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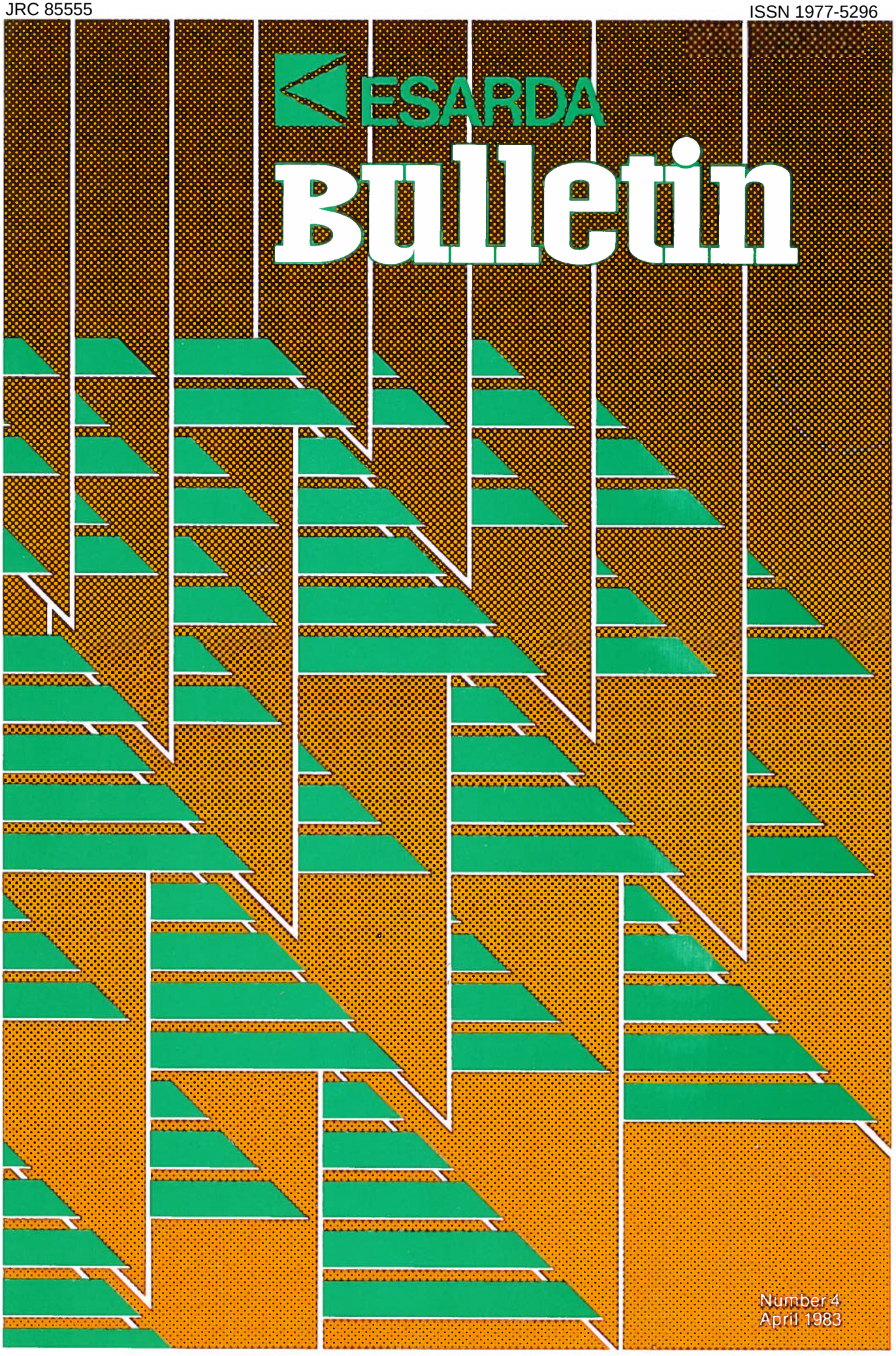

Number 4
April 1983

Editorial

Scanning the issue the reader can discover that it is mainly dedicated to the International Symposium on Recent Advances in Nuclear Material Safeguards organized by the IAEA in Vienna last November. As a matter of fact the issue contains 1) a survey of the symposium written by R.J.S. Harry, with his personal comments, 2) the full list of the abstracts of papers produced or co-authored by the countries participating in the ESARDA association and 3) the paper presented at the symposium by W.L. Zijp as chairman of ESARDA. The latter two items are reprinted with permission from "Nuclear Safeguards Technology - 1982" published by the IAEA.

The conclusion of Harry's paper allows me to extend the concept that "still a lot of technical work has to be done in order to optimize international safeguards" by remembering that ESARDA and INMM are actively operating in this field and the Annual ESARDA Symposium is one of the most specific means of clarifying the real necessities concerned. I would therefore like to give some details on the 5th Annual ESARDA Symposium which will be held in Versailles, France on 19th - 21st April 1983. Many of the readers have by now received the programme so that they know that the main theme is this year the "Interaction between Safeguards Authorities and Operators". This theme is essential for setting up good conditions for the application of international safeguards. Briefly the symposium will run on nine oral sessions in parallel with poster presentations which are scheduled so that similar subjects do not clash. Oral and poster sessions will deal with the subjects of Safeguards Perspectives, Operator's Experience, Plant Oriented Working Groups Experience, Data Evaluation Methodology and Nuclear Material Management. Chairmen and secretaries of sessions are instructed to adhere to the main theme of the symposium and will therefore direct the discussions accordingly. I wish to give here evidence to the first and last sessions, both containing invited papers. The first one will treat general problems from the point of view of the Commission of the European Communities (CEC), the International Atomic Energy Agency (IAEA) and the Commissariat à l'Energie Atomique (CEA). The last one will deal with the more specific problem of R&D in Support of Safeguards as seen by CEC, the U.S. DOE and the INMM. In addition I wish to draw attention to the invited paper opening Session 2 presenting the very important subject of Safeguards Interfaces seen from the point of view of the Euratom Safeguards Directorate.

The concept of engaging the specialists of the field in order to optimize international safeguards will be further investigated in the 6th Annual ESARDA Symposium which will be held in Venice in the central week of May 1984. Details will be given with the call for papers.

Returning to the scanning of the issue the reader can see that it starts with a paper of W.L. Zijp, chairman of ESARDA in 1982, describing the experience of one year of the association. It is an interesting report aimed to give information about the work going on in ESARDA. I take the occasion to add that the chairman of ESARDA for 1983 is Mr. E. Bastrup-Birk from Energistyrelsen, Denmark.

The issue also contains a paper of G. Bardone and F. Pozzi on the safeguards techniques applied by the operators to fuel reprocessing facilities. This paper is a compendium of technical developments under application in two different plants at the opposite extremes of the Italian peninsula. These are being developed by ENEA, the Italian Committee managing nuclear and alternative energies and replacing the preceding committee for nuclear energy (CNEN).

A paper from the Nuclear Research Centre of Karlsruhe, KfK, is presented by H. Ottmar on a reliable and timely nuclear material accountancy system for reprocessing plants. This is a technical paper describing an integrated system based on X-ray fluorescence and K-edge absorptiometry containing comments on the utility of the instrument in view of a design for routine operation.

Only one report on the activities of the ESARDA working groups is reported in this issue. The next issue will include the activity reports of most of the ESARDA Working groups.

Looking forward to seeing many of you in Versailles, I wish finally to remind you that the ESARDA Bulletin is open to contributions from ESARDA and non-ESARDA countries.

L. STANCHI

P.S. The Editorial Board would like to express its gratitude to Mr. A.G. Hamlin who has now retired from our community. His duties have been assumed by Mr. B.W. Hooton. The Editorial Board would like to thank Mr. Hamlin not only for his services in editing the ESARDA Bulletin but in general for all his work connected with the ESARDA association of which he was a member of the Steering Committee, and at the same time it would like to acknowledge his great human qualities and spirit of co-operation.

The Year 1982 in Retrospective

W.L. Zijp
ESARDA Chairman during 1982

The past year 1982 was the first calendar year under the new ESARDA contract, since it came into force in the middle of 1981.
Under the new contract the Steering Committee can have more than thirty members, and consequently it may lose some of its flexibility and readiness for decision.

For that reason an Executive Board was created with the following tasks:

A. The preparation of the agenda for the Steering Committee, and of proposals on which the Steering Committee will have to take decisions;
B. The management of day-to-day life, paying attention to several topics, such as:
   - proposals for the creation of new working groups, or ad hoc committees,
   - development and implementation of an internal and external information system,
   - the ESARDA-Bulletin,
   - relationships with external organizations (such as IAEA, INMM),
   - review of R&D orientation of ESARDA,
   - review of publications under auspices of ESARDA,
   - organization of annual symposia and topical meetings.

The Board has 5 members from different organizations (rotating according to an accepted scheme among the contract signatories), and a permanent secretary (J. Ley, JRC-Ispra, Italy, the secretary of ESARDA). The Board had three meetings (Brussels, 2 March 1982; the Hague, 8 June 1982; London, 4 October 1982), and prepared two Steering Committee Meetings (Brussels, 3 March 1982; London, 5 October 1982). The Board had extensive discussions on the work of ESARDA Working Groups, the R&D programmes, the organization of annual symposia (Petten 1982; Versailles 1983; Venice 1984), the Bulletin and some procedural matters. It turned out that the institution of the Board improved the decision making of the Steering Committee.

The Steering Committee itself reviewed the activities of all Working Groups. A new Working Group on Mathematical-Statistical Techniques was constituted, while the Working Group on Isotopic Correlation Techniques was reoriented as "Working Group on Reprocessing Input Verification". At this point it should be mentioned that two important ESARDA publications dealing with this topic were published in 1982 : the proceedings of the Stresa Symposium on "Isotopic Correlation and its Application to the Nuclear Fuel Cycle" (issued as ESARDA-14), and the final report on the "Isotopic Correlation Experiment" (available as report ESARDA-2/81; EUR 7766; KfK 3337).

With a view to the IAEA Safeguards Symposium in November 1982, it was decided to give the annual meeting in Petten more the character of a specialists' meeting than of a general symposium. The theme was "Harmonization and Standardization in Nuclear Safeguards". The proceedings were published within one month after the meeting as ESARDA-15. Valuable contributions to the subject were made by many of the more than 100 participants. It was appreciated that the time schedule of this meeting allowed ample time for technical discussions.

In 1982 the issues 2 and 3 of the ESARDA Bulletin were published, and we hope that the Bulletin fulfils its purpose as a means to promote interest in the activities of the Working Groups and the members of ESARDA. Here the important work of Mr. L. Stanchi (JRC-Ispra), the editor of the proceedings and the Bulletin, should be gratefully acknowledged.

Futher I would like to mention Mr. A.G. Hamlin (NMACT, Harwell), who has retired from his official duties. We thank him for his stimulating role in the past few years, and wish him many good and healthy years. Thanks are also due to Mr. R. Venchiarutti (previously CNEN, now ENEA), who for more than one decade represented his organization in the field of safeguards and who has now accepted a diplomatic appointment in Australia. Near the year's end British Nuclear Fuels Limited (BNFL) applied to become partner to the ESARDA contract, and a formal procedure to that purpose has been started. The BNFL initiative is welcomed since ESARDA has emphasized its desire to include operators of nuclear facilities in its ranks. For more information on this new orientation of ESARDA, see the chairman's paper presented at the 1982 Symposium in Vienna, which paper is reproduced in this issue of the Bulletin.

The interaction between safeguards authorities and the operators will be the main theme for the ESARDA Symposium at Versailles, 19-21 April 1983. I am looking forward to this symposium, also to meeting many of you, readers of the ESARDA Bulletin.

The Specialists' Meeting 1982 showed that ESARDA functions as a forum, not only for its European members, but also for participants from all regions of the world.

The preparations of the 1983 Symposium are well in progress, and I expect that this event will further contribute to the tradition of fruitful co-operation within our Association, to the benefit of international safeguards and public confidence in peaceful applications of nuclear energy.

International Symposium on

Recent Advances in Nuclear Material Safeguards

Vienna, Austria, 8-12 November 1982

A Short Review

R.J.S. Harry
Netherlands Energy Research Foundation, ECN, Petten

General Remarks

From 8 to 12 November 1982 the IAEA organized its fifth general safeguards symposium in Vienna. The title "Recent Advances in Nuclear Materials Safeguards" reflected quite well the contents of about a hundred papers that were presented. About 250 participants from about 25 Member States all over the world, the Commission of the European Communities and the IAEA used this opportunity to exchange information and to discuss new developments in safeguards.

A limited number of poster sessions was held, which allowed for a more direct contact between interested participants and the authors. The oral presentations were given in uninterrupted morning or afternoon sessions without coffee breaks, but the use of parallel sessions undoubtedly reduced the number of listeners to several presentations because one was obliged to choose (and unfortunately missed indicentally some other interesting presentations).

In a late stage of the programme planning 15 papers from the U.S.A. had to be withdrawn from the conference and the stimulating discussions and the thought provoking ideas from our colleagues from the U.S.A. were sadly missed.

Other contributions involving authors from the U.S.A. were presented in cases where the papers were the result of an international co-operation with the IAEA, the Commission of the European Communities and/or with other Member States. This illustrates clearly a trend in the research and development of safeguards, namely that international co-operation in the early stages of development and in testing under field conditions gets an increasing emphasis.

Many written contributions were made available during the conference, and the booklet containing all the "extended synopses" of the proposed papers supplemented this advance information on the contents of the contributions.

Some Basic Concepts

In the previous IAEA Symposium in 1978 two new ideas were promoted for safeguards, namely the concept of "extended containment and surveillance" and the concept of "near-real-time accountancy". The critical studies which were undertaken in the meantime have shown that for the application of such a containment and surveillance approach a lot more work has to be done on the containment and surveillance techniques before they can be integrated into a reliable safeguards system. Experiments with the implementation of near-real-time accountancy measures have resulted in positive judgement on the capabilities of this technique. In a paper which analysed critically and systematically the safeguards system as a whole, it was concluded that the two safeguards concepts of accountancy and containment and surveillance could not be separated as individual options, but they have to be dealt with as of a complementary nature.

A special methodology was suggested to assess the effectiveness of safeguards. In the process of making this assessment in an objective way several steps have to be taken that are of a more subjective nature. Essential input conditions to the methodology are given by the safeguards objectives quantified via threshold amounts and conversion times into detection goals. Some of the participants seemed to challenge the goals that have been in use for some time now.

For credible safeguards it is essential that the goals can be met, and especially in large future reprocessing plants where this might be a very difficult task, in terms of costs. Therefore one would refer back to the more political factors that could be used to allow for a more relaxed application of safeguards depending upon factors like the other fuel cycle capabilities in a State and the application of Full Scope Safeguards. The IAEA's Standing Advisory Group of Safeguards Implementation was asked to study on the "fuel cycle approach" and to review critically the safeguards objectives.

Safeguards Research and Development

It was interesting to note that the total amount of money that the States and the Commission of the European Communities are spending in the safeguards research and development in "support" or "co-operation" programmes for the IAEA exceeds considerably the total annual safeguards budget of the IAEA. These programmes are a real substantial factor in the development of safeguards. The exercise and tests in field conditions form an essential contribution to the development of an internationally credible safeguards system.

In the field of destructive analyses the preliminary results of the IDA '80 experiment were presented. This ESARDA sponsored project gives a good assessment of the state of the art in the isotope dilution analysis thanks to the possibility of making an intercomparison between 31 laboratories. The original way of presentation also indicated the years of experience that could be attached to the results, and it was evident that a long experience does not give an absolute guarantee for excellent results. One of the few other papers in the field of destructive analyses described how the results of automatized titrations were directly written on magnetic tape for further processing. This is a trend one can discern by all measurement techniques.

