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Analysis of the Nuclear Fuel Cycle in European Community Countries up to the Year 2000

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prepared by the
ESARDA Coordinators*

Introduction

The peaceful use of nuclear energy for electricity production has substantially increased over the past years and prognoses estimate a further increase in the near future. As a consequence the quantity of nuclear material in store or in use will increase accordingly. Furthermore, the conditions under which it will be available to the safeguards authorities for verification purposes will change in some cases. Both of these factors will affect safeguards.

Presently available safeguards technology provides means for efficient safeguards performance, but this does not mean that no further improvement could be made. The problems related to the assessment of performance for present safeguards technology were discussed at the 8th Annual ESARDA Meeting, held at Copenhagen in 1986 having as subject the «Capabilities and Objectives of the Use of NDA-DA-C/S Measures in Safeguards» /1/.

The projected evolution of the nuclear fuel cycle is expected to pose new technical challenges, mainly in two areas :

1) additional resources and technical developments will be required to safeguard the increasing amount of nuclear materials and the number of facilities. If efficient safeguards performance is to be maintained, both cost-effectiveness and inspection strategy will have to be looked at more carefully;
2) current practices for the operation of industrial facilities are expected to change. Examples are the automation of nuclear material handling, recycling of nuclear material and long term storage of spent fuel. As a consequence a number of safeguards relevant characteristics of the new facilities will change as well.

Experience has taught that years may pass between the development of safeguards techniques and their routine utilization. Therefore it is necessary to take account of the expected evolution of the fuel cycle in long term planning of R&D.

Aim of the Exercise

It was decided within ESARDA that an analysis of the evolution of the nuclear fuel cycle should be carried out to indicate changing features that might affect safeguards R & D activities of the ESARDA working groups. An analysis of the nuclear fuel cycle evolution in EC countries up to the year 2000 was undertaken, *based on the latest information given in the open literature in the period 1985/86. Changes in the data since that time have also been taken into account.* It is to be emphasized that the primary interest was in the trends of the data rather than their absolute values.

Working Procedure

In practice the study was subdivided into four phases.

The first phase of the action, "fuel cycle analysis", was to gather, from the open literature /2,3/ for each EC country, the data on the industrial activities related to the production, processing, fabrication and use of nuclear material between 1985 and the year 2000. The industrial activities are to be understood here as the number and type of facilities, the throughput and storage capacities of nuclear materials.

The second phase involved the organization of the collected data, checking the coherence between data of different origin and the preparation of estimate, for all EC countries, of the annual and cumulative capacities of industrial activities.

In the third phase, an analysis was performed of the characteristics of the different parts of the fuel cycle which are expected to change substantially and which, consequently, may influence the application of safeguards techniques.

Finally, the fourth phase was the analysis by ESARDA Working Groups of the adequacy of existing techniques to solve problems which arise from the qualitative and quantitative changes in the fuel cycle and the identification of future R & D work /4/.

Evolution of Fuel Cycle up to the Year 2000

First and Second Phases

These phases comprise the data base of nuclear fuel cycle evolution up to the year 2000. For the purpose of the analysis the fuel cycle was subdivided into five parts :

- power reactors,
- storage outside reactor sites,
- reprocessing and intermediate storage,
- mixed oxide fuel fabrication (MOX),
- low enriched uranium fuel fabrication (LEU).

For each of these parts, the flow and storage of nuclear material, focused on spent fuel, plutonium and low enriched uranium, was estimated. The years 1985, 1990, 1995, and 2000 were selected as milestones, all estimates being related to these years.

The following general conclusions may be drawn :

a) the number of power reactors (essentially PWR) will increase and the installed capacity is expected to pass from 70 GWe in 1985 to 127 GWe in 2000 when the share of nuclear electricity generation will be as high as 70% of the total production.
b) the annual discharge of spent fuel assemblies will almost double from 1300 t of heavy metal per year in 1985 to 2400 t in 2000.
c) the reprocessing capacities for LWR fuel indicate a considerable increase by a factor of 8, when the planned facilities in EC Countries are put into operation in the mid-nineties. It is interesting to note that in 1985 approximately 90% of the reprocessing capacity was used for spent fuel from gas cooled reactors (GCR), whereas in 2000 70% is to be attributed to LWR spent fuel.

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The annual quantity of Pu separated is expected to rise from 8 t to 36 t between 1985 and 2000.

d) the mixed oxide fuel fabrication capacities will increase from 60 t MOX per year in 1985 to 360 t in 2000, essentially for the fabrication of MOX fuel for LWRs.

e) for low enriched uranium fuel fabrication plants, the present capacity for LWR-fuel is 2800 t rising to 3500 t in 2000. These facilities have already reached their commercial size and are in practice ready to respond to the growing demands of light water reactors.

More recently, forecasts on the evolution of the EC fuel cycle have changed slightly. The main changes concern nuclear electricity production from LWR reactors, LWR fuel reprocessing capacities and the production of separated Pu. It is expected that by the year 2000 the growth in all these three industrial activities will be approximately 10% lower than was expected from the 1986 data /2,3/.

However, these changes do not affect the overall trend in the consideration on safeguards characteristics on the fuel cycle and the future R & D activities of ESARDA working groups.

The input and output stores of reprocessing and MOX fuel fabrication plants will contain larger amounts of nuclear materials, due to the substantial increase of potential throughput and due to the tendency to store materials over a period of several years of operation of these facilities.

Because the large reprocessing plants will become operational only around 1995, very large amounts of spent fuel from LWR are to be stored in storage ponds and in dry storage facilities in the meantime.

Outside the reactor sites, at present, the total storage capacity for spent fuel in the EC is more than 15,000 t for LWR fuel (corresponding to more than 30,000 fuel elements).

It should also be noted that the fuel cycle inside the EC has several important links with countries outside the EC, in particular for the reprocessing of spent fuel and for fuel fabrication. This explains the apparent imbalance between the capacities of the different stages in the fuel cycle in the EC.

Third Phase

With regard to the third phase, safeguards relevant characteristics which might be subject to change due to fuel cycle evolution were identified. In this connection, to make a complete and systematic analysis, the safeguards approach for each type of facility must be taken into account. However, there is considerable uncertainty concerning some future facilities and this fact may influence, in a substantial way, some of the conclusions drawn from the study.

For each type of facility, the following safeguards relevant characteristics were analysed in relation to input store, output store and process area.

Input and output store

- *material type* : chemical and isotopic composition of different categories of nuclear materials;
- number of items per category at any given time on inventory;
- type of storage;
- accessibility to nuclear materials (for example for assay);
- residence time of nuclear materials;
- general layout of input and output storage areas, frequency of shipments and receipts.

Process area

- throughput of nuclear material (or core load);
- process inventory and residence time;
- accessibility to the nuclear material (e.g. for assay of chemical and isotopic composition of nuclear materials at input and output of process area).

The main safeguards relevant characteristics influenced by the evolution of the fuel cycle may be summarized as follows :

Power reactors

- Use of MOX fuel assemblies
- Larger variety of uranium enrichments and use of dismountable fuel assemblies
- Higher burn-up, influencing the quantities and composition of isotopes produced (fission products, heavy metal).

Storage outside reactor sites

- Dry storage facilities are completely new from the safeguards point of view.

Spent fuel reprocessing plants

- Large increase in throughput and process inventories
- Number and size of storage ponds will increase substantially.

Mixed oxide fuel fabrication plants

- Use of Pu of LWR origin, thus with higher radiation levels (Am-241) compared with material originating from GCR
- Very large increase in production of MOX assemblies for LWR and increase of process inventories
- High level of automation of production processes and remote handling of nuclear materials in stores.

Low enriched uranium fuel fabrication plants

- Increased use of recycled uranium
- Large variety of uranium enrichments.

In addition, the number of nuclear material transports will increase substantially, in particular spent fuel, plutonium oxide and MOX assemblies.

Fourth Phase

This fourth phase of the exercise consisted of an analysis by the ESARDA Working Groups on further technical developments in order to satisfy future safeguards needs, taking into account the above quantitative and qualitative nuclear fuel cycle evolution. Their conclusions are given below.

A. Destructive Analysis (DA)

At present, the activities of the Working Group have concentrated on :

- REIMEPs (Regular Interlaboratory Measurement Evaluation Programme) related to uranium hexafluoride and plutonium dioxide;
- updating of measurement performances (target values);
- updating of target values for sampling uncertainties;
- evaluation of authentication procedures for inspection sampling;
- promotion of new techniques by evaluating their capabilities;
- support for the calibration of NDA techniques.

