composition	SIMS (directly), TIMS (if loaded on filament) ICPMS (using LASER ablation)
Other actinides	Isotopic composition	TIMS, ICPMS

[1] in TIMS usually by total evaporation, to minimize mass fractionation effects
[2] AMS has the lowest possible background for ^{236}U
[3] limited capabilities
[4] in GSMS using double standard method

etry [8]: sample is mixed with certified isotopic spike, the change in isotopic composition is used to determine elemental amount content)

- Isotopic amount content (isotopic assay): most commonly determined using TIMS, ICPMS, RIMS, in combination with IDMS (Isotope dilution mass spectrometry)
- Isotope abundance ratios: done using all mass spectrometer types mentioned in section 2.2., but for particular tasks/sample types particular types are being used preferentially, see table below:

For almost all instrument types and techniques, certified reference materials are needed for instrument calibration and/or quality control. In particular, they may be required in order to correct for mass fractionation effects. Reference materials certified for isotopic amount content and/or isotopic abundance ratios can be obtained from laboratories specialised in their certification, including the IRMM (http://www.irmm.jrc.be/html/homepage.htm) or NBL (http://www.nbl.doe.gov).

3. Useful references

[1] P. Steier, R. Golser, W. Kutschera, V. Liechtenstein, A. Priller, A. Valenta, C. Vockenhuber, (2002) Nuclear Instruments and Methods in Physics Research B, 188, 283-287.

[2] M.B. Andersen, Stirling, C.H., Potter, E.-K., Halliday, A.N., (2004), International Journal of Mass Spectrometry, 237, 107-118

[3] W. De Bolle, G. Muschenborn, P. De Bièvre, (1985), ESARDA 7th Annual Symposium; Liege, Belgium; 21-23 May.

[4] K. Wendt, N. Trautmann, (2005), International Journal of Mass Spectrometry 242, 161–168

[5] G. Tamborini, M. Betti, V. Forcina, T. Hiernot, B. Giovandone, L. Koch, (1998), Spectrochemica Acta Part B 53, 1289-1302.

[6] S. Richter, S. A. Goldberg, (2003), International Journal of Mass Spectrometry, 229, 181-197.

[7] I.T. Plazner, (2001), "Modern Isotope Ratio Mass Spectrometry", published by John Wiley & Sons, 1 edition January 1, 2001, ISBN-13: 978-0471974161, 530p.

[8] P.De Bièvre, H.S. Peiser, (1997), Fresenius' Journal of Analytical Chemistry, 359, 523-525

Call for papers

31st ESARDA annual meeting

Vilnius, Lithuania
26-28 May, 2009

The 31st ESARDA Annual Meeting will be a symposium on "Safeguards and Nuclear Material Management". It will be held in **Vilnius, Lithuania, 26-28 May 2009.**

This symposium will be an opportunity for research organisations, safeguards authorities and nuclear plant operators to exchange information on new aspects of international safeguards and non-proliferation recent developments in safeguards-related research activities and their implications for the safeguards community.

The symposium is anticipated to include a number of contributions from internationally-renowned authorities in the field.

The following two themes will give **direction** to the symposium:

- Integrated safeguards and a changing Europe
- Euratom safeguards in a global context

Contributions may cover but are not necessarily limited to the following **topics / disciplines**:

- Safeguards concepts (policies, perspectives, limitations, Strengthened and Integrated Safeguards, State and Regional Systems, Quality Assurance Approach)
- Euratom system: Beyond State level approach (European Union level approach)
- Nuclear safeguards implementation: experience, evaluation; plant specific experience on techniques, inspections and operations
- Experience in the implementation of Strengthened Safeguards systems and Integrated Safeguards
- Non-proliferation and future issues (Cut-off Treaty, CTBT and disarmament, excess materials, sub/cross-national threats, etc.)
- Export – Import control (and related activities)
- Synergies with other verification regimes (radiological, chemical, biological, dual use, etc.)
- Containment and surveillance methods and techniques; interface between safeguards and protection methods
- Co-operative programmes in safeguards; ESARDA
- Human resources and knowledge management issues including public information
- Measurements techniques and standards
- Data and information evaluation methodology, remote monitoring and secure data transmission
- Integrated measurement and monitoring systems
- Materials control and accounting, auditing and information systems
- Illicit Trafficking and borders control
- Applications of GPS and GIS and information security

Presentations with original content are strongly encouraged.

Papers will either be presented orally or in poster sessions. Contributions must be written and presented in English.

In submitting a contribution, the authors confirm having their organisation's approval for public release.

Authors are requested to submit the abstract(s) of their contribution(s) for reviewing by e-mail to:

ESARDA Symposium Secretary
European Commission Joint Research Centre
e-mail: esarda2009@jrc.it

in the following format:

- title, author(s), affiliation;
- an abstract in English of about 300 words to be used for paper selection (maximum 1 standard page).
- a maximum of 5 **keywords** related to the topic(s) of their paper
- whether they would like to present it orally or as a poster.

Abstract submission deadline:
21st November 2008

The abstracts will form the basis for accepting or rejecting contributions and allocation between paper and poster sessions. The Technical Programme Committee will decide whether or not an accepted paper will be presented as an oral presentation or as a poster. Authors will be informed of their decisions by the end of February 2009. The compendium of the accepted abstracts will be available on the ESARDA website and distributed at the meeting.

By sending an abstract, the authors agree to submit a paper suitable for inclusion in the symposium's proceedings and to present that paper at the symposium. The authors are requested to follow the "**instructions for authors**" available on the ESARDA web site for presenting abstracts and full contributions. Final papers / posters must be submitted in electronic format (e-mail).

Papers must be submitted electronically no later than:
15 May 2009

The Editorial Committee reserves the right to decline to accept any abstract or paper submitted after the deadlines.

The proceedings will be published shortly after the meeting and a copy sent to each participant.

Adequate space can be arranged for commercial presentations / exhibitions. For further information on commercial displays, please contact directly the symposium secretariat.

Registration forms, a copy of the programme and further information about the meeting will be available on the ESARDA web-site in due course:

www.jrc.ec.europa.eu/esarda

Ad Memoriam

Sergio Finzi

The ESARDA Secretariat was informed that Sergio Finzi, ESARDA chairman from 1983 to 1985, passed away in February 2008.

Sergio Finzi was born in 1928. He joined the European Commission in 1960, after a few years spent working in industry (EDISON, AGIP Nucleare) and as Assistant to Professor Silvestri at the Politecnico of Milan. In Ispra, he first worked in the Division of Nuclear Technology. In 1965 he was nominated Director of the Engineering Department, and then of the Programme Management. In 1974 he served as ad interim Director General of the Ispra site. Until 1982, he was Director of the Applied Science and Technology Department. In Brussels, he became Director for Nuclear Safety Research (DG XII) first, and then Director for Nuclear Safety (DG XI).

He retired in 1993.

The Secretariat extends its condolences to his family.