There was quite remarkable interest in isotopic correlation techniques for irradiated fuels. It seems that this safeguards technique is beginning to find its proper place in safeguards and nuclear material management. Plant operators and safeguards inspectors reported upon various applications which included many years of experience.

In the field of non-destructive analysis there is a tendency to use also in plant installed, and even operator owned equipment. Special attention has to be given in such circumstances to the so-called "authentication" of the results of measurements for safeguards acceptance. The advent of microprocessors has led to automation and

simple-to-operate equipment, but there is still a gap between the promises from research and development and the real performance in the inspection situation. Demonstrations and intercomparisons should help to close this gap and to supply data on performance for the statistical evaluation of the results.

Improvements and practical tests are reported in the field of containment and surveillance, in particular in relation with the CANDU-reactor. The Cerenkov-detector, which is used to detect the blue glow from irradiated fuel under water, can now also be used in a fuel storage facility where the normal illumination remains. In the field of identity verification several different ideas were described. In the application of seals, one is striving towards a standardization of seals and equipment to verify the seals. The ambitious RECOVER (REmote COntinuous VERification) system is tested now with the co-operation of several facilities, connected by ordinary telephone calls with the central equipment in the IAEA headquarters. Its future use is not yet clear.

For the statistical evaluation of safeguards data many test procedures are possible. An intercomparison of various tests which are sometimes used is not always easy or appropriate due to differences in underlying assumptions in the mathematical models. It is therefore advisable to make a choice for a standardized approach. One difficulty in assessing various models is that information on the quality of the measured data is often not available.

Practical Experiences

Many papers described real experiences with safeguards, in which also the standpoint of the operator was quite well represented. Real operating plants have been the subject of safeguards studies like e.g. the CANDU-reactor, the Tokai Mura reprocessing plant, several fuel fabrication plants and, in the special case of the Hexapartite Safeguards Project, all known commercial centrifuge enrichment plants. On this last project the chairman reported how inspectorates and technology holders work together to design the appropriate technical basis for safeguarding these facilities.

The safeguards burden for the operator was described very detailed in one paper, where it was stressed that in the low enriched uranium fuel fabrication the additional need for an annual physical inventory taking led to an effort of 63.5 operators working days involved in the placing of about 28,000 labels. The same paper stated that in a mixed oxide fuel production plant the extra operator burden for safeguards added up to 8% of the fabrication costs.

Another paper described how the inventory taking in a store was reduced from about 60 working days to about 5 days by the application of bar coded labels and computerization. It is clear that in the application of safeguards, cost effective improvements can be made if a good co-operation between safeguards authorities and the operator is established from the design phase onwards, including the design of the system of data gathering and processing and the sampling and verification procedures. At the final session of the symposium one participant remarked that probably the computerized auditing of the Viennese supermarket was more advanced than the auditing in many stores of the high technology facilities of the nuclear fuel cycle.

Conclusions

Many points have not been mentioned in this review, but it has become clear that safeguards is not any more a "new field" where a lot of "bright ideas" are born and investigated. Now the system is at work and it is a matter of adaptation, improvement, optimization. A good interaction with plant operators for field tests as well as for optimization of the safeguards system is essential.

Now the attention will have to focus on the way in which all results will fit into a coherent system. This will include the critical studies on the fuel cycle as a whole, on the data collection and evaluation systems, and on the fundamental questions on the safeguards goals in relation to the efforts involved to maintain an effective detection system for the diversions of the given size, in the appropriate time interval.

This symposium was successful in clarifying that still a lot of technical work has to be done in order to optimize international safeguards; and that researchers, developers and operators all over the world are eager to contribute to that aim.

Papers authored or co-authored by people of ESARDA Countries presented at the

International Symposium on Recent Advances in Nuclear Material Safeguards

Vienna, Austria, 8-12 November 1982

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"Nuclear Safeguards Technology - 1982", published by the IAEA

SM 260/5

R. Beedgen (KfK, Karlsruhe)

Truncated Sequential Test Procedure Using the CUMUF Statistic for a Timely Detection of Diversion

A sequential test procedure for materials accountancy is presented where a final decision at the end of a defined number of balance periods is made concerning the loss of material. It is shown how false-alarm probability at single-steps influences the false-alarm probability for the total time period.

SM 260/11

M.C. Moxon, E.W. Lees (UKAEA, Harwell)

Non-Invasive Determination of the Uranium Content in a Centrifuge Plant Dump Trap

The amount of uranium hexafluoride which has been absorbed onto NaF in the dump trap of a centrifuge enrichment plant has been measured using gamma-ray transmission techniques. This technique measures the transmission of ^{60}Co gamma-rays across a diameter of the trap and does not require the removal of the trap from plant. Because of the order of magnitude increase in the gamma-ray attenuation coefficient of UF_6 over any other material that should be present in the dump trap, a 10% accuracy in quantifying UF_6 content should be readily obtainable.

SM 260/12

D. Williams, R.J. Riley (UKAEA, Harwell)

Principal Features of a Design for a Portal Monitor for Nuclear Safeguards

A portal monitor for nuclear safeguards aims to detect the removal by personnel of significant amounts of special nuclear materials from a plant. Unlike similar devices intended for security use, it is not specifically intended to identify the individuals attempting such removal. Since diversion may be with the connivance of the plant operator, the monitor must be automatic, difficult to evade and both resist and record any determined attempt at tampering. Its output would normally be a log of events available only to an accredited inspector.

SM 260/14

A.G. Hamlin, F.J. Walford (UKAEA, Harwell)

In-Process Hold-Up as a Measure of Safeguards Significance

This paper examines the use of the in-process hold-up itself, as a measure of safeguards significance.

It is argued that for any process plant it is possible to define design limits for in-process hold-up, outside which the plant will not operate, or will operate in a detectably abnormal manner. It follows, therefore, that if the in-process hold-up can be derived at frequent intervals by input-output analysis from the start of the campaign, the only diversion that can be made from it during that campaign is limited to the quantity necessary to move the apparent in-process hold-up from its normal operating condition to the upper limiting condition. It also follows that detection of this diversion is as positive for protracted diversion as for abrupt diversion.

If that part of the in-process inventory that is only measurable by input/output analysis has an upper operating limit that differs from its normal operating limit by less than a significant safeguards quantity of the material in question, the Agency's criteria for both quantity and timeliness can be met by a combination of input/output analysis to determine in-process hold-up during the campaign, together with a material balance over the campaign.

The paper examines the possibility of applying this measure to process plants in general, discusses means of minimising the in-process inventory that must be determined by input/output analysis, and the performance required of the input and output analysis. It concludes that with current precision of measurement and with one input and one output batch per day each measured, the method would be satisfactory for a campaign lasting nearly a year and involving 6 tonnes of plutonium.

The paper examines the considerable advantages in verification that would arise from limiting safeguards analyses to the two points of input and output.

SM 260/15

F.J.G. Rogers (UKAEA, Harwell)

The Use of Micro-Computer with In-Field Non-Destructive Assay Instruments

The first version of the NMACT portable passive neutron coincidence counter (PPNCC) was equipped with individual scaler which recorded the count rates from both shift register and variable dead time counter pulse processing circuits. All subsequent data processing to calculate the plutonium mass of the sample was carried out on a programmable calculator.

Two years of practical experience with this system indicated that we would benefit by interfacing the counter outputs directly into a micro-computer, which could perform all the control and calculate functions required to operate the system efficiently.

This paper describes the computerised system and its operating features in detail, and discusses the improvements introduced in a two year operating period.

The experience gained from the use of a micro-computer in this role has led to other uses in conjunction with a portable gamma spectrometer, and for giving a superior record of the output of installed safeguards instruments such as doorway monitors and platform monitoring.

SM 260/20

M. Darrouzet, G. Fréjaville, P. Marimbeau, J. Pinel (CEA Cadarache), J. Regnier (COGEMA, Cap de la Hague)

Etablissement d'un Bilan d'Entrée de Combustibles. Réacteurs à Eau par Mesures non Destructives Gamma et Neutron

Deux méthodes de mesures non destructives sur des assemblages irradiés ont été développées pour les usines de retraitement.

Une méthode de spectrométrie gamma permet de déterminer le taux de combustion et le temps de refroidissement à partir des raies du ^{134}Cs, ^{137}Cs et ^{144}Ce.

Ces corrélations sont établies à partir de calculs "assemblage" effectués avec des codes de réacteur permettant de prendre en compte l'histoire du combustible en réacteur.

Une seconde méthode repose sur le comptage du flux neutronique émis par les assemblages. Une corrélation calculée permet de relier la masse de plutonium au flux mesuré.

Une campagne de mesures réalisée sur 3 lots d'assemblages C.N.A. ayant subi 3 cycles en réacteur a permis de vérifier la faisabilité de ce type de mesures dans un cadre industriel et d'effectuer une comparaison des résultats avec les valeurs annoncées par la centrale et les valeurs obtenues par analyses lors des dissolutions.

Bien que les particularités des assemblages C.N.A. soient défavorables, nous avons pu déterminer le taux de combustion des assemblages à mieux que 10% et le temps de refroidissement à mieux que 90 jours.

Les masses de plutonium déterminées par la mesure du flux neutronique donnent des résultats encourageants. Après normalisation à un assemblage référence, l'on obtient par rapport aux analyses dissolveurs : $+1,1 \pm 1.3$ % pour le lot 4 et $-0,005 \pm 0,65$ % pour le lot 5.

Une optimisation de l'appareillage et une prise en compte plus fine de l'historique d'irradiation devraient permettre sur des assemblages PWR actuels d'atteindre les précisions recherchées (5% sur le taux de combustion, 1% sur la masse de plutonium).

Un nouvel appareillage tenant compte de l'expérience acquise est en cours de montage. Il permettra, par des mesures d'assemblages PWR et BWR, de tirer des conclusions définitives, mais nous pensons, d'ores et déjà, que ces méthodes joueront un rôle très important dans le suivi des matières dans les usines de retraitement.

SM 260/21

M Desprès, J. Morel, B. Chauvenet, J. Legrand (CEA, Saclay)

Mesure de la Composition Isotopique du Plutonium et de la Teneur en Uranium ou en Plutonium

Un dispositif a été développé assurant automatiquement les mesures de concentration de plutonium et/ou d'uranium par la méthode d'atténuation différentielle de deux faisceaux de photons gamma et la détermination de la composition isotopique du plutonium par spectrométrie gamma.

L'appareil est constitué d'un ensemble mécanique assurant la mise en place par transfert pneumatique des sources d'irradiation, du volet obturateur et d'un système informatisé permettant le pilotage de cet ensemble et l'acquisition et le traitement des données. Des résultats accompagnés de leur incertitude globale de l'ordre de quelques pourcents pour les deux méthodes, sont présentés.

SM 260/22

J.P. Bariteau, W. Donato (CEA, Cadarache), G. Dean, J. Monier, M. Neuilly (CEA, Fontenay-aux-Roses), B. Thaurel (CEA, Saclay)

Industrial Applications for Safeguards Use of Neutronic Passive Measures for a Fast Determination of the Quantity of Plutonium

One important problem of those who are in charge of special nuclear materials is to characterize samples of plutonium (for example, quantity, quality, ...). They need many instruments and methods to do this quickly and surely. Neutron counting can be used for this purpose.

First of all theoretical neutronic emissions of PuO_2, detection instruments and different counting methods are briefly presented.

Two devices were built for our special applications :

- a large device which can measure very large quantities (3-12 kg) of plutonium in a nuclear factory (CEA, Cadarache) by using neutronic gross-counting.
- a portable device used by inspectors which can measure smaller and different samples of PuO_2 (100 mg - 3 kg) contained in small chests (upper limit: 0.4x0.4x0.3 m).

Shift register electronic is convenient for both applications. It gives a gross-counting and a real coincidence counting. A small calculator (HP 87 S) allows an automatic treatment of measurements.

Results of experiments on PuO_2 samples are presented and discussed. These results show that a gross-counting is adequate when there is always the same geometry and a negligible background : precision is better than 5%. When these conditions are not met, coincidence counting is necessary; accuracy on these measurements is better than 15%.

SM 260/23

G. Boisdé, P. Guillot, J. Monier, J.J. Perez (CEA, Fontenay-aux-Roses)

Non-Destructive Measurements of Uranium in Solution Using a Portable Optical Fibre Photometer

In low concentration ranges, French nuclear control inspectors do not have suitable measurement techniques at their disposal. Research and development project led to the design and construction of a portable optical fibre photometer intended for non-destructive measurements of uranium.

The spectral characteristics of uranium VI solutions are established for acidities ranging from 0.2 to 5 n in nitric medium. The absorbance-nitrates relationship is presented for different uranium concentrations. Measurements are taken at three wavelengths, one used as an internal reference, and the remaining two to solve a two-equation system with two unknowns.

The instrument called TELEPHOT 3N, uses small amounts of solution (25 ml) and has a response time of about one minute. Its performance has been checked and the measurement errors evaluated on some hundred samples.

This remote measurement technique will be used for plutonium and to monitor industrial processes.

SM 260/24

J. Bouchard, A. Giacometti, R. Girieud (CEA, Cadarache), M. Aries, P. Patigny (COGEMA, Cap de la Hague)

Quatre Ans d'Expérience d'Utilisation des Corrélations Isotopiques Calculées dans l'Etablissement du Bilan d'Entrée à l'Usine de la Hague

Les corrélations isotopiques calculées sont utilisées depuis plus de quatre ans par l'usine de retraitement de La Hague pour l'établissement et le contrôle de ses bilans d'entrée.

Les masses d'uranium et de plutonium entrant dans l'usine sont déterminées par la méthode du bilan gravimétrique. Celui-ci utilise le taux de combustion obtenu par corrélation isotopique calculée ainsi que le rapport Pu/U mesuré au dissolveur après une vérification par recoupement avec les valeurs obtenues par corrélation.

De plus, une vérification de tous les paramètres nécessaires à l'établissement de ces bilans qu'ils soient d'origine physique ou chimique, est systématiquement effectuée à l'aide d'un jeu de constantes de cohérence interne permettant la détection d'éventuelles anomalies dans les données de dissolution.

La qualification des corrélations isotopiques calculées a été effectuée lors de l'interprétation des analyses de nombreux échantillons représentatifs de combustibles irradiés ainsi que des résultats d'expérience d'irradiation d'isotopes séparés dans des spectres de réacteurs à eau.

La précision obtenue a été améliorée en prenant en compte dans les calculs neutroniques les effets inhérents au premier coeur du réacteur et en choisissant un jeu de fonction de corrélation qui atténue par effets de compensation les diverses perturbations de l'historique d'irradiation.

Les résultats obtenus à l'usine de La Hague sur près de 600 tonnes d'UO_2 retraités lors de l'utilisation des corrélations isotopiques calculées incitent par leur grand nombre et surtout par leur qualité élevée, à proposer l'extension de cette méthode à d'autres usines de retraitement. Celle-ci peut être mise en oeuvre par l'exploitant lui-même ou par des organismes de contrôle nationaux ou internationaux dans le cadre d'un contrôle de garantie.

SM 260/25

P. Mauchien, P. Cauchetier (CEA, Fontenay-aux-Roses), J. Grison (COGEMA, Cap de la Hague)

Dosage de Traces d'Uranium dans une Usine de Retraitement par Spectrofluorimétrie sur Solution

Pour établir précisément les tableaux d'inventaire et satisfaire aux exigences des garanties, comme pour contrôler la bonne marche d'une usine de retraitement, il est nécessaire de déterminer la teneur en uranium dans de nombreuses solutions où il se trouve à l'état de traces. Nous proposons, à cette fin, une méthode utilisant la propriété que présentent les solutions d'uranyle de fluorescer lorsqu'elles sont irradiées en lumière ultraviolette.