As far as nuclear fuel cycle evolution is concerned, the Working Group did not see any substantial need for development of new DA methods. However the changing feature, which DA measurements must cope with, was identified as follows.

Because of the large inventory and/or throughput of future facilities the number of safeguards samples to be analyzed by destructive assay will increase by a factor of 3 to 5 in the next five years.

This will require both more attention to measurement uncertainties and an increase in measurement capacity of analytical laboratories.

Accordingly, a series of topics, to be addressed in the future, was identified, namely :

- introduction of robotized systems to cope with larger numbers of samples and to reduce radiation exposure;
- use of mobile in-field DA instruments (mass spectrometer, potentiometric titrator and spectrophotometer);
- installation of on-site laboratories for safeguards DA (large reprocessing facilities and possibly large MOX fabrication plants);
- use of hybrid K-edge for routine measurements where possible;
- improvement of sample preparation techniques;
- increased need for interlaboratory intercomparison exercises;
- increased need for sufficient supply of certified reference materials;
- increased use of automatic devices to remove subjectivity on measurement interpretation.

B. Non Destructive Analysis (NDA)

The activities carried out by the Working Group have, up to now, concentrated on :

- Plutonium Isotopic Determination Intercomparison Exercise (PIDIE) to assess

the capabilities of gamma-ray spectrometry;
- Promotion of Pu pilot reference samples by CBNM for gamma-ray spectrometry;
- Promotion of REIMEP related to uranium hexafluoride;
- Procurement of uranium enrichment standard for gamma-ray spectrometry;
- Studies on the definition of measurement performances for NDA;
- Discussion on the application of particular nondestructive techniques, such as CIND (Controleur Isotopique Non Destructif) and K-edge densitometry;
- Combined meeting with plant oriented Working Groups for the discussion of subjects of mutual interest.

Although future developments in the fuel cycle indicate an increase in amounts of nuclear material, changes in material composition and changes in measurement conditions, the physical principles of physics on which measurement techniques are based will not change.

The main features, which NDA measurements must cope with, have been identified as follows :
- increase in the number of fuel assemblies to be measured;
- increase in the use of neutron poisons in fuel assemblies;
- use of MOX fuels stored underwater;
- increase of Pu-242 content due to Pu recycling;
- stacking of cans in a single container for PuO_2 storage;
- increase in the variety of the characteristics for items to be measured.

As a consequence of these changing features, the errors in both gamma-spectrometry and coincidence collar measurements will probably have to be reassessed, while the high burn-up will probably affect instrument electronics in general and calorimetry in particular. Finally the Group pointed out that fuel assembly movements should be minimized when measurements are required.

The analysis of this information identified a series of topics to be studied, namely :
- Development of appropriate measurement techniques for fuel assemblies together with the application of C/S measures to maintain continuity of knowledge;
- Correction techniques to take account of Gd in LWR fuels;
- Multiplication correction in the measurement of stacked cans by neutron coincidence techniques;
- Development of Monte Carlo codes for microcomputers to be used in measuring items having different characteristics from those for which calibration is available.

C. Containment and Surveillance (C/S)

Among the various activities carried out by the Working Group the following deserve mention :
- C/S devices for spent fuel final storage;
- verification of containment;
- application of different seals, namely VACOSS seals, adhesive paper seals, fibre optic and ultrasonic seals;
- application of LASSY device.

The first general consideration of the group on the evolution of the fuel cycle was that, because of the increase of material in inventory and throughput, it will not be possible to rely only on measurement techniques and there will be a greater demand for C/S systems.

The Group identified a number of fuel cycle topics of relevance to C/S and examined current C/S technology. The analysis of these enabled the Working Group to identify a series of generic topics to be tackled in the future, as follows :
- processing of C/S data;
- integration of C/S systems e.g. the combination of C/S devices to enhance the performance of surveillance;
- development of design criteria for C/S systems;
- use of surveillance in more function-specific applications;
- use of intrusion/penetration monitors versus optical surveillance;
- study on how to express C/S assurance/performance;
- authentication of C/S devices.

D. Low Enriched Uranium Conversion/ Fabrication Plant (LEU)

The Group has concentrated its activities on the following items :
- Intercomparison on measurements performed with weighing scales, potentiometric titration and in-field application of DA measurements;
- Investigation of the performance of NDA techniques to be used in LEU plants.

In general, the future evolution of the fuel cycle will not affect the activities of the Working Group, although some trends of particular interest were identified, namely :
- introduction of higher U enrichment;
- use of elements with different U enrichment in the same assembly;
- increase in use of neutron absorbers;
- use of MOX fuel (in relation with the MOX working group);
- use of dismountable fuel assemblies.

The future actions of the Group can be summarized as follows :
- intercomparison exercises on the use of weighing scales, potentiometric titration, powder sampling;
- investigation of the performance of NDA techniques, such as Neutron Coincidence Collar and PHONID, with particular attention to the measurement of recycled uranium, neutron poisoned fuel and multi-enriched fuel;
- application of NUMSAS (a statistical tool for the calculation of material balance uncertainties) and of computerization of nuclear material management;
- evaluation of safeguards costs in relation to the use of NDA techniques and introduction of in-field DA measurements by inspectors.

Moreover, expected changes in the design of LEU-fuel, such as use of mixed oxide, a larger range of uranium enrichment and use of recycled uranium, will require some studies. Furthermore the Group intends to evaluate the applicability of ultrasonic sealing systems to PWR fuel assemblies.

E. Mixed Oxide Fuel Fabrication Plant (MOX)

The MOX Working Group spent considerable time, in the past, to reviewing and discussing for each of the different MOX fuel fabrication facilities in the EC, the state of practice of measurement systems as applied to eight categories of nuclear materials encountered in such facilities. In particular the topics included were :
- random and systematic errors for the different measurement systems;
- measurement strategy at batch level in connection with the reference plant;
- definition of sampling errors;
- analysis of Shipper-Receiver Difference (SRD);
- assay of Pu nitrate and level monitoring.

The Group has decided to redefine its activities and areas of discussion in view of the new development in the use of MOX fuel. Taking into account the difficulties that were encountered in identifying subjects of common interest that are not industrially sensitive and not related to safeguards approaches, the Working Group will concentrate its activities on the analysis of safeguards techniques and practices, and exchange views on their applicability to large throughput facilities.

F. Reprocessing Input Verification (RIV)

The RIV Working Group has concentrated its activities on problems related to the determination of nuclear materials at the input to reprocessing plants and to their verification.

The activities carried out, up to now, by the Working Group include :
- exchange of experience among the participants (operators, inspectors, researchers);
- experiments in laboratories and reprocessing plants in order to check the applicability of ICT and to compare their performance with traditional verification techniques (ICE at WAK);
- the Benchmark Exercise to assess the performance achieved by the various laboratories in determining and verifying

the input inventory by means of their respective ICT methods;
- studies on the determination by weight of accountancy tank transfers;
- experiment at the former Eurochemic Plant to re-assess the accuracy and precision achievable in volume determination of input solutions (RITCEX);
- workshop on «Determination of the volume of reprocessing input solutions by tracer techniques»;
- development of procedures, mathematical models, computer programs and data bases.

The Group remarked that the changes in reprocessing to be expected in the next years exclusively concern LWR fuels, for which an increase in the amount of fuel processed is forecasted. Despite that, the number of input batches, as compared to the present situation, is expected to have only a slight increase which should not lead to a corresponding change in the number of verification measurements.

It was also recognized that some changes will occur in the practice of operator measurements, mainly related to the adoption of weighing and use of NDA techniques (K-edge densitometer).

From all these considerations the Working Group felt that its future activities will be only slightly influenced by the envisaged evolution in the fuel cycle and a list of possible actions was proposed as follows :
- Study of the representativity of samples, also taking into account the presence of undissolved material;
- Study and improvement of liquid sample stability;
- Study of methods to determine the amounts of nuclear material contained in hulls, filter residues or other residues;
- Research for improvement in volume determination of input solution taking into account calibration and recalibration procedures, use of tracer techniques, use of pneumatic systems, use of direct weight and density determination;
- Study of the implementation of K-edge densitometry to inspector verification activities;
- Final conclusions of the results of past experiments on the applicability of ICT to specific safeguards problems.

G. Mathematics/Statistics (MAT/STAT)

The Group is intended to supply technical advice to other working groups in the field of error propagation of measurement techniques, material balance evaluation and systems analysis.

The topics the Working Group was involved in were :
- guidelines on statistical analysis for the Isotopic Correlation Experiment;
- frequency of inventory verifications in MOX;
- statistical methods for recognition of seal patterns;
- evaluation of methods for fitting calibration curves;
- evaluation of safeguards effectiveness;
- statistical problems related to reprocessing tank calibration;
- studies of measurement errors in NDA;
- inventory sample size computations;
- error modelling in weighing scales;
- sequential MUF analysis of operator data in reprocessing.

Regular meetings are planned to start again in order to examine new subjects to be treated, in cooperation with the other Working Groups, taking into account past experience.

Conclusions

The analysis of the evolution of the nuclear fuel cycle in the EC has provided interesting indicators on future developments and implications for safeguards. Although the data used in the analysis are subject to some uncertainties, the trends are nevertheless clearly discernible.

The Working Groups have systematically analysed the safeguards characteristics and have compared their present activities with the requirements imposed by the evolution in the fuel cycle, whose most notable change is the steadily increasing amount of nuclear material. This implies an increase in the scale of MOX fuel fabrication, reprocessing and spent fuel storage. As a consequence, further development of safeguards techniques will be required.

The study provided a clear indication of the future R & D needs for safeguards techniques, especially in view of the characteristically long lead times involved in developing new methods and techniques.

References

/1/ S. FINZI, M. CUYPERS, «Capabilities and Objectives of the Use of NDA, DA and C/S Measures in Safeguards, ESARDA Point of View», Proceedings of the 27th INMM Annual Meeting, New Orleans 1986, p. 27-34.

/2/ Summary of Nuclear Power and Fuel Cycle Data in OECD Member Countries. Nuclear Energy Agency, April 1986.

/3/ M. KEMPKEN, «Verzeichnis der Kernkraftwerke der Welt», Atomwirtschaft/Atomtechnik, November 1986.

/4/ G. STIENNON, R. CARCHON, M. CUYPERS, «Analysis of the Nuclear Fuel Cycle in European Community Countries up to the Year 2000: ESARDA Point of View», Proceedings of the 29th INMM Annual Meeting, Las Vegas 1988, p. 194-200.

Performance Evaluation of Non-Destructive Assay Techniques and Characterization of Standards for NDA in Safeguards

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Abstract

One of the major issues in the application of Non-Destructive Assay (NDA) in nuclear Safeguards is the availability of adequate Reference Materials or Standards. The need for standards is obvious when one considers that any NDA measurement is a relative measurement. Without reliable standards, only instrument intercalibration, consistency and process control measurements are possible, but never a quantitative accountancy. Besides calibration curve preparation and normalization, well-characterized standards are also essential for:

a) evaluating NDA performances;
b) improving NDA performances;
c) resolving current discrepancies between claimed and achieved performances.

On the one hand, the standards should almost always be very similar or even identical to the items measured in the field, which complicates the NDA scenario further.

On the other hand, it is practically impossible to have an NDA standard for each material family measured. One way to reduce the number of standards needed is to calculate corrections (e.g. by Monte Carlo techniques) in order to relate the measurement of a family to a standard "similar" to that family.

Still, when the "similarity" becomes loose, the correction factors (and related errors) increase to such an extent that the creation of a new standard is required.

At the Institute for Safety Technology of the Joint Research Centre, Ispra, a PERformance LAboratory (PERLA) has been set up with the primary scope of assessing the performances of NDA techniques for Safeguards.

In this paper the following points will be discussed:

- the need for field and laboratory standards for NDA;
- their required level of accuracy related to the NDA field and laboratory performances;
- the procurement schemes and procedures to be followed in their characterization;
- the need for an interlaboratory network where standards are intercalibrated and/or related to those already existing in the field so that a "path" may be produced from which the inspector's field measurements can be traced back to primary standards.

Throughout the paper, reference is largely made to the characterization schemes and procedures adopted for the large standard inventory (Pu and U) of PERLA, as well as to the overall uncertainty of those standards.

1. Introduction

Non-Destructive Assay (NDA) techniques have, in the last few years, become more and more important and are being used to a large extent in nuclear material accountancy and control. This is essentially due to two reasons:

1) The various improvements in most NDA techniques led some of them (calorimetry, gamma-spectrometry) to have performances close to destructive analytical techniques (DA).
2) The parallel improvement of the statistical and procedural inspection approaches led to the traditional scheme:
   - NDA for semi quantitative or consistency checks,
   - DA for quantitative measurements,

being abandoned. As a consequence, NDA is now frequently used in scenarios which involve a quantitative (by "variable") analysis. But, this evolution in NDA application requires that more and more numerous and better characterized standards be available.

The uncertainty with which an NDA standard or a calibration curve is known is frequently a not negligible component in the overall NDA measurement uncertainty. Since the NDA measurement uncertainty is, in turn, one of the major components of the inventory verification uncertainty, one can see that only when providing suitable standards, having uncertainties negligible with respect to other sources of error, can one appreciably improve the Safeguards measurement performances in certain types of plants.

It is thus necessary to prepare ever more accurate standards for NDA Safeguards. In turn this determines the fact that for some parameters the analytical DA techniques do not provide sufficient accuracy to fulfil NDA standard requirements, as will be shown further in this paper.

A last, more general, introductory consideration: **the need for each NDA inspection measurement** (not only NDA, obviously) **to be traceable back to a primary standard becomes ever more evident to improve Safeguards objectivity and transparency**. Different paths, composed of different steps, are possible to make that connection: for instance, a measurement may be traced back directly to a primary standard when it exists. In other cases the way may be much longer, implying a comparison with a secondary standard and a calculation of correction factors, thus increasing the overall uncertainty.

Concluding these introductory remarks one can say that, besides performing quantitative and accurate material accountancy, well-characterized NDA standards play a primary role in:

- the normalisation and calibration of measurements;
- bias corrections;
- resolving discrepancies;
- authentication of instruments;
- NDA performance assessment.

This paper presents a number of considerations based on the experience gained at the JRC during two years of PERLA operation.

In chapter 2, sets of basic definitions are given on the concepts of standards, NDA performances and related error components.

Chapter 3 deals with the aspects of preparation and characterization of standards, and presents the list of the nuclear material available at PERLA.

Chapter 4 presents the analysis of the uncertainty of neutron, gamma and calorimetry techniques. The performance values are complemented with sensitivity studies to evaluate when an uncertainty value becomes appreciable.

In chapter 5 a preliminary set of performance tables is proposed, based on current JRC experience, with the sole aim of serving as a basis for defining NDA standard needs.

At present, at PERLA, in collaboration with Safeguards Authorities and ESARDA, tables of **NDA performance values** are being defined to be used:

1) in defining the accuracy level of NDA standards, which is within the scope of this paper, but also
2) in planning inspections by Safeguards authorities, and
3) in Safeguards verification and accountancy to analyse operator-inspector differences.

The required accuracy for NDA standards is given in chapter 6.

In chapter 7, tables are presented reporting the DA performances as defined by the laboratories which participated in the Pu PERLA standard characterization.

In chapter 8, the need for better characterized NDA standards for laboratory and field is examined.

Chapter 9 is a first proposal of a framework of NDA standards which links international reference laboratories and the field, through specialised laboratories.

Finally, conclusions are drawn in chapter 10.

2. Definitions

In order to explain the definitions used in this paper they are explicitly mentioned. The first set of definitions is taken from Ref. 1. It is acknowledged /1, p. 47/ that more accurate definitions are given in Ref. 2, but as it is outside the scope of this paper to enter into a discussion about definitions, the authors preferred to use the definitions accepted in a Safeguards framework and in particular for NDA, as proposed in Ref. 1.

Certified reference material (CRM)

This is a RM accompanied by, or traceable to, a certificate stating the property value(s) concerned, issued by an organisation which is generally accepted as technically competent. NDA-CRMs denote CRMs used as a basis to quantify NDA.

Primary standard

This is an NDA-CRM which, on the basis of established analytical methods and standards, is documented to be accurate within a stated uncertainty in the parameter of interest and all other specified properties.

Secondary standard

This is an NDA-CRM which, on the basis of established NDA methods and/or certified standards, is documented to be accurate within a stated uncertainty in the parameter of interest and other specified properties.

Normalization sample

This is a sample selected from production material with a nominal value for the parameter of interest assigned from production data. These samples are used to:

a) check instrument reproducibility;
b) revalidate physical standard integrity;
c) normalize data at a later date when additional characterization of the samples has been done.

Traceability

This is the ability to relate individual measurement results to internationally recognised standards or nationally accepted measurement systems through an unbroken chain of comparisons.