Après un bref rappel théorique, nous passons en revue les paramètres qui influent le plus sur la mesure : le milieu, la température, la nature de la matrice, le choix de la longueur d'onde de la radiation incidente. Nous en déduisons la nécessité d'effectuer la mesure par étalonnage interne en utilisant la méthode des ajouts dosés et l'intérêt d'obtenir le spectre de fluorescence qui permet d'authentifier la présence d'uranium.

Nous décrivons les applications de cette méthode à l'usine de la Hague où elle est utilisée depuis octobre 1981 par des équipes postées, en particulier pour contrôler les points suivants : l'acide d'attaque avant introduction du combustible, le pied de colonne du premier cycle, la solution carbonatée de lavage du solvant, les solutions de l'unité de traitement des effluents. Son utilisation se généralise à toute l'usine, même au contrôle de l'uranium dans l'oxyde PuO_2.

Cette méthode évite l'utilisation de réactifs organiques tels que la pyridine et, dans bien des cas, des séparations longues et génératrices d'effluents. Par exemple, le dosage est possible par simple dilution dans les concentrats de produits de fission et dans des solutions de plutonium dans lesquelles le rapport Pu/U atteint 10^3.

En solution pure, avec l'appareillage utilisé, la limite de détection est de quelques $\mu g l^{-1}$ dans la cuve de mesure. En général, la précision est de quelques pourcent.

SM 260/26

M. Luca, T. Goujard, G. Retali, R. Hagemann (CEA, Saclay)

Utilisation d'un Etalon Interne Constitué d'un Mélange d'Isotopes 233 et 236 de l'Uranium pour Améliorer la Justesse des Analyses Isotopiques et Celle des Dosages par Dilution Isotopique de l'Uranium

La dilution isotopique associée à la spectrométrie de masse à thermoionisation est l'une des techniques utilisées pour la mesure précise des quantités d'uranium contenues dans les solutions de combustibles irradiés. La justesse des résultats obtenus est, en particulier, directement liée à la justesse de l'étalonnage de la solution de traceur (uranium 233) utilisée. Cette dernière est généralement étalonnée par dilution isotopique à partir d'uranium métallique naturel de pureté garantie, et la mesure qui limite la justesse de cet étalonnage est celle du rapport 233/238, dont la valeur peut être entachée d'une erreur systématique due à une correction imparfaite du fractionnement isotopique. Afin de corriger les effets du fractionnement isotopique qui limitent la précision des analysis isotopiques, une méthode basée sur l'utilisation d'un mélange étalon $^{233}U/^{236}U$ a été mise au point.

En mélangeant l'étalon interne à des standards isotopiques de rapports 235/238 certifiés, il est possible de déterminer le rapport 233/236 par normalisation interne à une valeur dont la justesse est celle des standards utilisés. Cet étalon interne est ajouté à l'échantillon à analyser et les rapports 233/236 mesurés sont normalisés à la valeur précédemment déterminée et le même facteur de normalisation est appliqué aux rapports 235/238 mesurés. Ces derniers rapports sont alors connus avec la même justesse que celle des standards isotopiques.

La même méthode peut être appliquée à la mesure des concentrations d'uranium par dilution isotopique en employant un traceur double 233/236 à la place du traceur simple actuellement utilisé. Le fractionnement isotopique sur les rapports mesurés 233/238 peut alors être corrigé avec précision car il est connu à partir de la comparaison des rapports 233/236 mesurés à la valeur 233/236 théorique déterminée précédemment.

Une comparaison des résultats obtenus par la méthode classique et par la méthode proposée ici sera présentée. L'utilisation généralisée de cette méthode pour l'étalonnage des solutions de traceur et les mesures effectuées sur les échantillons réels permettrait certainement de supprimer ou au moins de limiter de manière très appréciable les écarts de justesse qu'ils soient accidentels ou systématiques.

SM 260/27

R. Bödege, E. Leitner (DWK, Hannover), R. Avenhaus (Hochschule der Bundeswehr, München)

Efficiency of a Material Accountancy Safeguards System Including Extended C/S-Measures

In the IWG-RPS Overview Report extended containment/surveillance (C/S) measures have been defined as those safeguards measures which attempt to provide assurance that all transfers are through key measurement points (KMP). In that report it has been suggested that further work on the models on material accoutancy (MA) and C/S-systems in a complete safeguards approach should be continued at least to the point where well founded and detailed analyses on the individual assurances and their combinations can be carried out. This paper presents the results of the efforts of various research groups as well as of the authors in order to solve some of the problems indicated above.

As shown in earlier papers, under appropriate assumptions the MUF-D-test is optimal for a safeguards system based on material balance and data verification principles, where material has been

diverted by methods the diverter considers most favourable. In this paper the justification for choosing the MUF-D-test and applying it also to C/S-measures will be explained, as will be its use in detecting shipments of special nuclear material (SNM) through access points not initially declared for this purpose. The analyses made hitherto will be expanded and the generalized test statistics together with the optimal detection probability will be determined once again for cases where the diverter uses a strategy which is most advantageous to him. This will show the maximum detection probability increase obtainable by this method.

SM 260/28

U. Schuett, F. Schinzer, K. Stoefen (NUKEM, Hanau)

Experience with a Computerized Accountancy System at a Fabrication Plant for Highly Enriched Uranium

In 1975, a computerized accountancy system for safeguards was installed and in the mean time has been improved at the NUKEM Fabrication Plant at Hanau, F.R. of Germany. The nuclear material is controlled on the basis of batch data during its flow through the different storage and fabrication areas.

In a nearly real-time accountancy manner all material movements are reported to the central nuclear material accountancy group of the plant who feeds the data into the computer. Advantages are a quick overview and an immediate listing of total and itemized inventories of plant sections. The physical inventory taking can be performed by a factor 4 faster than without computer. For users no special computer knowledge is necessary, the system is easy to manage and delivers suitable data to the international inspectors, who did not request a system change. But it is planned to improve the system by more terminals and additional software features.

SM 260/29

D. Gupta (KfK, Karlsruhe)

International Workshop on the Near-Real-Time Accountancy Measure Overview Report

An International Workshop on the near-real-time accountancy (NRTA) measure was established in December 1980 to investigate the capabilities and limitations of this measure for a large-scale reprocessing facility. The present overview report summarizes the activities and the results of this workshop as of July 1982. After establishing the process and accountancy data base for a 1000 t HM/a reference reprocessing facility, the workshop developed simulation models for the sequential generation of data for throughput and inventory of plutonium in the process material balance area (MBA). A well defined set of boundary conditions and parameter values for measurement uncertainties and loss patterns were established, on the basis of which a number of sequential statistical test procedures were evaluated. One important condition for the application of the NRTA measure was the stipulation that routinely measured Pu inventories in process tanks only, would be used, since more than 95% of Pu inventories in the process MBA are in these tanks. About 12 kg of Pu, expected to be the normal inventory in six pulse columns, was assumed to be constant. In spite of the simplifications made and the fact that mainly simulated data were used, these investigations permit the conclusion that the NRTA measure provides a greater sensitivity in terms of the amounts wich can be detected and the timeliness of detection, than the conventional material accountancy. Since measurements are restricted to process tanks only, routinely available measurement techniques can be used. The main thrust of R&D activities has to lie in the practical demonstrations of this measure under operating conditions, some of which are already under way.

SM 260/30

H. Wuerz (KfK, Karlsruhe)

A Non-Destructive Measurement System for Spent LWR Fuel Assemblies

A spent LWR fuel assembly monitor based on combined active and passive neutron counting is presented. The monitor primarily developed for the objective of criticality control in the head end of a reprocessing plant can be used for safeguards applications as well. It can be used for an independent verification of the shipper's data.

An extended demonstration programme for German LWR fuel assemblies was performed. Up to now 72 spent uranium fuel assemblies of 4 PWR and 3 BWR reactors and 20 spent MOX fuel assemblies were measured. The initial enrichment of the uranium fuel assemblies varied between 1.9 and 3.29 % ^{235}U.

Results and experiences of the demonstration programme are presented. The characteristics of the assay of LWR fuel assemblies are discussed. Accurate and representative burnup dependent neutron emission data of spent LWR fuel are given. These data are compared with results of destructive analysis and with theoretical results obtained from burnup codes. Finally, a description of the non-destructive measurement system is given.

The demonstration programme has shown that without knowledge of any fuel assembly's data the burnup of uranium fuel assemblies can be determined with an uncertainty of ± 1200 MWd/tHM and the initial enrichment of uranium fuel assemblies with an accuracy of ±5%. The demonstration programme moreover showed to what extent the irradiation history influences the results of NDA measurement. Using these data and accuracies the total plutonium content can be determined from isotopic correlations with an accuracy better than ±0.3 kg/tHM for PWR and ±0.5 kg/tHM for BWR fuel assemblies.

SM 260/31

C. Brückner (KfK, Karlsruhe), S. Crutzen, B.C. d'Agraives (CEC, Ispra), H. Heger (VDEW, Frankfurt), L. Pachl (WAK, Kahl)

Experience Accumulated to Date in the Fuel Assembly Seal Demonstration at the Kahl Experimental Nuclear Power Station

A demonstration programme on fuel assembly sealing systems is being performed in the Kahl Experimental Nuclear Power Station within the framework of the IAEA support programme of the Federal Republic of Germany.

Its aim is to demonstrate the sealing of LWR fuel assemblies.

Ultrasonic seals developed at the Ispra Joint Research Centre are used after completion of the licensing procedure. Fuel assemblies fabricated by Kraftwerke Union/RBU and by EXXON Nuclear Corporation (ENC) were prepared for eactor loading in November 1981. The seal is adapted to the special design features of the fuel rod.

Effort is directed at drafting the criteria to be met by an in-situ re-verification procedure, in which the ultrasonic signal of a seal is compared with the primary pattern by calculation of a correlation factor. Thinking along lines also incorporates developments by the Sandia Laboratories, U.S.A.

SM 260/32

E. Mainka, J. Neuber, H. Wertenbach (KfK, Karlsruhe), R. Berg, E. Stojanik, B. Fueger (WAK, Karlsruhe)

Fully Automated, Wavelength Dispersive X-Ray Fluorescence Analysis of Uranium and Plutonium in Reprocessing Plant Solutions

A fully automated, computer operated X-ray fluorescence system is described. Based on experiences gained with earlier systems and taking into account a plant operator's requirements a robust, pneumatically operated modular system was constructed.

The problems encountered in adapting the system to glove-boxes, the features of the sample preparation, control of the system and the calculation of the results are outlined.

A comparison between a scanning and a 7-channel X-ray fluorescence spectrometer was experimentally established. Measurement results are reported.

SM 260/33

W. Beyrich (CEC, Karlsruhe), W. Golly, G. Spannager (KFK, Karlsruhe), P. De Bièvre, W. Wolters (CEC, Geel), L.A. Machlan, J.W. Gramlich, J.A. Fassett (NBS, U.S.A.)

The IDA-80 Measurement Evaluation Programme on Mass Spectrometric Isotope Dilution Analysis of Uranium and Plutonium : Some Preliminary Results

The IDA-80 measurement evaluation programme is concerned with the determination of uranium and plutonium in reprocessing input solutions by mass spectrometric isotope dilution.

The participants were requested to report, as a part of the programme, the element concentrations and isotope abundances of uranium and plutonium in two sample solutions "B" and "R". Solution B had been prepared by dilution with nitric acid from a sample of original feed solution taken at the WAK-reprocessing plant and contained fission products. The reference solution R was prepared by CBNM and did not contain fission products.

The paper presented includes graphs of the above requested results. They show the spread of the values reported by the participating laboratories as compared against certified values supplied by CBNM, Geel and NBS, Washington. Results of laboratories claiming to perform isotope dilution analyses frequently or even continuously for more than 5 years, are marked especially.

The data presented should be considered as preliminary. They must still be discussed by the participants at the meeting scheduled in 1983.

SM 260/34

H. Ottmar, H. Eberle, P. Matussek, I. Michel-Piper (KfK, Karlsruhe)

Qualification of K-Absorption Edge Densitometry for Applications in International Safeguards

The distinguished features and performances of K-absorption edge densitometry using continuum X-ray transmission sources for measurements of special nuclear material concentrations in solutions are briefly outlined. It is shown that the measurement accuracy and reliability, the simplicity of operation, and the ease of authentication of K-absorption edge densitometry can notably enhance safeguards technology, and qualify the technique for safeguards applications in plutonium reprocessing and nitrate-to-oxide conversion facilities. An advanced solution-assay system combining simultaneous K-absorption edge densitometry and K-X-ray fluorescence analysis is presented. Prospective applications of this versatile instrument for verification measurements in a reprocessing plant are discussed.

SM260/35

D. Sellinschegg, U. Bicking (KfK, Karlsruhe)

MUF-Residuals Tested by a Sequential Test with Power One

Near-real-time materials accountancy is an ongoing safeguards development to extend the current capability of IAEA safeguards. The evaluation of the observed "Material Unaccounted For" (MUF) time series is an important part in a near-real-time materials accountancy regime. The maximum capability of the sequential data evaluation procedure is demonstrated by applying this procedure to the

material balance area of the chemical separation process of a reference reprocessing facility with a throughput of 1,000 tons heavy metal per year, as an example. It is shown that, compared to a conventional materials accountancy approach, the detection time as well as the detection probability is significantly improved.

SM 260/36

M.J. Canty (KFA, Jülich), G. Spannagel, F. Voss, A. Berliner (KfK, Karlsruhe)

Computer Simulation of the Process MBA for a 1000 t Reprocessing Facility

Current investigations of the application of near-real-time material accountancy methods to safeguarding large scale commercial reprocessing facilities suffer from a lack of availability of plant data. Few commercial plants are in operation, and those that are do not provide routinely and in near-real-time the required material balance data. Detailed process simulations on digital computers can provide an alternative until real in-process simulations on digital computers can provide an alternative real in-process inventory and throughput measurement results become available. In addition they provide a relatively inexpensive means of experimenting with different operating schemes and material accountancy strategies with a view to optimization of safeguards effectiveness. In particular, the robustness of statistical test procedures may be investigated.

A computer program has been developed for simulating the operation of the process MBA of a 1000 t reference reprocessing facility running on a Purex flowsheet. The program describes dynamically all Pu flows and hold-ups throughout the process, beginning at the input accountability tank and ending at Pu-nitrate load out. It writes "true" values for volumes, concentrations and flow rates to an output file for subsequent simulation of throughput and in-process inventory measurements taken for near-real-time accounting.

In order to produce data that are as realistic as possible not only the individual process units were modelled in detail, close consideration was also given to operating modes (rework, independent operators for each Pu-cycle, etc.). The program serves primarily as an input for testing the effectiveness (detection probability, timeliness, false alarm rate) of evaluation procedures for detecting abrupt and protracted material losses under "realistic" conditions.

SM 260/37

I.C. Campsie, H.D. Rattray (UKAEA, Springfields)

Development of a Vitrification Technique for Rolled Seamed and Other Containers

One of the projects included in the U.K. Safeguards Programme has been the development of a method of establishing the identity and integrity of rolled seam cans, i.e. finger prints. This technique can also be applied to other containers, instruments and equipment.

The requirement was to permanently mark the surfaces of the containers with a "signature" which was not easily forged, even with sophisticated equipment. It was proposed to strategically position the unique "signature(s)" so that they would be damaged by any attempt to enter, change the contents and reseal the container. Therefore on reading the "signature" the damage would be detected and the auditors alerted on the need to examine the container's contents.

It has been established that surface texture traces through vibro-etched serial numbers produce unique profiles, i.e. signatures which can only be reproduced when retracing the same path through the original etched serial numbers.

The system has the advantage that the serial number that can be visually read yet contains a unique non-forgeable signature which can be traced onto a paper chart or converted into a numerical code.

Work has concentrated on developing an automatic method of profile analysis using a pattern recognition technique which extracts a number of the largest peaks for each profile and translates the horizontal distance between the peaks into a numerical code.

A feasibility study in which some 100 profiles were analysed has been completed. Its main conclusions are :

- The data reduction exercise can be done compactly and reliably from the profile signals
- Implementation of a small micro-computer will be straightforward
- Analysis of at least 4-6 peaks will be required to provide a numerical code which has sufficient verification confidence
- No start or finish position is required, the system compensates for horizontal shift.

Future work will consist of building a prototype verification unit for the analysis and using this unit to analyse a large number of profiles to provide statistical confidence levels on the probability of false rejection. It is also intended to manufacture a hand held traversing unit.

SM 260/41

E. Dermendjiev (IAEA, Vienna), W. Lauppe (KFA, Jülich), F. Schinzer (NUKEM, Hanau)

Mass Verification of UF_6 Stored in 30" Cylinders

A Load Cell Based Weighing System (LCBS) has been tested for safeguards mass verification of low enriched UF_6 stored in 30" cylinders. It was found that the LCBS is simple in operation and reliable in performance. The accuracy has been estimated to be about 1 kg. Some modifications of the original system have been tested. The results of numerous weight measurements of 30" UF_6 cylinders at NUKEM show that the LCBS can be successfully used by safeguards inspectors as an instrument for independent UF_6 mass verification.

SM 260/48

W. Jaek, E. Münch, B. Richter, G. Stein (KFA, Jülich)

Safeguards Aspects of Multinational Facilities

In order to take account of the many and varied economic, ecological and sociological requirements, particular mechanisms for co-operation must be applied so that a transfer of nuclear energy from the industrialized countries to the developing countries is facilitated. In this context, aspects of non-proliferation and supply assurance attain a special significance.

A possible interaction with safeguards in a multinational facility should be analysed, particularly the way in which by implementing INFCIRC/153, §81 by a type of internal control on the basis of "interrelation with other states" inspection effort and expense could be reduced in such a facility and thus a multinational facility could be given a safeguards credit. Previous studies, particularly by the IAEA within the framework of the study on "Regional Nuclear Fuel Cycle Centres" as well as at the INFCE, have not taken this aspect of a safeguards credit into consideration but have rather regarded the multinationalization or internationalization of the fuel cycle as an additive measure.

With respect to the uses of nuclear energy in industrially developing countries, a phased plan is suggested for the development of international co-operation. A typical feature is that each form of co-operation is subject to the national legislation of the host country in question.

The results presented here cannot be regarded as final conclusions but rather as an initial approach by means of which possible interdependences between various countries and their interaction with the effectiveness of safeguards can be qualitatively represented.

SM 260/50

H.J. Arenz (CEC, Luxembourg), L. Koch (CEC, Karlsruhe), S. Schoof (KfK, Karlsruhe)

A Comprehensive Fuel Nuclide Analysis at the Reprocessing Input

The composition of spent fuel can be determined by various methods. They rely partially on different information. Therefore, the synopsis of the results of all methods permits a detection of systematic errors and their explanation.

Methods for determining the masses of fuel nuclides at the reprocessing input point range from pure calculations (shipper data) to mere experimental determinations (volumetric analysis). Inbetween a mix of "fresh" experimental results and "historical" data is used to establish a material balance.

Deviations in the results obtained by the individual methods can be attributed to the information source which is unique for the method in question. The methodology of the approach consists of three steps : by paired comparison of the operator analysis (usually volumetric or gravimetric) with remeasurements the error components are determined on a batch by batch basis.

Using the isotope correlation technique the operator data as well as the remeasurements are checked on an interbatch basis for outliers, precision and bias.

Systermatic errors can be uncovered by interlab comparison of remeasurements and confirmed by using historical information. Experience collected during the reprocessing of LWR fuel at two reprocessing plants prove the flexibility and effectiveness of this approach. An example is presented to demonstrate its capability in detecting outliers and determining systematic errors.

SM 260/51

G. Guzzi, A. Federico (CEC, Ispra)

Use of a Transportable Quadrupole Mass Spectrometer for Isotope Analysis of Uranium Hexafluoride

JRC is carrying out a work in the field of Safeguards of fissile materials aimed at developing a transportable quadrupole mass spectrometer with a twofold aim :

- the isotope and impurity analysis of UF_6 with an electron impact ion source;
- the isotopic analysis of U and Pu of different chemical forms by a special thermal ionisation source.

The first phase of development is almost completed. It is thought worthwhile to present the main characteristics of the prototype instrument and the results obtained during experiments conducted at enrichment plants. The main specifications of the instrument are complemented by a description of the recently developed parts of the equipment. The procedures followed during the measurements are summarized and the results obtained in three in-field tests for $^{235}U/^{238}U$ ratio measurements and for minor isotope determinations are presented and discussed. The second phase of development is at present under study by two firms specialised in the field of mass spectrometry.

SM 260/52

F. Argentesi (CEC, Ispra), J.P. Shipley (Los Alamos)

Loss Pattern Identification in Near-Real-Time Accountancy Systems

One important aspect of the loss detection in nuclear materials accountancy is related to the unavailability of statistical methods specifically designed for analysing near-real-time accountancy data. However, in the case of multiple periods the onset and extent of loss are not known a priori; consequently, specially developed procedures that accommodate this uncertainty, are essential.

Starting from the already developed statistical methodologies for multiple balances, an extension to achieve controllable properties is considered in this paper. The extension proposed is based on the analysis of all the possible contiguous subsequences of data, augmented by pattern identification techniques to assist in the detection and classification of loss scenarios stemming from whatever cause.

On the basis of Monte Carlo simulations a restricted set of reference patterns and frequencies has been generated and used as templates for the classification procedure. The classification algorithm is based in the concept of minimum distance between the observed alarm pattern and a reference loss pattern. The results obtained seem to indicate improved performance over that obtainable from more common statistical methods.

SM 260/54

L. Bondar (CEC, Ispra)

Passive Neutron Assay

An assessment of the various systems for passive neutron assay has been carried out. Four different types of time-correlation analysis were considered in the assessment, carried out by a Plessey minicomputer. These are the Variable Dead-Time counter, Shift Register, Pulse-to-Pulse and the Pulse Fluctuation Analysis. A coherent treatment of the various systems has been introduced. A common interpretation model based on elementary probability theory and basic nuclear data is applied to all the various time-correlation analyses. Measurements have been performed using a well-counter and the Euratom Time Correlation Analyser on various Pu sources.

SM 260/57

F. Brown (Dept. of Energy, London)

The Hexapartite Safeguards Project

Commercial exploitation of the gas centrifuge enrichment process began in earnest in early 1970's. The general principles for fulfilling safeguards obligations were easily established, since the physical simplicity of the process permits accurate material accountancy measurements. Elaboration of this basic approach has proven to be considerably more difficult, the commercial sensitivity and non-proliferation aspects of the process raising particular concerns.

The Hexapartite Safeguards Project (HSP) was initiated in November 1980, the participants being Australia, Euratom, the IAEA, Japan, the "TROIKA" States (F.R. of Germany, the Netherlands and the United Kingdom) and the United States of America. Four working teams were established, and each team met at approximately four-month intervals for a period of two years. The result has been a plan for a practical demonstration of a Limited-Frequency Unannounced-Access' strategy. It is confidently expected that this exercise will successfully demonstrate the practicability of the HSP conclusions and vindicate the considerable investment of time, effort and goodwill by all participants in the project.

SM 260/59

W. Gmelin, P. Bommelle, B.W. Sharpe, B. Love (Safeguards Directorate, Luxembourg)

Recent Developments in the Implementation of Euratom Safeguards

The Euratom safeguards system is based legally on the 1958 Treaty of Rome establishing the original Community of 6 (now 10) countries. Under this safeguards system, the Commission has, inter alia, "to satisfy itself that any particular safeguarding obligations assumed by the Community under an agreement concluded with a third state or an international organization are complied with" (art. 77b). The practical implementation, of safeguards within the Community is significantly influenced by the requirements of:

(a) the three different agreements between the Community, its Member States and the IAEA, concerning the application of IAEA safeguards to some or all of the civil nuclear materials in the Community, and

(b) the various agreements between the Community and certain third countries, concerning inter alia the application of safeguards within the Community to nuclear materials supplied, directly or indirectly, by these third countries.

Within the past four years significant developments have occurred in both groups of agreements. The NPT (Non-Nuclear Weapon State) Agreement of course continues in force, with the inclusion since 1981 of Greece in the group of 8 NNWS Member States partly to the Agreement; the Euratom/U.K./IAEA Agreement which has been negotiated following the British "Voluntary Offer" in the context of the NPT entered into force in 1978; the Euratom/France/IAEA Agreement concerning the implementation of IAEA safeguards in France entered into force in 1981. An agreement between Australia and the Community was signed and came into force in 1981, and a further amendment to the Euratom/Canada Agreement came into force also in 1981.

The Euratom safeguards organisation is the only multinational safeguards organisation in the world, and currently has a staff of some 120 inspectors, with appropriate administrative support, and can draw for research and development work on the resources of the Community's Joint Research Centre.

The recent changes in inspection techniques, particularly in relation to Non Destructive Assay techniques, and the implementation of Containment and Surveillance measures, are described. A description is given of the experience gained in recent years in the operation of "Joint Teams" of Euratom and IAEA inspectors in certain plants as well as the continuing experience gained under the normal regime, using the observation principle, as foreseen in the respective agreements.

SM 260/64

W. Stanners, R. Schenkel, M. Crosbie, D. Landat (Safeguards Directorate, Luxembourg)

Euratom In-Field Experience with Non-Destructive Assay of Plutonium

This paper presents recent advances in the verification of plutonium using NDA-techniques in the field. The examples given are not routine applications of NDA-techniques by Euratom inspectors, but serve to show how special requirements - either from the operators side, or from the safeguards directorate side - can be approached under field conditions using standard equipment, such as the SAM II/SNAP system, the VDC and the HLNCC.

1. In a Pu store, total neutron measurements (using SAM II/SNAP) have been performed on individual birdcages in their storage positions. The source strength s, was determined from the counts c, of the detector obtained at position n and from a factor f_{in} giving the contribution of source i to the counts c, at detector position n. Analysis of the data shows that the consistency between measured and calculated source strengths is as good as the results obtained by similar measurements done on single containers in a location remote from the store.

2. Large drums (about 0.6 m diameter and 1.75 m high) with natural and enriched MOX-plutonium were measured with the HLNCC. The detector was dismantled and rearranged around the drum, to "see" as much of the drum as possible. The efficiency for this very unusual geometry was about 0.33% for total and 0.00047 % for spontaneous fission neutrons. Within the counting statistics error of about 0.4% for total neutrons and about 19% for coincident neutrons, the results gave confirmation of the operators declarations.

3. Plutonium end product samples from a reprocessing plant have been measured with a VDC and an HLNCC to overcome transport problems and delays in obtaining analytical results. Comparison of the results against the operators declarations give agreement to within about 2.4% for total and about 2.8% for coincident neutrons. The method is useful for short response time purposes and it also provides safeguards authorities with a basis to reduce the frequency of chemical analysis on samples by application of a random sequence or on the basis of only verifying outliers from NDA-measurements.

SM 260/65

F. Argentesi, R. Benoit, M. Cuypers, S. Guardini, G. De Grandi, M. Franklin, K. Müller (CEC, Ispra), A. Rota (CEC, Luxembourg)

A Functional Integrated Data Evaluation System for Safeguards

The quantitative assurance provided by the safeguards of nuclear material is based on the analysis of a very large amount of data from different origins and quality. They are generated by operators on a regular basis, or provided during discussions of facility attachments or declaration of production schedules. They are also generated by inspectors during their verification activities which are related to accountancy data, auditing, independent measurements, sealing, surveillance, etc.

The Joint Research Centre and the Safeguards Directorate of the Commission of the European Communities are studying a Functional Integrated Data Evaluation System (FIDES) for safeguards. A preliminary outline of such a system was presented at the 3rd ESARDA Symposium at Karlsruhe.

The scope of this paper has been firstly to emphasise the decisional thread which underlies the activities and secondly, to give a progress report on JRC work which is designed to give effect to these ideas. The progress reported covers two activities. The first is the automatic coordination of the operator's measurement system information with the operator's accounting declaration. The second element is the development of a functional structure for NDA data generation evaluation and transmission.

SM 260/66

A. MacDonald, A.H. Kerr, D.J. Savage (UKAEA, Dounreay)

Operational Experience with the Ceric Oxidation, Ferrous Reduction and Dichromate Titration Method for Plutonium in Fast Reactor Fuel Reprocessing

A new titrimetric procedure was described at the 1978 IAEA nuclear safeguards technology conference in which plutonium is oxidised to plutonium VI by cerium IV in nitric acid solution, the excess oxidant is destroyed in a series of redox reactions, and the plutonium VI is reduced by a measured excess of iron II which is then back titrated with potassium dichromate. This paper describes the further development of this method, and the operational experience gained in routine use.

The absence of bias in the basic procedure was demonstrated using NBS plutonium metal standard. Iron, chromium manganese, zinc, molybdenum, uranium, americium, and mixed fission products are shown not to interfere at the levels expected in nitric acid solutions of irradiated fast breeder fuel. Both vanadium and neptunium interfere quantitatively. Examination of the reaction between plutonium VI and the arsenic III used to reduce excess cerium IV suggests that it could be a source of bias, but detailed examination of the recommended procedure shows there is no problem. Temperature variations between 15°C and 30°C are shown to be tolerable. Routine operational experience for accountancy and safeguards purposes has been very good for both input and product streams.

SM 260/68

W.B. Bremner, M.L. Clark, B.W. Spence (UKAEA, Dounreay)

Operational Experience Relating to Measurement of Plutonium in Solid and Liquid Waste Streams from Fast Breeder Reactor Fuel Reprocessing

The reprocessing of fuel at the Dounreay Nuclear Power Development Establishment (DNPDE) from the Prototype Fast Reactor has been routinely carried out for about two years and has been reported elsewhere. These reprocessing operations have been subject to continuous safeguards both by Euratom and IAEA. A primary safeguards feature of the reprocessing plant is by the use of nuclear material accountancy and this paper deals with techniques used for the measurement of plutonium in solid and liquid waste streams during two reprocessing campaigns.

The solid waste streams have been designed to allow eleven non-destructive analysis (NDA) systems to be used in order to make plutonium measurements for plant control, accountancy and safeguards thus also ensuring that the correct waste disposal route is selected. The major problems associated with these NDA measurements have been in the head-end area of the reprocessing plant, as a result of the high associated γ-activity present and of the neutron emission from ingrown curium isotopes.

The liquid waste stream measurements are mainly achieved by tank sampling followed by wet chemical spectro-photometric or radiometric analysis in support laboratories. In the case of highly active raffinates analyses are carried out in a suite of shielded $\alpha\beta\gamma$-facilities using remote handling techniques. The medium and low active streams are analysed by similar techniques but the sample handling problems are much less severe.

The quantities of plutonium which were measured during the first two campaigns are given for the various waste streams. In the design of the accountancy system it was considered that chemical cross checks of the NDA measurements should be made. Results are given of cross-check analysis of leached hulls which compares the NDA values to destructive analyses. A quality control system is used to continuously evaluate and update the performance of the wet chemical colorimetric methods.

A co-operative programme with Euratom to investigate the optimisation of solid waste measurement systems and the comparison of theoretical studies with practical measurements, has been in progress for over two years and will shortly be concluded. Some data on certain aspects of this NDA evaluation work is presented.

There is a continuing requirement for improvement in these measurement techniques. In the case of solid wastes the main development effort is towards replacement of sealed tube neutron generators, for neutron interrogation of head-end wastes, by a 252-californium system or by 144-praseodymium gamma measurements. For liquid raffinate streams the emphasis is on carrying out analysis by on-plant equipment. Evaluation of the differential die-away technique for analysis of highly active raffinates is being carried out.