A second set of definitions concerns the concepts of:

- **random error (or random uncertainty);**
- **systematic error (or systematic uncertainty);**
- **bias.**

To explain the interpretation given to the above concepts better, it is useful to describe briefly a typical NDA measurement procedure. Note that the measurement procedure itself /3,4/ generates a specific error structure: a different procedure (different calibration or normalisation patterns, etc.) would produce a different error structure (see also /5,8/).

The typical measurement procedure considered (simplified for the sake of clarity) is reported in the following.

A measured count rate (e.g. in a neutron detector) of an unknown sample "i" is normalised to a normalisation sample. The normalised count rate is then interpreted in terms of the parameter measured (e.g. Pu-240$_{eq}$) through a previously established calibration curve.

There are, obviously, other parameters, not always negligible, such as background count rate, dead time corrections, etc., that are neglected here. Now, in the above example we define the following **"error structure"**:

- the *random* component of the *uncertainty* as derived from the counting of the item "i";
- the *short-term systematic error* from the normalization sample counting: it is shared by measurements which are carried out in between two successive normalisations;
- from the counting statistical uncertainty of the calibration curve parameters (e.g. var a, var b, cov a,b in the case of a straight line calibration curve) and the uncertainty in the knowledge of the standards we derive the evaluation of two components of the *long-term systematic error*: calibration curve setting usually takes place less frequently than normalisation, this error component is therefore shared by all measurements that have been calibrated with the same curve.

Some measurement methods do not exhibit all of these elements. A case in point is the Pu isotopic composition measurement by means of gamma spectrometry using "instrinsic calibration". Here the error components are at most a bias plus a random error. /9,12/.

If, after having evaluated the total standard deviation propagating the above components correctly, the average discrepancies found with the certified or declared values can not be statistically explained by the uncertainty evaluated, then we are in the presence of a *bias*.

The distinction between a systematic error and the bias in this paper is whether or not it is reasonable, in the specific context, to consider the error component as having a probability distribution.

See also Refs. 13 and 14 where substantially similar definitions are given, with possibly the only exception concerning the definition of the bias.

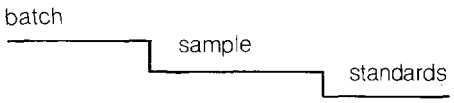
It is useful to illustrate some of the above definitions as applied in NDA.

The origin of the bias is frequently due to the fact that the standards used for creating the calibration curves do not come from a very well-known and/or characterized material, or even though standards are of high quality, the items measured belong to more spread batches with different characteristics. Typical examples are:

- fuel MOX pins with an isotopic composition which is very similar to that of the standards, but not identical: if not properly corrected, the measurement results will be biased;
- the presence of impurities, moderating matrices, in the items, but not in the standards;
- density, granulometry effects, etc.

All the above examples imply that the standards and calibration used and/or the items measured are no longer representative of the measured batch, which is a situation that frequently also occurs in the DA world, with obviously some differences.

In the application of NDA to a verification stratum, the stratum is rarely composed of homogeneous items and hence it is likely that the sample of items chosen for measurement also consists of items which, from an NDA measurement point of view, are different in matrix or geometry or whatsoever. If the inspector is obliged to measure these items with a calibration curve which may be a source of bias for some of them, then the D statistic created from these measurements would also be biased. Given the amplification that occurs in creating the D statistic, this could be a non negligible problem. More specifically we are frequently confronted with a three steps problem:



We have a batch, not always homogeneous; we take samples which may be not 100% representative of the batch, and perform measurements calibrated with standards that, in turn, may not be 100% representative of the samples.

In DA the situation is conceptually similar: the bias is almost always due to the non representativeness of the sampling, but since in DA the sample is treated to fit the standard, only the last mismatching is reduced or is "absent".

So, to summarise, the structure of a performance values Table for NDA should be rather more complicated than simply giving one figure for the uncertainty, or two figures: random and systematic components. A complete pattern should rather look as in the Table 1.

The uncertainty breakdown might be too extensive in certain NDA applications, or even too limited in others; in cases when, for instance, large background or other corrections are made, or when a simple Poisson model might be not adequate, a different error pattern should be studied.

Other definitions used in this paper are:

Precision

The precision of a measuring procedure is a measure of the closeness together of successive independent measurements generated by repeated application of the process under specified conditions /15/. This is usually quantified by the random error standard deviation.

Accuracy

The accuracy of a measurement procedure is the closeness of the mean of the measured values to the true value (Ref. 15). In this paper the "total uncertainty" is the evaluated inaccuracy of measurement results or of a measuring procedure.

Error/uncertainty structure

Is a table (as Table 1) of error sources or error components into which the total error is split.

Error/uncertainty pattern

Is the set of numeric values to be included in the above table.
Examples are Tables 3,4 and 5 where error patterns are given for various techniques (but for error structures nevertheless different from Table 1).

Performance, performance values

A general definition of performance is the following /42/: "whereas the knowledge of the overall uncertainty and error sources associated with measurement systems is the basic question, other important parameters must be considered in evaluating performance. These parameters are, for example:

- reliability-ease of implementation
- representativeness and authenticity
- intrusiveness to plant operation
- time to obtain a result
- cost

The relative importance of these parameters may also be different for each type of user".

Target values

"The target values are intended to describe the level of performance which many laboratories can achieve, or should be able to achieve, on a routine basis. More experienced laboratories would be expected to perform considerably better. Laboratories with less experience or entering in the field should aim at achieving this "target" performance level. In other words, Target Values attempt to describe a reasonably achievable "state of the practice" (as opposed to "state of the art", which is considerably better) /72,73/.

3. Procurement and Characterization Scheme of NDA Standards

One of the most important aspects of the preparation of standards for NDA is the characterization level: intended as the total uncertainty assigned to the various "certified" parameters. It is definitely clear that a standard must be planned for one specific technique (e.g. Neutron Coincidence Counting: NCC) or several techniques (NCC and HRGS = High Resolution Gamma Spectrometry), and must have a characterization level tailored for each of the techniques considered. To prepare a standard for calorimetry or for NCC does not necessarily require the same uncertainty level because their performances are rather different. Therefore, the first step in each standard preparation and characterization scheme is to define the required level of uncertainty of the NDA standard, based on the "known" performances of the NDA technique(s) for which the standard is prepared.

This uncertainty or characterization level will directly determine the amount of effort and procedure control and consequently of resources dedicated to the preparation of the standard and, therefore, its cost. Having good knowledge of the performance levels (uncertainty levels) of NDA techniques, when applied to various kinds of materials and in different operating conditions (field, laboratory) allows us to plan a well suited cost/effective policy of NDA standard preparation.

Table 1: NDA error structure

Component	Origin
Random error	Item counting statistics
Short-term systematic error	Normalization sample counting
Long-term systematic error	Statistical error from calibration curve set-up
Long-term systematic error	Characterization error on standards used for calibration
Bias	Non representativeness of standards and/or sampling
Other components (random or systematic depending on the measurement procedures)	Corrections (e.g. background, dead time, etc.)

One of the tasks of PERLA (PERformance LAboratory), created at the Joint Research Centre (JRC) of the Commission of the European Communities (CEC), Ispra, is to improve the knowledge of NDA performance values /16-17/.

An extensive set of Pu and U bearing standards (Table 2) was acquired and characterized /18-20/ to allow laboratories and Safeguards Authorities and Operators to calibrate their instruments and methods, and to assess NDA performances through tailored experiments. Table 2 also contains indications on the level of certification.

Summarising, the logical scheme for NDA standard preparation and characterization is the following:

- define NDA performances, structured for preparation of standards;
- define consequently NDA standard accuracy requirements;
- compare NDA requirements with DA performances;
- define preparation and characterization schemes suitable to attain the above requirements.
- set up a Quality Control programme to ensure that the above requirements are satisfied.

4. NDA Error Analysis

In this chapter the behaviour of the uncertainty components of some NDA techniques is discussed, with the main aim of defining their performances in different conditions such as counting time, burn-up value and required accuracy.

With this scope in mind, sets of sensitivity tables were produced concerning plutonium assay through calorimetry, passive and active neutron counting and High Resolution Gamma Spectrometry (HRGS), and uranium assay through active neutron counting techniques and again HRGS.

4.1 Plutonium Assay

4.1.1 Introduction

This section deals with the determination of the Pu mass in PuO_2 or MOX powders, MOX pellets and pins. The Pu assay, following the most commonly used approaches requires two measurements:

- an isotopic determination by NDA or DA;
- a neutron measurement (high level neutron coincidence counting or similar) or a calorimetric measurement for mass determination.