A high availability of NDA system coupled with a quick analysis of raffinates is an essential requirement for plant safeguards. The performance of the system in use at DNPDE will be considered from this view point.

SM 260/83

H.P. Filss (KFA, Jülich)

Non-Destructive Assay of Fresh and Irradiated Fissile Material in Waste Barrels by Selective Neutron Transport Techniques

Various fissile material configurations are handled in a research centre such as KFA, Jülich. Large containers are an important sample type, mainly barrels containing waste-type materials. This NDA system is intended for this type of samples, containing mixtures of fissile, radioactive and matrix material. It is an active neutron assay system suitable for irradiated material as well and operable in a hot cell. Its development is based on previous work with a system for small and medium-sized samples which has also been tested with single HTR fuel elements (fresh and irradiated) and Pu-bearing samples. In the present system, the sample to be interrogated, which may be a waste barrel, is placed in the middle of a rotating table. The Sb-Be-neutron source is on one side and the selective neutron counter on the other side of the sample. The counter is specially designed to register preferably fast prompt fission neutrons as the fissile material signal. According to a test measurement with unirradiated rod pieces in a waste barrel, the background is equivalent to approximately 2 g ^{235}U in this NDA system. The dependence on different sites in the barrel is ±20% at present. Not only are the neutron count rates greatly reduced in the case of a concreted barrel, but the fissile material rates and the background rates are reduced as well. The relation of background/fissile material signal remains practically unchanged. This result leads to the expectation that even concreted barrels can be directly measured by this system if they contain more than 2 g of ^{235}U. For accurate measurement additional work on calibration seems to be necessary, but in any case, the system offers the possibility of a direct control measurement in large final containers.

SM 260/86

C. Beets (CEN/SCK Mol), P. Boermans (FBFC, Dessel), H. De Canck, R. Ingels (Belgonucléaire, Dessel), J. Satinet (Tihange Power Plant)

Belgian Experience in Safeguards Implementation and Related R&D Activities

As a Non-Nuclear-Weapon State Party to the Non-Proliferation Treaty, Belgium considers the Agency's safeguards system as one of the basic components of the Treaty. Therefore, in our particular case, the effectiveness of the Agency's safeguards system as a result of the co-operation between the Agency, Euratom and the State has to be clearly stated and demonstrated.

This paper will follow this idea as a guideline, considering the implementation of international safeguards in our main representative nuclear facilities.

It should be borne in mind that nuclear energy contributes a significant part of the production of electrical energy (about 25% in the year 1980) so that the impact of safeguards on the Belgian nuclear fuel cycle constitutes an important information related to the Agency's safeguards system.

This impact has led Belgium to propose a Research Support Programme to the Agency, the three parties involved being IAEA, Euratom and Belgium. The first two studies in this programme concern :

- the optimization of safeguards measures in mixed oxide fabrication plants,
- the impact of improvement in verification capabilities on the safeguards approach in LEU fabrication plants.

SM 260/102

A.S. Goldman (Los Alamos), R. Beedgen (KfK, Karlsruhe)

The Use of (D,MUF) and Maximum Likelihood Methods for Detecting Falsification and Diversion in Data Verification Problems

The investigation of data falsification and/or diversion is of major concern in nuclear materials accounting procedures used in international safeguards. In this paper, two procedures denoted by (D,MUF) and LR (Likelihood Ration) are discussed and compared when testing the hypothesis that neither diversion nor falsification has taken place versus the one-sided alternative that at least one of these parameters is positive. Critical regions and detection probabilities are given for both tests. It is shown that the LR method outperforms (D,MUF) when diversion and falsification take place.

SM 260/108

C. Beets (CEN/SCK Mol), M. De Carolis, A. Keddar (IAEA, Vienna), H.O. Menlove (Los Alamos)

Links between Different Ultrasonic Techniques Proposed around the World for Items Unique Identification

In the application of NDA techniques for safeguards verification it is important to realize that the transition from laboratory development to routine use requires fully documented field tests in order to define practicable NDA standards accepted both by the operator and by the Agency.

The CEN/SCK with the nuclear facilities installed nearby, namely oxide (Franco-Belge de Fabrication de Combustible, FBFC, Dessel) and mixed oxide (Belgonucléaire) fabrication plants constitutes a unique site where the IAEA has the opportunities to test instruments and methods in realistic field conditions.

The objective of the IAEA contract No. 2274 R/B, initiated in 1980, concerns the optimization of NDA measurements in field conditions for safeguards purposes. It foresees the following programme of measurements :

- Determination of ^{240}Pu equivalent of Pu in bulk by passive neutron coincidence using the High Level Neutron Coincidence Counter (HLNCC),
- Determination of Pu isotopic ratios of Pu in bulk by transmission gamma spectrometry using intrinsic germanium detector and portable SILENA MCA,
- Verification of ^{235}U and ^{236}U contents in PWR fuel assemblies by fission neutron counting in active and passive modes using the neutron collar technique,
- Pu total verification by calorimetry and gamma spectrometry for small samples (1st type calorimeter), for bulk samples up to 2 kg PuO_2 (2nd type calorimeter) and fast reactor rods (3rd type calorimeter).

The varioius tests effected, or to be effected, in the programme combine representative items encountered in flow and inventory verification and relevant NDA techniques.

Realistic error estimates have to be deduced from each of these tests, taking into account the error contributions due to the significant parameters in the applied technique. This is not an easy task. In order to design appropriate experiments and to get the best analysis of the data, the instrument developer, the operator and the inspectors have been closely associated in each experiment. Furthermore, to permit further analyses, raw data are published in the various progress reports.

Representative (and costly) items, such as medium and high burn-up PuO_2 cans, being available, part of the programme has been extended to some of the ESARDA Working Group partners (UKAEA, KfK, ECN) and L.A.S.L. (U.S.A.) in order to have an inter-comparison of the techniques in field conditions.

It is hoped that this programme will contribute to fulfill a part of the matrix giving the set of error values typical for the tested combinations item - NDA. This matrix will notably provide for a quantitative basis for establishing powerful sampling plans and inspection strategy.

SM 260/109

S. Crutzen, P. Jehenson (CEC, Ispra), J. Mc Kenzie, I. Waddoups (Sandia Nat. Lab., Albuquerque)

Links between Different Ultrasonic Techniques Proposed around the World for Items Unique Identification

Over the last two years, several research establishments involved in R&D for International

Safeguards have been investigating the possibility of using the "Ultrasonic Signature" principle to identify uniquely items such as containers and fuel elements or bundles.

The ways in which general principles are applied, always appear to be very different.

A careful systems analysis shows that the solution chosen can be inserted in a general scheme resulting in three major approaches involving only two different families of electronic devices for the identity, pick-up, and treatment.

SM 260/124

A.P. Knight (UKAEA, Winfrith)

Rapid Inventory Taking by Electronic Data Gathering

A Plessey bar-code reading system has been installed in a nuclear material store at AEE Winfrith and is used to identify discrete packages of various nuclear materials. A mini-computer is utilized as a data store which, coupled with the Plessey bar-code system, produces physical inventory listings or inventory discrepancy reports very quickly. The time taken for a complete inventory of the store has been reduced from 60 man-days to 5 man-days.

MS 260/135

D.J. Pike, A.J. Woods (Univ. of Reading)

Statistical methods in Nuclear Materials Accountancy : Past, Present and Future

The analysis of nuclear material inventory data is motivated by the desire to detect any loss or diversion of nuclear material, insofar as such detection may be feasible by statistical analysis of repeated inventory and throughput measurements. The early regulations, which laid down the specifications for the analysis of inventory data, was framed without acknowledging the essentially sequential nature of the data. It is the broad aim of this paper to discuss the historical nature of statistical analysis of inventory data including an evaluation of why statistical methods should be required at all. If it is accpeted that statistical techniques are required, then two main areas require extensive discussion. First, it is important to assess the extent to which stated safeguards aims can be met in practice. Second, there is a vital need for re-assessment of the statistical techniques which have been proposed for use in nuclear material accountancy. Part of this reassessment must involve a reconciliation of the **apparent** differences in philosopy shown by statisticians; but, in addition, the techniques themselves need comparative study to see to what extent they are capable of meeting realistic safeguards aims. This paper will contain a brief review of techniques with an attempt to compare and contrast the approaches. It will be suggested that much current research is following closely similar lines, and that national and international bodies should encourage collaborative research and practical in-plant implementation. The techniques proposed require credibility and power; but at this point in time statisticians require credibility and a greater level of unanimity in their approach. A way ahead is proposed based on a clear specification of realistic safeguards aims, and a development of a unified statistical approach with encouragement for the performance of joint research.

SM 260/138

R. Abedin-Zadeh, T. Beetle, E. Kuhn, D. Terrey, S. Turel (IAEA, Vienna), G. Busca (CEC, Luxembourg), S. Guardini (CEC, Ispra)

Preparation of Plant Specific NDA Reference Materials

The importance of having suitable and well characterized Non-Destructive Assay (NDA) reference materials for the verification activities of the safeguards control authorities is stressed. The Euratom Inspectorate and the IAEA have initiated an extensive programme for the procurement and preparation of Joint Euratom/IAEA safeguards NDA reference materials with the active participation of the Ispra Establishment of the CEC Joint Research Centre. The different type and nature of materials, condition of measurements, plant characteristics and provisions required specific considerations for plant specific NDA reference materials.

The preparation of each reference material was planned case by case and specific criteria such as limitations in different facilities, measurement capabilities, conditions, product availability and population variability are ascertained. A procurement scheme was prepared describing step by step procedures detailing responsibilities, measurement conditions, destructive analysis schemes, desired characteristics and methods of data evaluation.

This paper describes the principles and procedures carried out for the preparation of a reference MOX pin, low enriched uranium reference rods, low enriched uranium reference drums, reference MTR assemblies and THTR reference pebbles. The scheme for each characterization technique is presented.

SM 260/145

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Near-Real-Time Materials Accountancy. A Technical Status Report

Near-real-time materials accountancy as applied to reprocessing facilities involves two major elements, the measurement or estimation of the in-process physical inventory at frequent intervals and the statistical evaluation of the resulting sequential material balance data. For most reprocessing facilities the bulk of the in-process inventory normally is in intermediate "buffer" tanks, and is directly measurable with little or no added effort. In contrast, the plutonium inventory in the solvent extraction system does not appear to be directly measurable. Variations in this inventory, however, could cause a reduction in the sensitivity of the sequential data analysis. Although some studies are in progress, to date an acceptable means for accounting for these variations has not been found. Statistical tests for evaluating sequential material balance data are still being studied at several laboratories, but consultants at a meeting in January 1982 agreed that under all circumstances there is some increase in both detection timeliness and detection sensitivity using n.r.t. accountancy. IAEA verification of operator-generated measurement data is an area requiring significantly increased effort. It is hoped that a major demonstration over a period close to a full year can be arranged starting in 1983.

Cooperative Development of Safeguards within the European Safeguards Research and Development Association (ESARDA)

IAEA SM 260/69

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Paper presented at the International Symposium on Recent Advances in Nuclear Materials Safeguards, Vienna, Austria, 8-12 November 1982.
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Abstract

After a listing of the partners in the ESARDA organization attention is given to the various working groups, which are active in the field of safeguards research and development.

Recently some working groups have been established or reorganized with a view to better orientation to the actual problems existing in todays nuclear facilities.

In these working groups representatives from research organizations, safeguards authorities and plant operators are participating to solve the most urgent problems.

The objectives and activities of the working groups are summarized in appendices.

The priorities for the various tasks are set by the ESARDA Steering Committee, in which, since 1981, plant operators are also represented. Information exchange between interested persons occurs not only at the working group level, but also at the annual ESARDA symposia.

The European Safeguards Research and Development Association (better known as ESARDA) provides an international forum for the exchange of information and ideas between nuclear facility operators and safeguarding authorities.

Its contract partners are at present:

- the European Atomic Energy Community (Euratom, since 1969);
- the Kernforschungszentrum Karlsruhe, F.R. Germany (KfK, since 1969);
- the Centre d'Etude de l'Energie Nucléaire /Studiecentrum voor Kernenergie, Belgium (CEN/SCK, since 1970);
- the Comitato Nazionale per la Ricerca e per lo Sviluppo dell'Energia Nucleare e delle Energie Alternative, Italy (ENEA, since 1971);
- the Stichting Energieonderzoek Centrum Nederland, the Netherlands (ECN, since 1971);
- the United Kingdom Atomic Energy Authority, Great Britain (UKAEA, since 1973);
- the Energistyrelsen, Denmark (ENS, since 1973);
- the Commissariat à l'Energie Atomique, France (CEA, since 1981);
- British Nuclear Fuels Ltd., Great Britain (BNFL, very soon).

ESARDA has one of the largest coordinated safeguards R&D programmes in the world. The history of the first 10 years of ESARDA has well been described by D. Gupta in 1979 at the first annual ESARDA Symposium at Brussels[1]. Originally ESARDA was constituted as a group of research establishments (in principle one from each member state of the European Community), plus the Joint Research Centre, which were concerned with advancement and harmonization of research and development work for safeguards. Its main objective was to secure voluntary exchange of information on programmes, avoidance of unnecessary duplication, and a joint approach in identifying future work. This constitution was broadened by a change in the Contract of Association in 1981, described later. The Steering Committee discusses and reviews regularly a comprehensive document with a systematic presentation of the efforts by all partners. The discussion leads to a setting of priorities and to a harmonization and optimization of the R&D work.

The actual R&D work of ESARDA is performed mainly within the structure of working groups which were originally devoted to particular techniques. Active working groups of this type are :

- WG for non-destructive analysis (NDA)[2],
- WG for destructive analysis (DA)[3],
- WG for containment and surveilance (C/S)[4,5],
- and more recently, the WG on mathematical/statistical techniques (M/S).

Their objectives and present main activities are described in detail in the appendices 1, 2, 3 and 4.

These Working Groups attracted participants and observers from outside the member research establishments.

From 1 January 1981, ESARDA has been so constituted that participation and representation in the Steering Committee has been open to any organization having substantial interest in the application of safeguards. Thus nuclear plant operators now have direct representation in the Steering Committee, which has the highest decision level. This new legal situation reinforces the tendency which started some years ago, to take advantage of the cooperation of the plant operators. They have been recognizing the fact that an effective and efficient application of safeguards measures can be harmonized with measures which favour economic and cost-effective plant operation.

As a consequence of this new orientation of ESARDA, important Working Groups have been established, which have a clear plant specific character[7]. These working groups bring together plant operators, research scientists, and safeguarding authorities, to discuss, and, as far as possible, to cooperate in the solution of problems which arise when safeguards measures are applied in actual plants.

The following "plant specific" working groups have been formed :

- the WG for Low Enriched Uranium fuel fabrication plant (LEU)[8],
- the WG for Mixed Oxide Fuel fabrication plant (MOX),
- the WG for Reprocessing Input Verification (RIV)[9-11].

The Objectives and present main activities are described in some detail in the appendices 5, 6 and 7.

With respect to the oldest plant specific Working Group (that on LEU) it is worthwhile mentioning that in addition to research workers, and the Euratom Safeguards Inspectorate representatives of some nine European fuel fabricators are also participating.

This Working Group discussed problems essentially from the point of view of establishing cost-effective methods of safeguards application in operating plants, and of avoiding time consuming reconciliation of differences between measurement capabilities at different plants. A study is being made of accountancy problems and verification techniques. A cooperative effort was undertaken on the intercomparison of weighing, intercomparison of analytical performance using reference uranium oxide pellets, the use of rod scanners for nuclear material control, and the application of special computerized accountancy systems, also applicable to Near Real Time Accountancy, and their interface with safeguards needs.

This work is currently continuing, and results will be reported when they become available.