In section 4.1.2, a short introduction is given to High Resolution Gamma Spectrometry (HRGS) for plutonium and in sections 4.1.3, 4.1.4 and 4.1.5, respectively, the gamma techniques are related to:

- passive neutron determination;
- active neutron determination;
- calorimetric determination

to determine error propagation and sensitivity parameters.

4.1.2 Plutonium isotopic ratios determination by HRGS

The NDA technique most commonly used in the determination of Pu isotopic ratios applies the intrinsic calibration approach. When using intrinsic calibration, strictly speaking, no calibration standards are needed. Nevertheless, for a performance evaluation, to improve the technique and, finally, to meet the requirements of those techniques which use external calibrations (e.g. the so-called quasi-infinite thickness "approach" /21,22/, standards for the Pu isotopic determination are widely used and still more needed.

The evaluation of the uncertainty requirements for HRGS-Pu standards must be based, as for other techniques, on the performance values reached by HRGS for each isotope.

This is possibly one point which is still open: the correct definition of the NDA performance of HRGS when applied to plutonium isotopics, both in the laboratory and in the field are quite complex. Particularly uncertainties on sensitive isotopes like Pu-238, Pu-240, Pu-242 are the most difficult to assess, and a large gap can be seen between field and laboratory performances.

Tailored experiments have been done /21,52,53/ to throw light on HRGS performances, in both field and laboratory conditions.

As far as DA capabilities to fulfil NDA standard requirements are concerned at present, DA techniques are capable of providing sufficient accuracy for the characterization of standards for HRGS (see Chapter 6).

Table 2: Nuclear materials for PERLA

Material type			Certification level*
HEU	MTR platelets, plates	3 enrichments	4
	MTR assemblies (18)		4
	UO_2 powders, pellets (g. kg)	6 enrichments	3
	THTR particles, pebbles		3
	Metal buttons (kg)		4
LEU	UO_2 powder, pellets (g. kg)		
	UO_2 pins	not yet procured	
	Short assemblies		
	U_3O_8 CBNM/NBS	5 enrichments	1
PuO_2	Small cans (g)	3 burn-ups	2
	Large cans (kg)	3 burn-ups	2
	CBNM	4 samples	1
	PIDIE	7 samples	5
MOX	Pins	fast thermal	2
	Pellets	recycle	2
	Powders		2

* The certification levels are as follows:
1) International reference material or many laboratories
2) PERLA certificate (3 laboratories)
3) PERLA certificate (2 laboratories)
4) PERLA certificate (1 laboratory)
5) Others

4.1.3 Plutonium mass determination by Passive Neutron Assay and HRGS

The technique most frequently used to determine plutonium mass uses passive neutron counting from spontaneous fissions from Pu-238, Pu-240 and Pu-242. A weighed sum of these isotopes is called Pu-240 equivalent ($Pu\text{-}240_{eq}$). The emitted passive neutron flux is normally detected through coincidence circuitry (Shift Registers) or so-called multiplicity counters that use the feature that neutrons from the same fissions are correlated.

Hence, when considering the use of Neutron Coincidence Counting (NCC) for plutonium mass determination in combination with HRGS, the following major error sources must be taken into account:

- uncertainty in Pu-238 abundance = e_8
- uncertainty in Pu-240 abundance = e_0
- uncertainty in Pu-242 abundance = e_2
- uncertainty in $Pu\text{-}240_{eq}$ = e_{40} (NCC neutron)

A parametric sensitivity study, performed in 1985 /23/, for a LWR MOX fuel is shown

in Fig. 1: experimental data are taken from analyses performed at the JRC Ispra /24,25,26/ The total uncertainty of NCC measurements was then evaluated at different isotopic compositions (i.e. burn-ups). The study assumed as uncertainties the values shown in Table 3, which was a typical error pattern for NDA when applied in the field.

Table 3: Error pattern for NCC error evaluation

Source of uncertainty	Uncertainty
HRGS on Pu-238	e_8 = 5.0%
HRGS on Pu-240	e_0 = 5.0%
ICT for Pu-242 (*)	e_2 = 5.0%
NCC (neutron) on $Pu\text{-}240_{eq}$	e_{40} = 2.0%

(*) ICT: Isotopic Correlation Technique

The result of the study showed that, with the error pattern considered, the major contribution to the total error in the Pu weight determination comes from the HRGS measurement of Pu-240.

The situation today has evolved for some parameters: particularly we should quote the following as attainable performances (see also Table 9):

Pu-238 (HRGS) : e_8 = 0.5-2%
$Pu\text{-}240_{ec}$ (NCC neutron) e_{40} = 1%

But the two parameters mentioned above are, unfortunately, negligible in the total error, as is shown in Figure 1.

Instead, the performance in measuring the Pu-240 abundance is still quite low. 5% is not an underestimate of the present capabilities: new spectrum analysis tools claim better results /51,52,53/, but must still be extensively implemented.

Note that the quoted uncertainty for Pu-242 (e_2 = 5%) was quite optimistic; at the present time 10-20% might be a more realistic field value.

But in a future scenario, where:

- plutonium will always contain high Pu-242 fractions,
- Pu will be more and more blended,
- The Pu-240 abundance uncertainty is going to decrease

the major component of uncertainty in the determination of Pu mass with NCC plus HRGS could be represented by the Pu-242 uncertainty, as one might suspect from looking at Fig. 1, unless better Pu-242 abundance determination methods become available.

4.1.4 Plutonium mass determination by Active Neutron Assay and HRGS

A similar error sensitivity study /23/ showed that the uncertainty build-up in a Pu active assay of PuO_2 powders is different.

Plutonium active assay is performed by irradiating a Pu sample with a neutron source. In this case both passive and active signals can be used in the elaboration, or only the active signals coming from fissile isotopes (Pu-239 equivalent).

In Fig. 2, when assuming the error pattern given in Table 4:

Table 4: Error pattern for active neutron error evaluation

Source of Uncertainty	Uncertainty
HRGS on Pu-239	e_9 = 1.0%
HRGS on Pu-241	e_1 = 1.0%
Active Assay on $Pu\text{-}239_{eq}$	e_{39} = 2.0%

the results show that the highest (and constant over the whole burn-up range) contribution to the overall uncertainty comes, in this case, from the neutron measurement. In this case it is reasonable to try to reduce the neutron measurement errors, because this will decrease the Pu mass overall uncertainty significantly: if one could reach 1% in active neutron measurement accuracy, this would still be at the end a component comparable with HRGS components, all of the order of 1%.

4.1.5 Plutonium mass determination by Calorimetry and HRGS

Preparing NDA standards for calorimetry is one of the most challenging issues.

Calorimetry can be used to determine or verify the Pu content of a sample under the condition that its isotopic composition is known. The Pu mass (M) is calculated following the formula

$$M = W/P_{eff} \quad (1)$$

where W is the power output of the sample, and P_{eff} the effective specific power: the latter can be obtained by calculating the

Error Patten

Source of Uncertainty	Uncertainty
HRGS on Pu-238	e_8 = 5.0%
HRGS on Pu-240	e_0 = 5.0%
ICT for Pu-242 (*)	e_2 = 5.0%
NCC (neutron) on $Pu\text{-}240_{eq}$	e_{40} = 2.0%

(*) ICT: Isotopic Correlation Technique

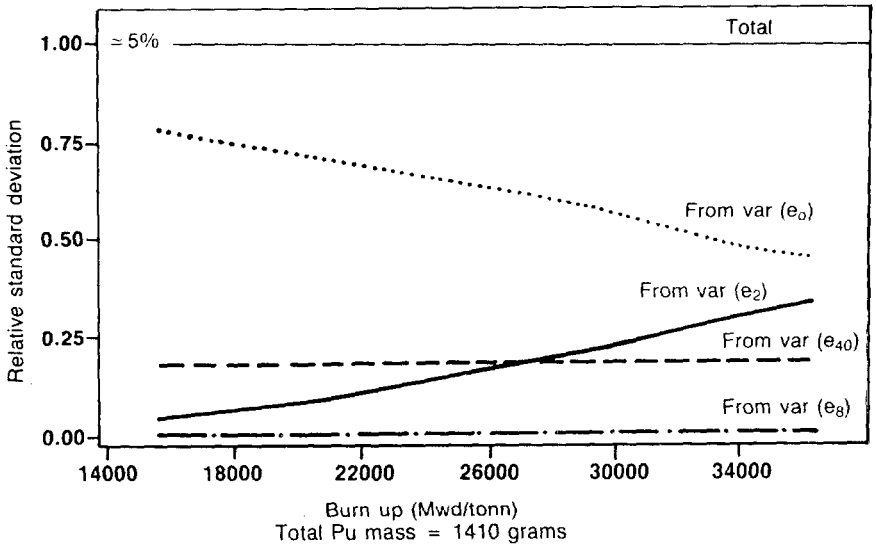


Fig. 1: Plutonium error propagation analysis: Passive Neutrons.

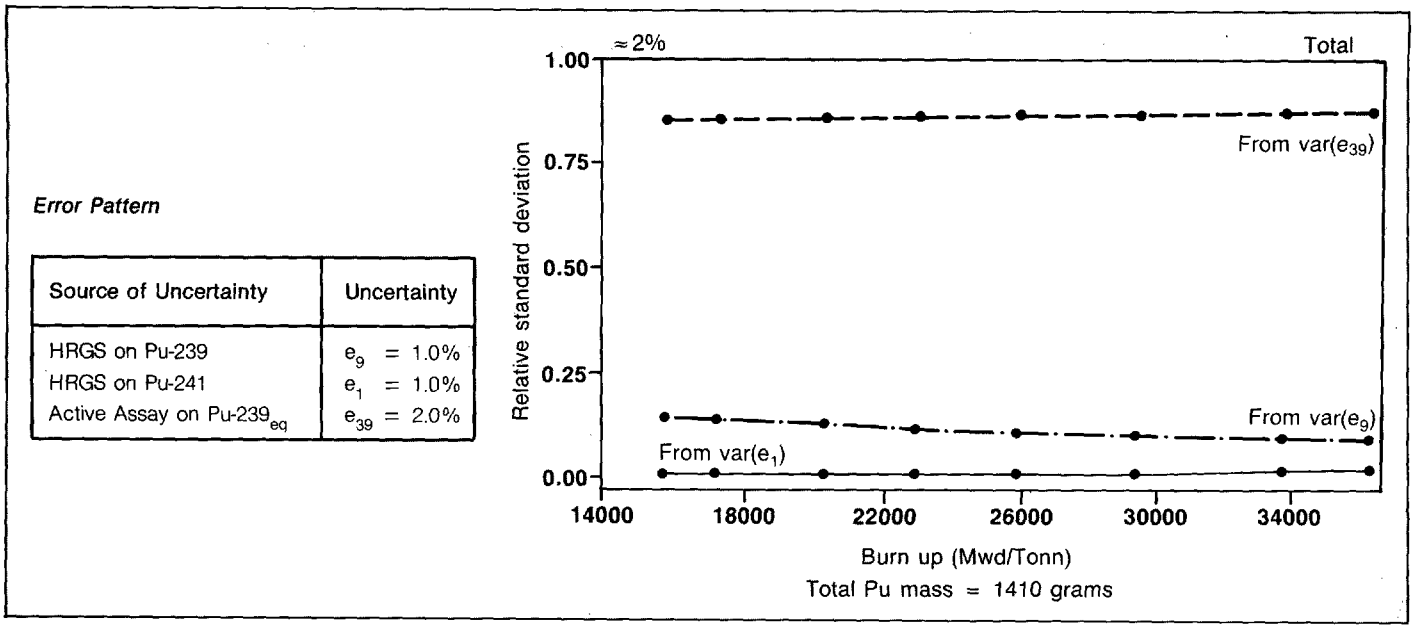


Fig. 2: Plutonium error propagation analysis: Active Neutrons

contribution of the different isotopes, according to the formula:

$$P_{eff} = \sum_i R_i P_i \qquad (2)$$

R_i being the isotopic fractions and P_i the corresponding specific powers.

The specific heat increases with burn-up, as shown in Table 5, where different burn-up values calculated for light water reactor MOX fuel are associated to the isotopic abundances of Pu and Am-241.

An error sensitivity study was carried out in Ref. 27 assuming nuclear data values from Ref. 28 and the uncertainty patterns in the knowledge of isotopic abundances, given in Table 6.

The measurement of the power was assumed to have negligible error contribution. Combining different "fuels" with the above error patterns, one could have a rather precise knowledge of the accuracies achievable by calorimetry in different conditions.

A summary of performances that can be achieved is given in Table 7 where results of error analysis are given for some of the above scenarios.

The values in the table show that, assuming the error pattern of Table 6, calorimetry can measure with error levels ranging from 0.3% to more than 1%, according to the different isotopic abundances and related accuracies.

Note that at very high accuracy level (0.1%) the assumption of negligible uncertainty from power determination may no longer be valid.

In Ref. 29 it is shown that the above uncertainty levels are experimentally achievable.

Table 5: Pu isotopic compositions and Am-241 content for different burn-ups (from Ref. 27)

Fuel No.	Burn-up MWd/t x1000	Pu-238 (wt%)	Pu-239 (wt%)	Pu-240 (wt%)	Pu-241 (wt%)	Pu-242 (wt%)	Am-241 (wt%)	Spec. power (mW/g)
I		0.01	93.	6.4	0.55	0.04		2.3239
II	8-10	0.60	85.68	12.11	1.94	0.22	0.16	2.9189
III	16-18	0.24	75.65	18.42	4.59	1.11	0.74	4.2850
IV	25-27	1.451	58.945	24.822	10.549	4.233	2.53	11.4910
V	38-40	2.0	45.	27.	15.	11.		14.6490
Spec. power (mW/g) (from ANSI 15-22, 1975)		567.16 ± 0.57 (0.10%)	1.9293 ± 0.0053 (0.27%)	7.098 ± 0.015 (0.2%)	3.390 ± 0.002 (0.06%)	0.1146	114.23 ± 0.16 (0.14%)	

Table 6: Error pattern for calorimetry error evaluation

Source of uncertainty		Uncertainty			
		Pattern 1	Pattern 2	Pattern 3	Pattern 4
HRGS on Pu-238	e_8	2.0%	1.0%	1.0%	1.0%
HRGS on Pu-239	e_9	0.3%	0.3%	0.3%	0.1%
HRGS on Pu-240	e_0	0.5%	0.5%	0.5%	0.5%
HRGS on Am-241	e_{41}	2.0%	2.0%	1.0%	2.0%

Table 7: Overall percentage error on the P_{eff} light water reactor fuel

Fuel number	burn-up (1000 MWd/t)	from Error Pattern 1	from Error Pattern 2	from Error Pattern 3	from Error Pattern 4
II	8-10	0.36	0.31	0.29	0.87
III	16-18	0.64	0.45	0.35	
IV	25-27	1.21	0.70	0.61	

To evaluate the individual isotope contribution to the overall error one can look at the difference between the results of error patterns 1 and 2 where the only difference is the uncertainty in Pu-238 which is 2% in pattern 1 and 1% in pattern 2. The effect of decreasing the Pu-238 uncertainty in high burn-up by a factor 2 is also to decrease the overall uncertainty (from 1.2 to 0.7) by roughly a factor 2. A reduction by a factor 2 in the Am-241 uncertainty has much less influence (see the comparison between patterns 2 and 3 in Table 7).

Nevertheless, in fuels with a low Pu-238 content and a high Am-241 percentage, a larger contribution (the largest perhaps) must be expected from the Am uncertainty (see also Table 5: the specific power of Am-241).

The results are also presented in the form of curves in Fig. 3.

In conclusion, when planning plutonium standards for calorimetry, one must consider that the contribution of sensitive isotopes such as Pu-238 and Am-241 is essential in the uncertainty build-up: a detailed quantitative and sensitivity analysis of the overall uncertainty is needed, and accuracies better than those provided by routine DA are necessary.

4.2 Uranium Assay

4.2.1 U-235 enrichment determination by HRGS

Another domain where NDA has reached a high level of accuracy is HRGS for U-235 enrichment. In Refs. 30,31 it is shown that the current technology allows accuracies in the determination of U-235 abundance of the order of few tenths of a percent, provided one has well characterized standards, with negligible uncertainty in comparison with the measurement errors.

The U-235 abundance is evaluated, determining the net peak area (A_{net}) of the unknown sample (A_x) and of the known standards (A_1,A_2), with an appropriate HRGS set-up. In Ref. 30 it is shown that the random uncertainty on A_{net} is, in a certain range, a function of the counting time. In a situation where an accuracy of the order of 0.1% in the random parameters is achievable in a counting time of the order of hours (see Fig. 4), a challenging demand is made of the DA characterization of standards: levels of accuracy of the order of 0.05% are required, such as those achieved material for gamma spectrometry /32/.