The good spirit of cooperation, experienced in the LEU Working Group was reason for the Steering Committee to establish the MOX Working Group. This working group has the support of all European MOX fuel fabricators. It will extend the recognized value of the efforts of the LEU group into the more specialized field of mixed oxide fuel fabrication. At present work was started on the exchange of uniformation on possible nuclear material management procedures, on measurements techniques, and on procedures for Physical Inventory Taking, which are of use for safeguards application. This Working Group is also analysing in detail the plant specific safeguards approaches.

Some specialized problems with a more mathematical/statistical background, raised by the LEU Working Group and which will probably be also of importance in the MOX field, have been separated from the plant specific working group. These clearly needed the cooperation of mathematicians and statisticians for their solution, and the Steering Committee decided to establish a special working group for these mathematical/statistical techniques with participation of experts on this field.

This Working Group will mainly act upon request from the plant specific working groups : thus ensuring that its efforts are applied to practical rather than theoretical problems.

Quite recently the old Working Group on Isotopic Correlation Techniques (ICT) has been reoriented by the ESARDA Steering Committee to consider techniques applicable to the verification of input to reprocessing plants, including the calibration of the input accountability tank, ICT would thus become one of several techniques to be studied in a comparative manner. It is expected that this will prove an attractive forum for reprocessing plant operators to cooperate in resolving these difficult problems in an effective and practical approach. Furthermore, considerable effort was spent to integral testing of ICT in the Isotope Correlation Experiment (ICE)[11]. This experiment will have a follow-up in the Reprocessing Input Verification Experiment (RIVE), which is now being planned.

The Working Group on Containment and Surveillance is not purely concerned with techniques.

Although nominally more a technique-orientated than a plant orientated working group, it is attracting considerable and valuable support from plant operators. These operators are concerned in evaluating the operational and/or economic advantages which might be gained by these measures, which may be used to supplement or partly to substitute for the traditional Nuclear Material Accountancy approach.

This change in emphasis from the original research-oriented working groups to more practical and plant-specific working groups, has not been without problems for the organization of ESARDA, but the success obtained up till now is an encouragement to continue in this direction, and to find effective ways of overcoming these organizational difficulties. Rules have to be developed or improved for the creation of appropriate working groups, for their suspension or termination after finishing the defined jobs and for reporting the results.

With respect to the publication of results and dissemination of the experience obtained, ESARDA has already published (see the attached list) a series of technical reports, and has organized, with great success, a series of annual meetings (symposia and specialist's meetings). These meetings are organized around a theme of current safeguards interest and constitute an additional forum for information exchange. They have been well attended also from outside the European Community. Contacts were established with the R&D work in the U.S.A. through the channels of the Institute of Nuclear Materials Management (INMM). ESARDA had twice the opportunity to present its activities in special session of two annual INMM meetings (Albuquerque, 1979, and Palm Beach, 1980). Reciprocally INMM representatives participated in the series of ESARDA symposia in Europe.

The 1982 specialist meeting, held in Petten, was devoted to the theme "Harmonization and Standardization in the Field of Safeguards". The 1983 symposium, which will be held at Versailles, has a main theme "The Interaction between Safeguards Inspectors and Plant Operators in Safeguards".

In conclusion, one may note that the ESARDA organization has set-up a unique cooperative structure for improving the practical application of safeguards, which draws upon the expertise of all interested parties (research collaborators, safeguards inspectors and plant operators).

Such a structure should increasingly facilitate the strengthening of the safeguards system with minimum intrusions into the productive processes of the nuclear industry.

Acknowledgement

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Appendix 1

Terms of reference for the ESARDA Working Group on Techniques and Standards for Non-Destructive Analysis

Objective

To evaluate and recommend criteria for non-destructive analysis of nuclear materials for use by plant operators and the safeguarding authorities.

Present activities

1. Intercomparison of gamma and neutron measurements on cans of plutonium-oxide, containing either 0.5 or 3 kg Pu (in real plant conditions)
2. Plutonium Isotopic Determination Intercomparison Exercise (PIDIE), using 0.5 g Pu samples
3. Preparation of uranium-oxide enrichment standards for gamma spectrometric determination of enrichment
4. Feasibility study of NDA measurement on finished fuel assemblies
5. Updating of the list of reference materials for non-destructive assay of U, Th and Pu isotopes (report ESARDA-5, EUR 6089, 1978)
6. Updating of the report on established application of non-destructive techniques for nuclear materials measurements control or verification (report ESARDA-6, AERE-R-9167, August 1978).

Appendix 2

Terms of reference for the ESARDA Working Group on Techniques and Standards for Destructive Analysis

Objective

To evaluate and recommend criteria for destructive analysis of nuclear materials for use by plant operators and safeguarding authorities

Present activities

1. Publication of the 1979 list of target values for measurement uncertainties for various destructive analysis methods
2. Preparation of a clarifying document on significant systematic differences between declared and verified values
3. Establishment of a qualified list of destructive analysis methods with well-known performances for consideration as international standards
4. Preparation of an updated list of reference materials
5. Participation (and if necessary, organization) of regular inter-laboratory measurement programmes to determine the reliability of measurements with the least possible effort on the part of participating and organizing laboratories.

Appendix 3

Terms of reference for the ESARDA Working Group on Containment and Surveillance Techniques

Objective

To evaluate and recommend procedures for containment and surveillance methods for use by safeguarding authorities to reduce the inspection effort at nuclear facilities.

Present activities

1. Preparation of a joint U.S.A./ESARDA compendium on C/S techniques
2. Collect information on the following C/S devices:
   - general purpose seals
   - doorway monitors
   - cap seals
   - electronic seals
3. Collect information on the sensitivity function of these devices.

Appendix 4

Terms of reference for the ESARDA Working Group on Mathematical-Statistical Problems

Objective

Study of the problem of mathematical-statistical nature which are important for the application and implementation of safeguards and nuclear material accountancy.

Present activities

1. Preparation of statistical guidelines for scale calibration, to analyse the outcome of the project of establishing performance data (variances and biases) of weighing scales under plant operation conditions, by means of measurements on reference masses, prepared and circulated by JRC.
2. Perform a modelling of process flows and measurement system in a mixed oxide fuel fabrication plant, to obtain a mix of dynamic verification and complete inventory verification, which meet the inspection requirements in terms of detection time and goal quantity.
3. Study the statistical approach for performing automatic (i.e. computerized) control of analog recordings of identity patterns obtained by ultrasonic testing of seals (with random conclusions).
   Two approaches have been mentioned, the Ispra apporach, investigating the characteristics of the patterns (peak heights and peak positions), and the Sandia approach, investigating the correlation between two plots, represented by a series of equidistant points.
4. Perform a critical review of existing techniques, and make a recommendation for the following topics encountered in interlaboratory intercomparison of results of measurements on various characteristics of a series of items:
   - procedure for detecting outliers, not only for the univariate case, but also for the multivariate case,
   - procedure for deriving estimates for the individual measurement variances for the laboratories involved,
   - procedure for deriving estimates for the individual measurement biases for the laboratories involved.
5. Perform a study of the various procedures of tank calibrations in reprocessing plants, in order to arrive at practical recommendations.

6. perform a study on the relation between verification effort and goal quantity in a Low Enriched Uranium fuel fabrication plant. The verification effort has here to be related to the number of statistical samples, the accuracy of NDA measurements on unsealed items present, and the required measurement time.

Appendix 5

Terms of reference for the ESARDA Working Group on LEU Conversion/Fabrication Plants

Objective

1. To identify problems encountered both by plant operators and safeguards authorities while implementing international safeguards regulations
2. To propose acceptable solutions to the parties concerned which should optimize the effort in man-power and hardware with the benefit of a higher efficiency and credibility of the actual control.

Present activities

1. Continuation of the study and verification scheme for a reference LEU plant, based on a fast response computerized accountancy system (dynamic inventory verification), together with statistically based sample verification.
2. Continuation of the parallel study of a verification scheme for the same reference plant, based on the verification of input and output, and on the verification of the physical inventory once per year.

Appendix 6

Terms of reference for the ESARDA Working Group on Mixed Oxide FuelFabrication Plant Safeguards

Objective

Study of the problems related to the implementation of safeguards measures in mixed oxide fuel fabrication plants.

Present activities

1. Investigation of Near Real Time Accountancy capabilities
2. Study of the implications of a biweekly inspection strategy considering partial inventories, in combination with a total Physical Inventory Taking at the end of the normal material balance period.
3. Critical analysis of the IAEA document AG-244 (''Some safeguards considerations for a reference mixed oxide fuel element fabrication plant with an annual throughput of 500 kg PuO_2'') including new concepts and extrapolations to a reference plant with a throughput larger than 1 tonne PuO_2/year.

Appendix 7

Terms of reference for the ESARDA Working Group on Reprocessing Input Verification

Objective

Analysis of the measurement/verification system at the input of reprocessing plants for LWR fuel, in view of making safeguards procedures more effective and efficient.

Present activities

1. To carry out an exercise of application of the various existing procedures for Isotope Correlation Techniques (ICT), in order to demonstrate their sensitivity in detecting anomalous data
2. To lay-down detailed procedures for practical use of ICT
3. To evaluate procedures for sampling and for determining the head-end losses
4. To update the ESARDA data bank of isotopic compositions in irradiated fuels and to critically evaluate its contents.

Development and Testing of Systems and Techniques for Safeguards of Spent Fuel Reprocessing Facilities

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Introduction

ENEA is at present engaged in a comprehensive study of safeguard systems and techniques to be applied in fuel reprocessing facilities both for process control and for International Safeguards purposes.

Designers and operators of reprocessing plants are acquainted and actively involved with the progress, development and utilization of the measuring systems and techniques.

The aims are the following :

- Testing and demonstration of already developed instrumental techniques in operating reprocessing facilities (i.e. EUREX - Saluggia (North Italy), ITREC - Trisaia (South Italy)).
- Optimization of the installed instrumentation in order to improve performances for Safeguards and Accountability applications (EUREX - Saluggia, ITREC - Trisaia).
- Research and Development for new techniques (Research Lab. - Casaccia - Rome).
- Development of automation for data acquisition, handling and treatment as a very useful feature for process/safeguards controls implementation (EUREX - ITREC - Research Lab.).

Some of the work is being carried out in the form of cooperation programmes with the IAEA, EEC and the University of Rome.

At present the areas under investigation at ENEA are :

- Input Accountability (ITREC plant)
- Hulls Monitoring (EUREX and ITREC plants)
- Process Control and Intermediate Product Measurements (EUREX, ITREC)
- Product Accountability (EUREX)
- Liquid Wastes (EUREX)
- Solid Wastes (EUREX).

In particular the following systems and techniques are under development testing and demonstration :

- TDR technique (1) for input/output accountability volume measurement
- Gamma-ray spectrometry and weight technique (load cells) for N.M. losses detection in the leached hulls (1)
- X-ray absorptiometry (MAX-1) (1); dual energy X-ray absorptiometry (DEXA) (2); gamma ray spectrometry for product characterization and accountability
- X-ray and alpha monitoring systems for Pu loss detection in liquid waste streams (3)
- Gamma spectrometry for Pu loss detection in solid wastes (4).

Some more details related to this comprehensive study, are presented in the following.

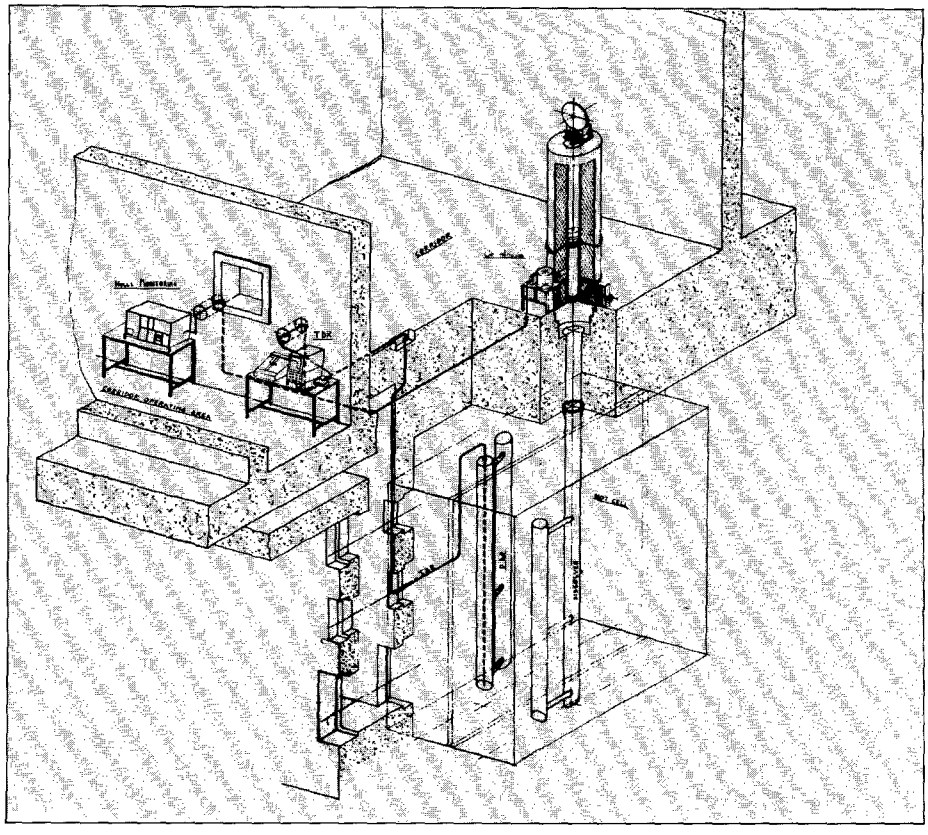


Fig. 1 - ITREC Plant: Layout of the TDR and of the hulls monitoring system installation

Input Accountability

Volume measurement

A TDR* apparatus has been built, tested and compared with the DIP tube manometric system, under various and extreme experimental conditions, in the input accountability vessel of the ITREC plant (Fig. 1). Four years of testing** have allowed a great deal of experimental data to be collected and a complete evaluation of the performance of the equipment to be made. Although quite satisfactory results have been obtained, a set of recommendations has been forwarded to the IAEA for further improvements of the design (5).

Hull Monitoring

Gamma Spectrometry

Two experimental set up have been designed, built and tested : one in EUREX (Fig. 2) and one in the ITREC plant. Both methods are based upon a gamma measurement of the basket containing the leached hulls. The main difference between the two methods is the choice of the gamma ray detectors. In EUREX an NaI crystal was used while in ITREC an advanced Ge(Li) coaxial detector was used. Both systems have performed extremely well. Losses lower than 0.1% can be detected easily.

Weighing Technique

To relate the N.M. losses in the leached hulls, EUREX has tested a weighing system as an alternative to gamma spectrometry (Fig. 2). A very simple method is applied, which is based on the weight of the leached hulls and the successive comparison of the value obtained with the weight of the canning material made available by the supplier.

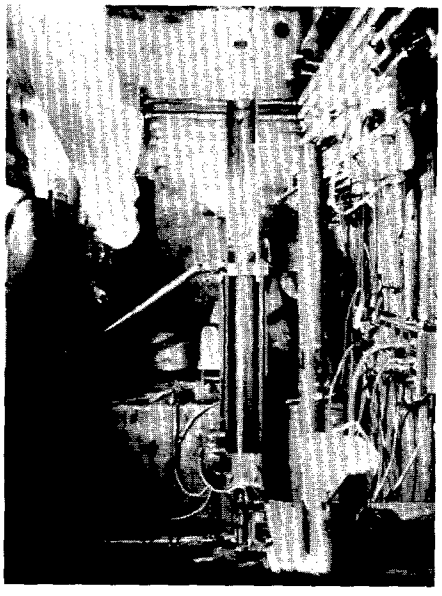


Fig. 2 - EUREX Plant: Leached hulls weighing and gamma monitoring

The measuring system is affected by an uncertainty : the liquid remaining in the hull which cannot be evaluated a priori. On the basis of the data obtained after a series of 15 weighings the amount of liquid remaining in the hulls is estimated to be about 300 grams.