Table 8: Error pattern for U-235 abundance (HRGS) error evaluation

Source of uncertainty	Uncertainty
Net peak area of unknown sample	A_x = 0.1%
Net peak area of standard No. 1	A_1 = 0.1%
Net peak area of standard No. 2	A_2 = 0.1%
Enrichment of standard No. 1	E_1 = 0.1%
Enrichment of standard No. 2	E_2 = 0.1%

In fact, with the error pattern of table 8, where two standard enrichments (E_1,E_2) would have an uncertainty of 0.1%, one can see that the variance components coming from them are not negligible in the overall uncertainty build-up (0.14%) of the unknown enrichment E_x. A characterization level better than 0.1% is therefore required for the standards of the U-235 abundance determination.

4.2.2 Uranium-235 mass determination by Active Neutron Interrogation

Active neutron interrogation for the determination of the U-235 mass, both in total counting as in PHONID type instruments /23,33,34/ and coincidence counting in AWCC type instruments /35,36/, is not particularly demanding from the point of view of standard characterization.

The measurements consist in irradiating an uranium sample by a neutron source and in detecting induced fission neutrons. The signal is then interpreted in terms of U-235 trough a calibration curve

The best performance in those NDA techniques is of the order of 1%, while the U-235 mass can easily be measured by DA plus weighing with 0.1 - 0.2% even in the case of bulk samples. So, in the neutron-gamma combination, the standard for gamma measurements of the enrichment is, in general, the one that determines the highest accuracy constraint (see Table 6, chapter 5).

A slightly different scenario may occur in some specific applications, such as, for instance, the case of SIGMA /37,38/. SIGMA is a active interrogation-delayed neutron counting instrument belonging to the

Error Patterns (from Table 6)

	Uncertainty			
	e_8	e_9	e_o	e_{41}
Pattern 1	2.0%	0.3%	0.5%	2.0%
Pattern 2	1.0%	0.3%	0.5%	2.0%
Pattern 3	1.0%	0.3%	0.5%	1.0%

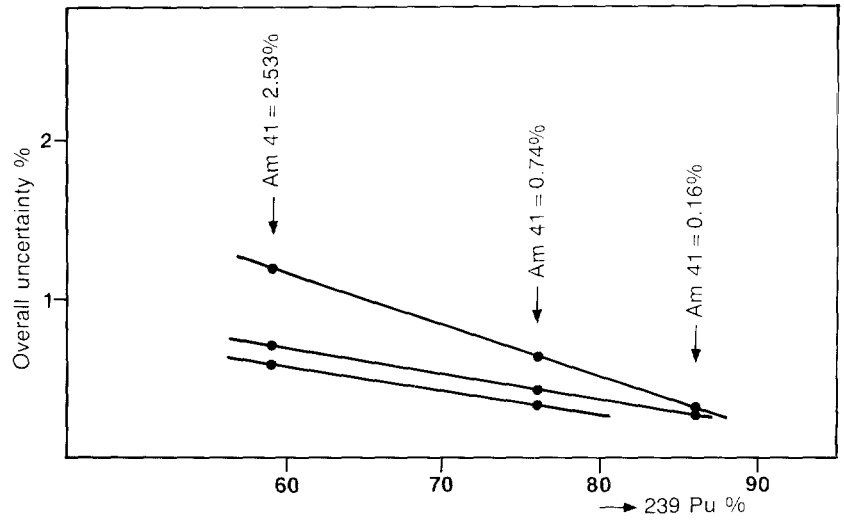


Fig. 3: Plutonium calorimetry: overall P_{eff} uncertainty evaluated at different burn-up levels.

Error Pattern

Source of Uncertainty	Uncertainty
Net peak area of unknown sample	A_x = 0.1%
Net peak area of standard No. 1	A_1 = 0.1%
Net peak area of standard No. 2	A_2 = 0.1%
Enrichment of standard No. 1	E_1 = 0.1%
Enrichment of standard No. 2	E_2 = 0.1%

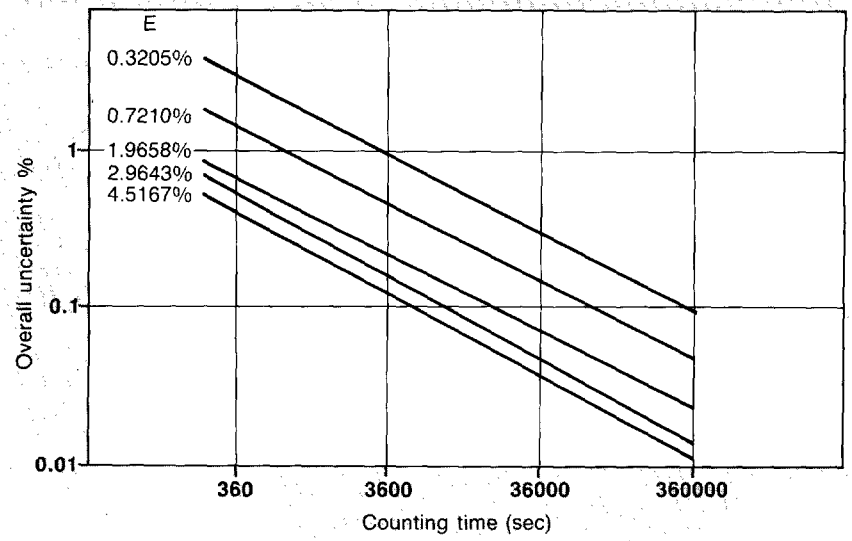


Fig. 4: Predicted A_{net} uncertainty as a function of counting time.

Euratom Safeguards Directorate, which has been used in a fuel fabrication plant since 1974 to verify U content in THTR fuel elements. The fuel element is a sphere (a pebble) composed of a 50 mm diameter core of graphite and coated particles containing a uranium-thorium mixture. The core is surrounded by a 5 mm thick layer of graphite. Each fuel element contains approximately 1 g of U-235. The device (see Fig. 5) was designed and made by JRC Ispra specifically for these fuel elements. This characteristic of having **always the same sample** determines its high accuracy, since no perturbation from exotic parameters such as humidity, geometry, multiplication, etc. can influence the instrument response. Provided one has a good calibration curve, SIGMA can give results of the U-235 content of the order of tenths of a percent.

The calibration is carried out on the basis of a linear regression fitted to the net corrected counts determined for the eight Reference Standard pebbles with a U-235 weight ranging from about 0.9 to 1 g (Fig. 6).

The THTR calibration standards were prepared as part of a large campaign of procurement and characterization of Plant Specific Reference Materials conducted jointly by Euratom Safeguards Directorate, IAEA and JRC staff. The standards were produced by HOBEG under IAEA-Euratom agreed specifications /39,40,41/.

The U-235 enrichment of the standard pebbles is 93% and there are three different weights: 0.899 g (two pebbles), 0.959 g (four pebbles) and 0.999 g (two pebbles), with a quoted uncertainty of 0.0009 g.