By applying operating procedures which can dry the hulls (for example, by heating the basket inside the dissolver), the technique can be improved. The technique should be considered, after more promising results, because of its quick operating procedure and its low cost.

Intermediate and Product Measurements

Output Accountability Volume Measurements

A system similar to the TDR applied for the input accountability vessel of the ITREC plant has been designed, built and tested for the plutonium final product accountability vessel of the EUREX plant. Previous experience gained with the ITREC equipment has allowed us to make substantial improvements in the design and fabrication of this equipment. Furthermore a much better operating procedure and data acquisition (Fig. 3) and evaluation was made possible during the Candu campaigns.

* Patented

**This work has been carried out as part of a cooperation programme with the IAEA.

U and Pu Concentration Measurements

Two NDA techniques have been applied for N.M. concentration measurements : the X-ray densitometer for the measurement of the total absorption of heavy metals (MAX-1) and the Dual Energy X-ray absorptiometer (DEXA).

The MAX-1 is a non-specific system, since it measures the total absorption of heavy metal in solution, while DEXA is a specific one, since it enables simultaneous measurement of two elements in a mixed actinide solution.

The two sets of equipment were designed*, built and tested, the first one in the EUREX plant (Fig. 4) and the second one in the ITREC plant. Further development is under way for the second set in view of its possible application for safeguards purposes. The performance of MAX-1 has been very good. For the Candu campaign the NaI crystal was, however, replaced by a gas-filled proportional counter, in order to overcome the interference due to the spontaneous emission of Pu.

The results as regards the DEXA obtained during a Thorex campaign where a centrifugal contactor was used, in addition to providing good agreement with the analytical data, showed the ability of the method to timely detect transient phenomena.

High Resolution Gamma Spectrometry

ENEA has great interest in developing a routine method of Pu isotopic composition measurements with gamma ray spectrometry. A set of gamma ray spectra of plutonium samples with known isotopic composition has been taken and evaluated with a general computer program for gamma spectrometry. Furthermore measurements have been performed on a Candu Pu sample, whose isotopic composition is known by mass spectrometry, which was specially prepared and made available by the EUREX plant.

On this set of gamma ray spectra a mathematical evaluation is under way using different methods.

The future development of this research programme envisages the following steps :

- to acquire Pu isotopic standard accepted and/or certified by international authorities (IAEA, Euratom, NBS)
- to optimize both gamma spectrometry measurements and mathematical methods for their evaluation with the aim of obtaining a level of accuracy close to that of mass spectrometry.

Pu and U Measurements in Liquid Waste

The following systems are being tested at ENEA :

a. Pu X-ray monitoring system
b. Alpha particle monitoring system using a silicon surface barrier detector of large area (600 mm²)
c. OLAM system, based on the detection of alpha particles by cerium activated glass scintillators.

* Patented

Fig. 3 - EUREX Plant: Block diagram of the microprocessor system for the TDR automatization

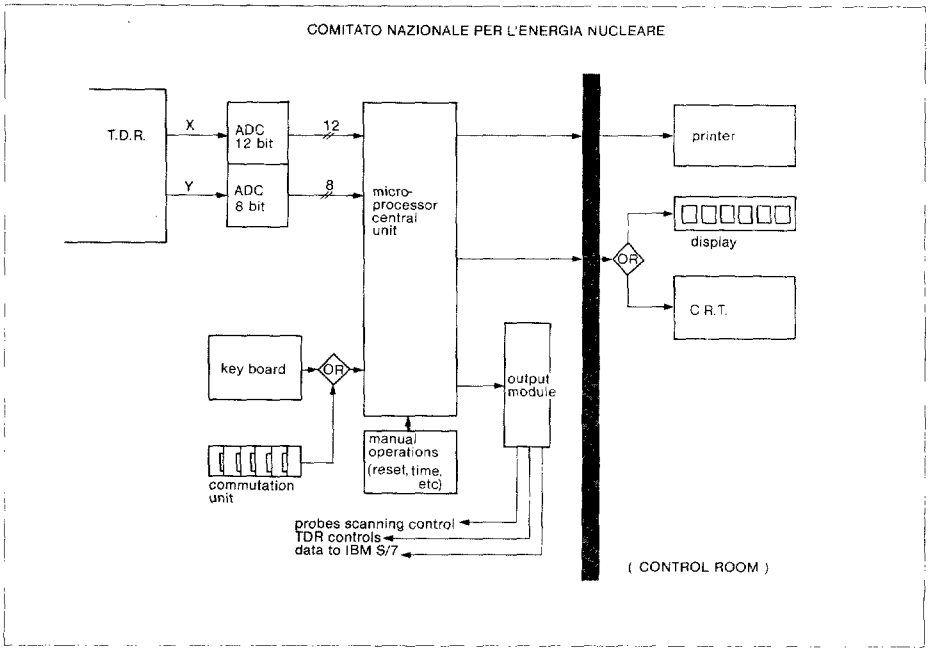
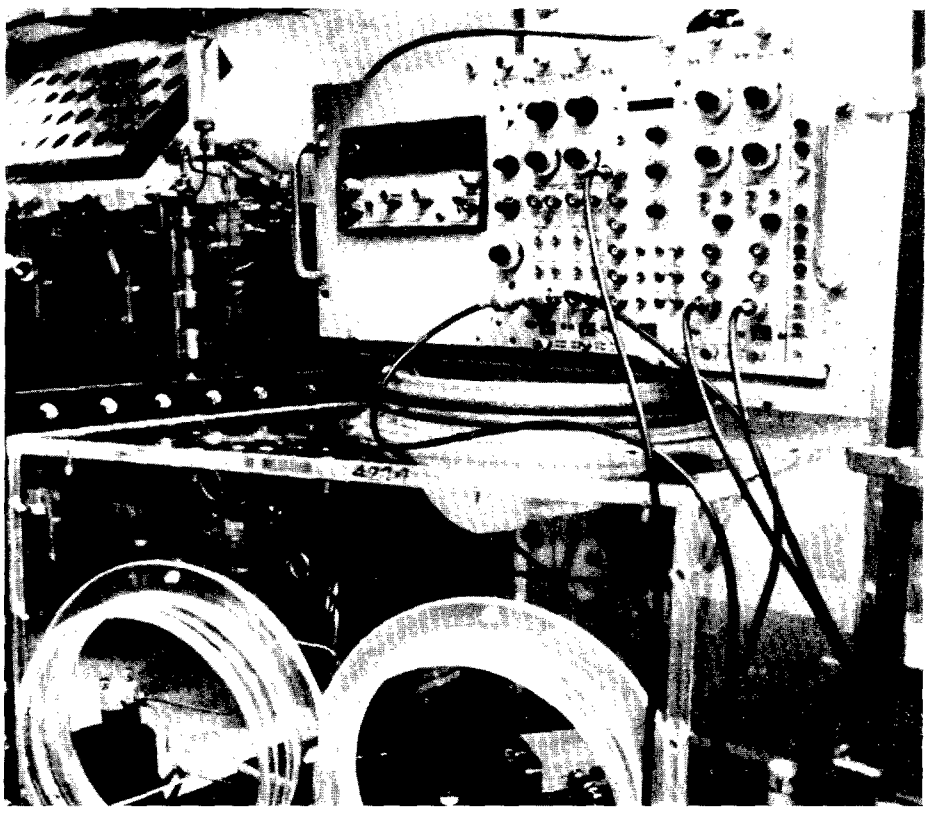


Fig. 4 - EUREX Plant: Max-1 absorptiometer installed on the second extraction cycle product stream



The choice between the three systems is made depending on the stream to be controlled, the location of the plant in which the control is carried out and the environmental conditions.

The units as in items a. and b. have been designed by CNEN while OLAM is an IRT equipment supplied by the JRC-Ispra following a CNEN-EUREX/Euratom agreement to test it in hot conditions.

The X-ray monitoring system uses a shielded X-ray NaI (Tl) scintillation detector installed on a stainless steel flow-cell with inlet and outlet positioned to give homogeneity and full liquid exchange in the cell.

The sensitivity during calibration runs performed using Ba-133 to simulate Pu was found to be 2.10^{-3} Ci/l for Pu-238 and 2.5 mg/l for Pu-239 respectively.

The unit has been installed on the EUREX plant to control leakage on the steam condensate stream of the Pu final product evaporator (Fig. 5). It was in operation during the Candu reprocessing campaigns

The semiconductor alpha-particle detector system is being evaluated at the Casaccia research laboratory.

At present the Pu sensitivity measured is 0.1 mg/l of Pu-239. The sample cell of small volume has been designed to have a thin layer (2 mm) of liquid facing the detector in order to minimize the beta radiation interference.

OLAM (on-line alpha monitor)

This consists of an on-line cesium activated glass alpha detection system designed to operate on highly active and corrosive process streams. Designed and developed by Gozani et al., it is distributed by IRT Corporation.

The technical specifications reported show excellent performance, namely :

- max sensitivity 10^{-8} Ci/l alpha in the presence of low beta activity
- max allowed fission product activity 30 Ci/l.

The high speed electronics option allows its use with a very high background of beta activity.

The system, which is being developed as part of a Euratom (Ispra) - CNEN (EUREX) contract, was preliminarily tested with uranium solutions and alpha and beta dry sources at Casaccia Research Laboratory, and at present is being evaluated at the EUREX plant.

Fig. 5 - EUREX Plant: Plutonium X-ray monitoring system installed on the final product evaporator condensate stream

Plutonium Measurement in Solid Waste

The first approach to the problem of solid waste disposal for reprocessing plants was to standardize the container.

Until 1978 wastes were placed into 200 l drums (height 85 cm, diameter 57 cm, thickness 1.5 cm).

Taking into consideration that the solid waste produced in reprocessing plants are mainly solids of low matrix density (gloves, paper, glass, plastic) and particularly with a low content of plutonium, CNEN has decided to apply a disposal procedure which uses smaller containers whose dimensions have been standardized as being 40 cm height, 22 cm diameter.

Eight such containers are, after the plutonium content has been measured, transferred into the 200 l drums. The smaller containment has been suggested in order to increase the effectiveness of accounting and safeguards measurements, to improve the measurement sensitivity and to avoid the need for drum segmentation reducing the cost of sophisticated instrumentation and counting time.

Among the available NDA techniques, gamma spectrometry was selected as being the most suitable and widely used.

The system set up at LRR-CNEN-Casaccia is based on one-shot measurement of a rotating container using a coaxial intrinsic Ge detector of 43 cm^3 volume.

The system has been transferred to the EUREX plant for the checking of the low matrix solid waste produced during the Candu fuel reprocessing campaign in order to meet the safety authority's prescriptions. The sensitivity obtained is within 1 mg Pu.

Furthermore, the Casaccia Research Laboratory will participate within the framework of an EEC contract in an intercomparison programme of measurements on containers prepared and made available by CEA.

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An Advanced Nondestructive Assay System for Reliable and Timely Nuclear Materials Accountancy in Reprocessing

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Introduction

Nuclear materials accountancy in reprocessing, and its independent verification by international safeguards authorities, requires the accurate quantitation of uranium and plutonium in liquid process streams, which may carry considerable levels of fission products and other interfering elements. The precise quantitative assay of plutonium in the highly active dissolver solution at the reprocessing headend is a well known and illustrative example for the analytical challenges encountered in the respective accountancy and verification measurements.

Three different analytical techniques have so far demonstrated their principal applicability to the plutonium analysis in reprocessing input solutions :

- mass spectrometric isotope dilution analysis, which is the currently adopted standard method for this assay problem;
- wavelength-dispersive X-ray fluorescence spectrometry of L X-rays, utilized at KfK for many years on a laboratory scale[1,2];
- spectral photometry of Pu VI [3].

All three methods are immune to the high activity of the solution samples, but require more or less elaborated and skillful procedures for sample preparation and calibration in order to arrive at accurate quantitative results.

In a recent paper[4] we have proposed an alternative approach, based on a purely nondestructive assay without need for any sample treatment prior to the analysis. The proposed instrument makes use of two proven nondestructive radiometric techniques : energy-dispersive K-edge X-ray absorptiometry (K-edge XRA), and energy-dispersive X-ray fluorescence (XRF) analysis of K X-rays. Both techniques have been integrated in a single instrument, where they can be utilized simultaneously, or individually, depending on the application. The hybrid K-edge XRA/K-XRF system thus offers the possibility for rapid and reliable accountancy and verification measurements for a wide range of applications. In reprocessing it is equally applicable to both input, intermediate process and product stream analysis. We shall take the reprocessing input analysis here as an example for illustrating major features of the hybrid system.

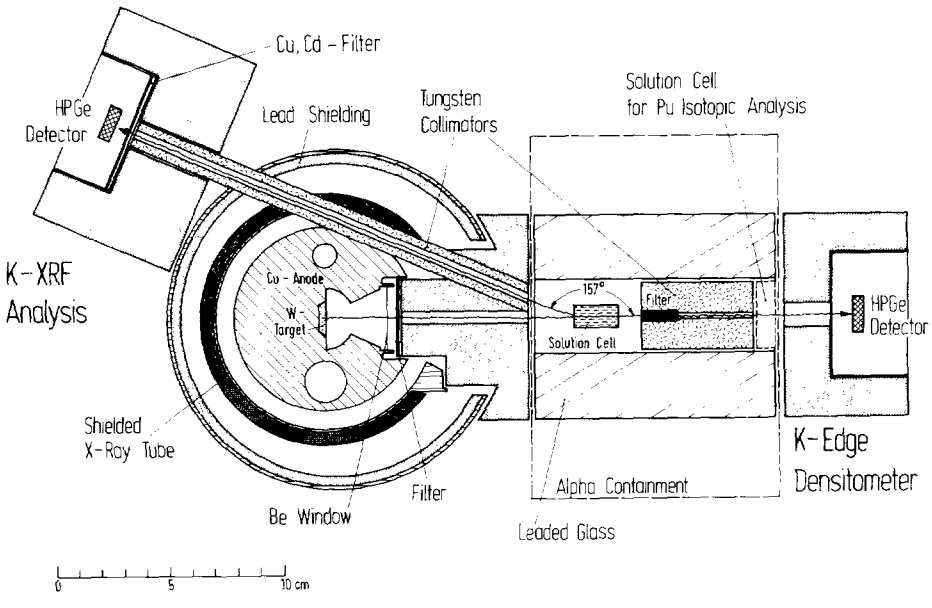
Instrument design

The hybrid system represents an upgrade of the previous K-edge densitometer developed at KfK under the Support Programme of the Federal Republic of Germany to the to the IAEA[4,5,6]. This instrument has been used during the last 3 years for a thorough performance evaluation of K-edge XRA using an interrogating X-ray continuum. The KfK densitometer incorporating this technique has demonstrated high accuracy, reliability and long-term stability.

Figure 1 shows a cross-sectional view of the hybrid system. A second X-ray detector has been added to the previous K-edge densitometer to provide the capability for simultaneous K-edge XRA and tube-excited K-XRF measurements for the same sample. The X-ray unit utilized in the hybrid system is equipped with a standard constant potential X-ray tube with maximum ratings of 60 kV/19 mA. The small and compact metal ceramic tube has shown excellent reliability during the previous 3 years of operation in the K-edge densitometer.

The K-XRF assay geometry shown in Fig. 1 has been chosen to optimize the assay precision and detection sensitivity. It represents a compromise between shortest possible distance between tube

Fig. 1 - Cross-sectional view of the hybrid system



focus spot and sample position, and largest possible backward angle for the observation of the fluoresced K X-rays. A large backward angle for the X-ray channel is desirable in order to remove inelastically scattered primary radiation from the energy region of fluoresced uranium and plutonium K X-rays. This goal has been achieved in the present set-up for tube voltage settings < 160 kV. The reduced background in the K X-ray regions is illustrated in Fig. 2, which shows a typical measurement example from a plutonium solution sample. The large bump arising from inelastically scattered primary radiation is energetically shifted below the plutonium X-rays.

The K-XRF part added to the K-edge densitometer provides the large dynamic range required for simultaneous element assay at large ratios. The resolving power of energy-dispersive K-XRF for uranium and plutonium is comparable to that obtained from typical diffractive L-XRF systems, with the additional advantage of strongly reduced sensitivity to matrix effects because of the much higher penetrability of K X-rays. On the other hand, energy-dispersive XRF systems do not exhibit the high immunity to sample self-radiation, as do the diffractive systems. We have overcome this difficulty of K-XRF analysis through proper instrument design, which now makes precise K-XRF assays feasible at solution radiation levels of a few 100 Ci/l).

The present set-up is designed for discrete sample measurements in removable solution cells. The transfer of the original solution sample into the solution cell, and its positioning in the spectrometer, represent the only sample handling operations needed for the measurement. This procedure, and the subsequent analysis, are easily accessible to fully automated operation.

Role of K-edge XRA and K-XRF

The high immunity to matrix effects, including high radiation levels from the sample, and the simple and straightforward manner for absolute element quantitation recommended K-edge XRA as a reliable reference method for absolute heavy element concentration measurements at levels ≥ 20 g/l.

In the dissolver solution analysis K-edge XRA provides the measurement reference basis in terms of an accurately determined reference value for uranium. The K-edge X-ray transmission spectrum from a typical LWR dissolver solution, shown in Fig. 3, displays the characteristic jump of the X-ray transmission across the uranium K-edge, from which the uranium concentration is deduced in a simple manner. The external nondestructive K-edge XRA measurement in the hybrid system thus takes the role of the internal isotope or element sample spiking procedure used in mass spectrometric isotope dilution analysis and wavelength-dispersive L-XRF analysis.

The problem of accurate quantitation from XRF - a traditional difficulty with any XRF measurement - is solved in the hybrid system through the simultaneous K-XRF and K-edge XRA measurements for the minor and major elements, respectively. The whole assay process for analysis of uranium and plutonium in dissolver solutions thus is simply reduced to the measurement of two ratios : the X-ray transmission ratio at the K-edge of uranium, and the intensity ratio of fluoresced K X-rays from uranium and plutonium.

Fig. 2 - K-XRF spectrum from an inactive plutonium solution (4.5 g Pu/l)

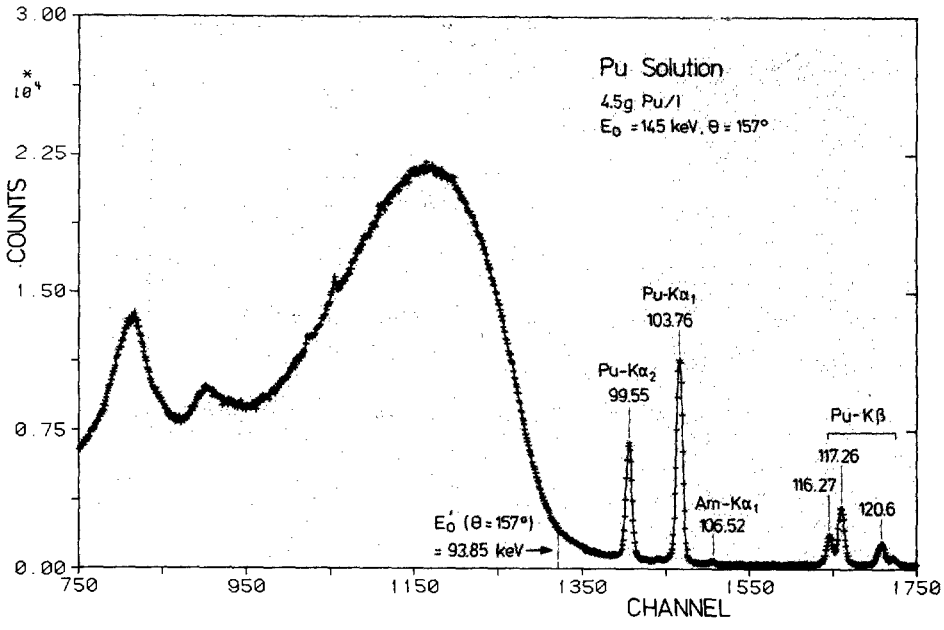
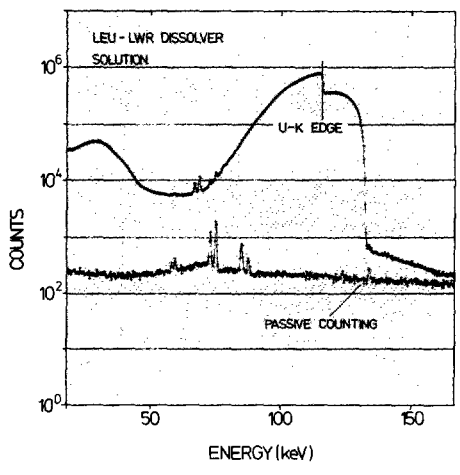


Fig. 3 - K-edge transmission spectrum (top) and passive spectrum from an LWR dissolver solution



Performance

The performance of K-edge XRA has been demonstrated previously for different applications, including the analysis of highly active dissolver solutions. An assay accuracy in the range of 0.2 to 0.3% for uranium in any type of dissolver solutions is safely within the capability of K-edge XRA.

Recent investigations have been directed towards the evaluation of typical performance characteristics of the new K-XRF part incorporated into the hybrid system. Priority has been given to the evaluation of its applicability to the plutonium assay in LWR dissolver solutions, which represented one of the major design objectives for the new K-XRF part.

The calculated and experimentally verified production rate for plutonium $K\alpha_1$ X-rays from a 1 ml solution sample containing 1 mg Pu amounts to about 2×10^7/sec in the present set-up, using tube settings of 155 kV/15 mA and 1 mm Cd for filtering the primary X-ray beam. This fluorescence yield is sufficient to compete with the fission product activity of typical dissolver solutions. On the other hand, the dose rate exposure to the sample of about 100 R/min is acceptably low to prevent the formation of photolytically produced gas bubbles in the solution, which could affect the K-edge XRA assay.

Exploratory proof-of-principle measurements have been carried out on representative dissolver solutions. Fig. 4 shows K-XRF spectra obtained from LEU-LWR and MOX-LWR dissolver solutions. The spectra represent 2000-sec counts from 0.3 ml samples. Burn-up and cooling time of the spent fuels were 42 GWd/t and 3 years for the LEU-LWR fuel, and 27 GWd/t and 2.7 years for the MOX-LWR fuel, respectively. The fission product activity, measured in the passive mode of the hybrid system from the long-lived fission products ^{106}Ru-Rh, ^{125}Sb, ^{134,137}Cs, ^{144}Ce-Pr and ^{154,155}Eu, adds in both samples to about 100 Ci/l. It has no significant impact on the plutonium assay precision. The only fission product gamma-ray line in the K X-ray region, the 105.3 keV line from ^{155}Eu, is well resolved from the plutonium Kα_1 X-ray.

The precisions in the measured ratio of uranium and plutonium Kα_1 peak intensities, deduced from repeat measurements of 2000-sec counting periods in the present prototype instrument, were 0.5% and 1% for the MOX and LEU-LWR fuel type dissolver samples, respectively. Neptunium in the LEU-LWR sample, present at a U/Np ratio of ~1500, was still measured with a precision of 5%. An upgraded compact hybrid system, currently under design for LWR dissolver solution analysis, will improve the above plutonium assay precisions by about a factor of 2.

The establishment of a single calibration constant will be sufficient to convert the uranium and plutonium X-ray peak intensity ratio into the corresponding atom ratio. This has been verified from mixed U/Pu solution measurements, which covered a wide range of total heavy element content and U/Pu ratios. The calibration approach thus turns out to be of similar operational simplicity as in the K-edge XRA assay.

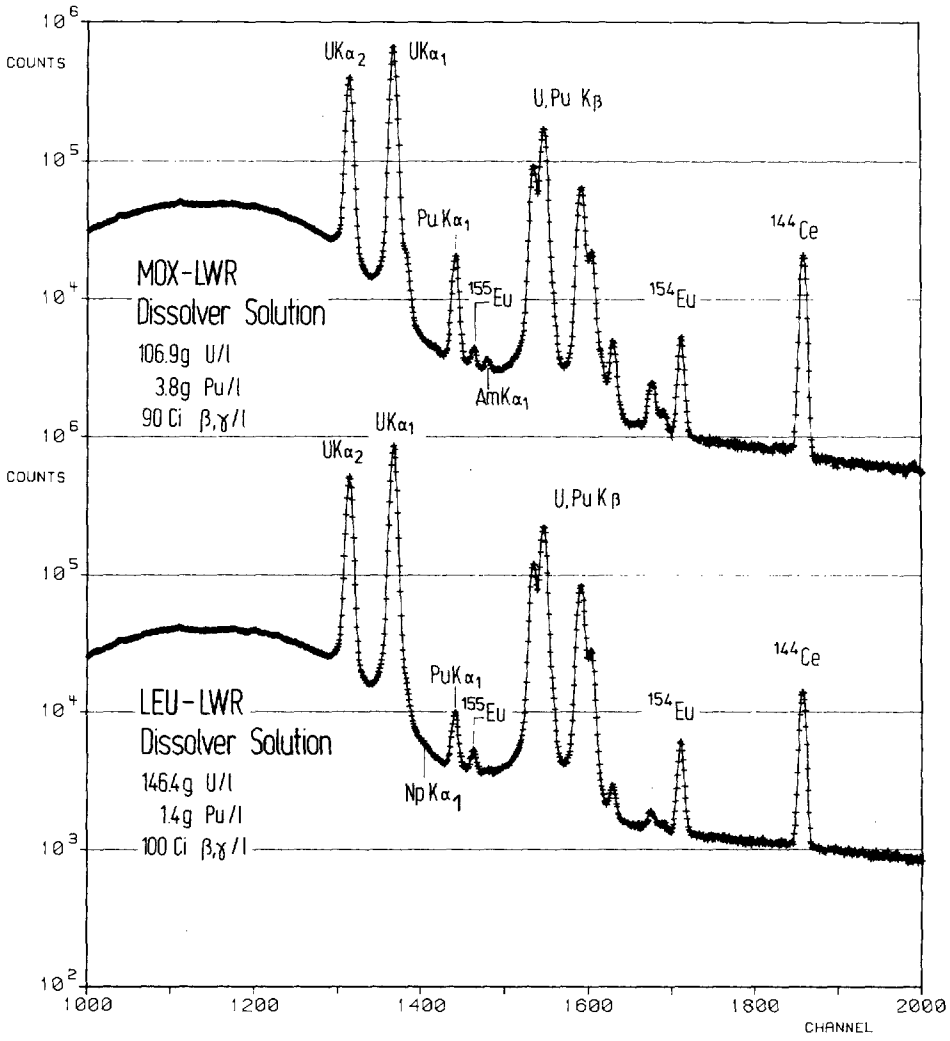
Outlook

The hybrid K-edge XRA/K-XRF system presented here has demonstrated the feasibility of a purely nondestructive assay approach to nuclear materials accountancy in reprocessing. All informations on uranium ***and*** plutonium concentrations in feed, intermediate process and product streams are obtained from a single instrument in a timely, reliable and transparent manner.

We should particularly emphasize the advantage of using a single technique of demonstrated inherent simplicity, transparency and operational stability, namely K-edge XRA, as reference method for both input and output measurements. Efforts needed for calibration, and for its validation and verification, are thus largely reduced. This fact, and the elimination of any sample treatment procedures, will certainly facilitate the assessment of both random and systematic measurement errors.

The combined K-edge XRA/K-XRF system marks a step forward in NDA measurement technology, to the benefit of both plant operators and safeguards authorities. Our previous positive experiences with K-edge XRA gives us confidence that the advanced integrated system currently designed at KfK will fully prove its expected performances under routine operation conditions.

Fig. 4 - K-XRF spectra from typical LWR dissolver solutions



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Activities of ESARDA Working Groups

Reprocessing Input Verification

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ESARDA Working Group ICT-RIA Changes to RIV

In 1982 the Working Group for Isotopic Correlation Techniques and Reprocessing Input Analysis (ICT-RIA) completed one of its major projects, the "Isotope Correlation Experiment - ICE", for which the final report was issued in July 1983.*

Following the experiment, the Group reviewed its past activities and achievements, and came to the conclusion that the isotope correlation technique has by now emerged from the research stage; in many applications concerning light water reactor fuels it can be (and, in fact, is) counted among the number of techniques available for accountancy and verification purposes.

The Group also noted that more extensive analysis of the of the whole measurement/verification system at the head-end is needed. In fact, it appears that i) the quality of the present analytical work could be further improved, since it falls short of its potential accuracy, and ii) the overall performance of the present measurement/verification systems is not yet quantified. These problems certainly rank higher in priority in respect of those related to the further development of ICT.

Consequently, the Group decided to reorient its activities and to focus its efforts on the possible improvements to be obtained in the measurement/verification systems at the head-end. This task also includes the evaluation of the effect of the penetration of other measurement techniques, now proposed for improving the verification system (γ and n measurements).

The proposed changes were endorsed by the Managing Board and by the Steering Committee of ESARDA, and are reflected in the title of the Group, which is now : **"Working Group for Reprocessing Input Verification - RIV".**

In consideration of the new orientation, various coordinators of the ESARDA projects appointed new members to the Group which, at present, includes permanent representatives from the following organizations:

Belgium: CEN/SCK
Commission of the European Communities
France: CEA, COGEMA
Germany (F.R.): DWK, KFA, KfK
International Atomic Energy Agency
Italy: ENEA
The Netherlands: ECN
United Kingdom: BNFL, UKAEA
U.S.A.: BNWL

Also, from November 1982, the Tokai Mura Establishment of the JAERI (Japan) is permanently represented on the Working Group.

Present activities of the Working Group

The Working Group is now engaged in three main activities:

1. In collaboration with the ESARDA W.G. for Mathematical and Statistical Techniques, an analysis is being performed of the procedures used by operators and inspectors in evaluating the data generated at the reprocessing input. General recommendations will be produced, in view of a possible harmonization of the procedures.
2. Concerning isotopic correlations, the group noted that the procedures of applications are not yet standardized, and different approaches are used by the different laboratories. A "Benchmark Exercise" was therefore initiated, to compare the merits and the performances of the various procedures proposed. These are being tested on a common set of data in respect of their ability: i) to assess the material balance at the reprocessing input, on the basis of the isotopic composition data provided by the plant operator, and ii) to discover various types of delibrate errors artificially introduced into the original data. The data for the exercise have been provided by the CEA; they concern two reprocessing campaigns performed at the Cap-de-la-Hague plant of COGEMA on fuel irradiated in the German reactor KWO, for a total of 53 batches.
   Several laboratories are taking part into this evaluation, namely CEA, JRC, KfK, CEN/SCK, ECN, IAEA, JAERI. Preliminary results are expected by mid 1983.
3. A reprocessing input integral experiment is planned in collaboration with DWK. The aims can be divided into: i) performance of a complete material balance of the head-end process, and ii) comparison of the efficiency of different verification procedures. This second task should include verification procedures still under discussion, such as: methods based on the analysis of fuel nuclides other than those used by the operator, NDA, fission product ICT.

*"The Isotope Correlation Experiment", edited by L. Koch (CEC, JRC-Karlsruhe) and S. Schoof (KfK-Karlsruhe). The report can be obtained from the editors or from the Convenor of the Working Group.

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