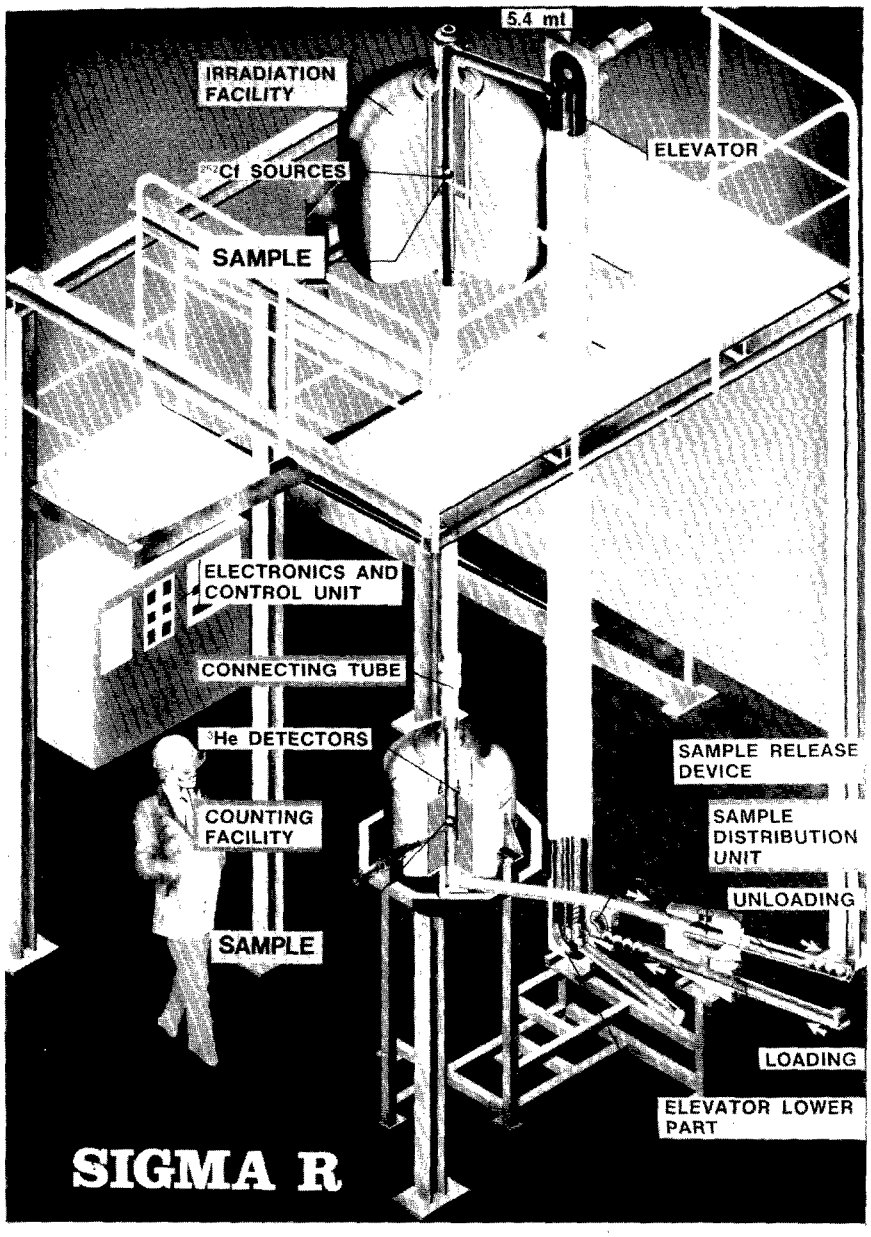


Fig. 5: General view of the SIGMA instrument.

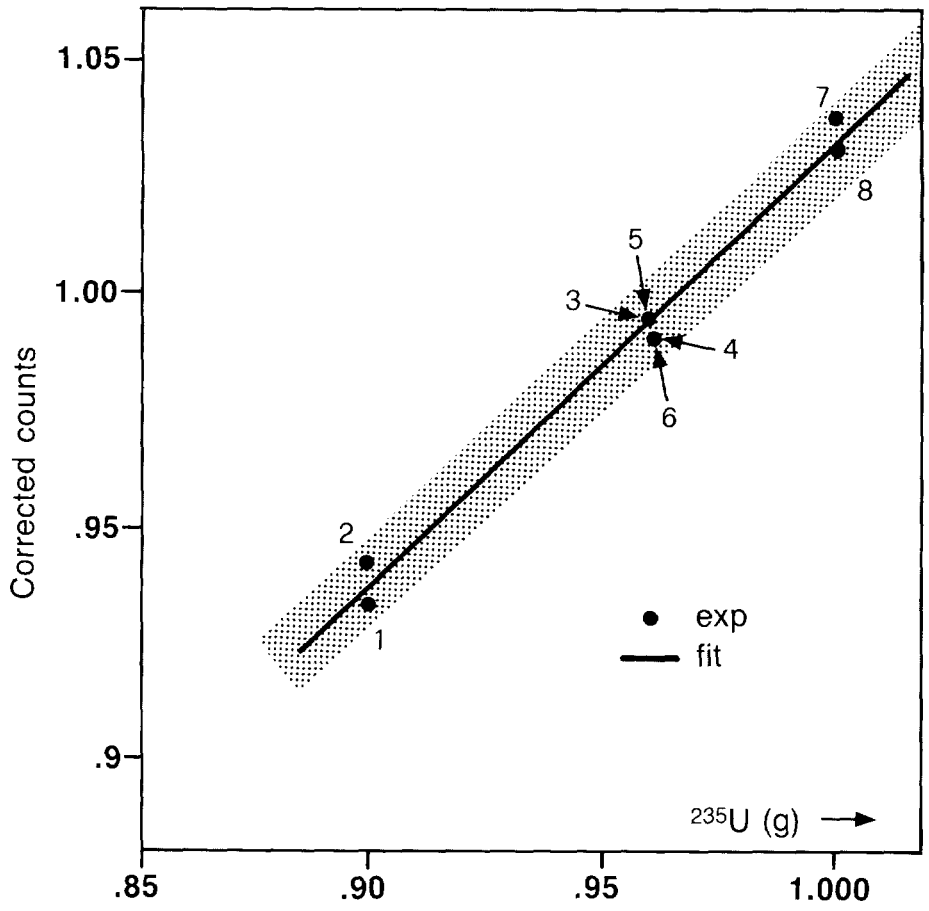


Fig. 6: Calibration curve with Plant Specific Standards.

The overall calibration error with the reference set remains small (between 0.15 and 0.3%) provided the weight remains within 5% of the central nominal weight "x".

In an interval "x" = ± 1%, the calibration error is negligible.

The example of SIGMA, even though on a very limited scale, is indicative of how well a good standard can fit in with an inspection strategy and reduce the NDA uncertainties to such a low level that a very accurate quantitative accountancy can be made just on the basis of NDA.

5. NDA Performance Values

A general definition of "performance" was given in chapter 2, but it is taken up again here for the sake of clarity:
"whereas the knowledge of the overall uncertainty and error sources associated with measurement systems is the basic question, other important parameters must be considered in evaluating performance. These parameters are, for example:

- reliability-ease of implementation
- representativeness and authenticity
- intrusiveness to plant operation
- time to obtain a result
- cost

The relative importance of these parameters may also be different for each type of user" /42/.

The authors of this paper share and assume the definition, but in the framework of this paper only the measurement uncertainty is analysed.

The previous chapters were a brief review of some important aspects concerning performances of NDA techniques when applied to certain types of materials and provided some specific examples encountered in field measurements.

In this chapter, based on the above discussion, we propose a set of "NDA performance values" in "laboratory" and "field" conditions. A generic structure of the NDA uncertainty in random and systematic components common to all techniques is in fact frequently insufficient to describe the error behaviour. As mentioned in chapter 2, not only is the error structure different from one technique to another, but also the same technique applied to different materials can determine a completely different error structure, such as, for instance, HRGS when applied to U-235 abundance or to Pu isotope measurements. Furthermore, the same technique applied to the same material with a different procedural scheme, can give rise to different error patterns and values.

For instance, in the active neutron techniques for U-235 mass determination, a different recalibration strategy may lead to completely different values of the short-term systematic error /3,43,44/. To enter into details in this complex field is beyond the scope of this paper. We limit ourselves here to giving only preliminary laboratory performances (uncertainty) for some NDA techniques that are represented by one figure called "overall uncertainty".

The table concerning the "laboratory performance values" (Table 9) is derived from the considerations expressed in the previous chapters and represents the present status of elaboration at the JRC PERLA laboratory. Giving only one figure for the overall uncertainty, not structured as suggested above, is in this case justified by the following considerations:

- Laboratory measurements are generally carried out in favourable and closely controlled conditions, so that the random error is negligible. The figures given in the table are, therefore, almost representative of the systematic component of the uncertainty.
- The standards used in the laboratory are generally very well characterized and documented, so justifying the absence of biases.
- Therefore as our scope is limited to using these values for establishing NDA standard uncertainty requirements, this structure is sufficiently good for the purpose.

The "laboratory performance values" given as well as the "field performances" given thereafter are also derived from the following literature:

for Pu passive neutrons:	Refs. 45-50
for HRGS on Pu:	Refs. 51-53
for calorimetry:	Refs. 54-57
for U mass determination:	Refs. 58-61
for U enrichment:	Ref. 62

Any further use of these performance values should strictly be considered under the above considerations. In field conditions. the other parameters which are considered negligible, could become very important.

The "field performances" given in Table 9 are a preliminary attempt to establish typical uncertainty values for field measurements, and they are intended (in the frame of this paper) to be used to define criteria for the preparation of secondary standards to be used in the field. The values in the table are dominated by the "systematic error + bias" and represent typical operator-inspector discrepancies. For "field" performances see also Refs. 63-69; for general NDA performance evaluation tables see also Ref. 70.

Some remarks on Table 9:

- The table is a summary of more extended tables in which particularly the material type and the error components

Table 9: NDA performance values (%)

Parameter	Material	Laboratory overall Uncertainty	Field		Technique
			random	system. + bias	
Pu conc.	PuO_2	(0.2)		(2-4)	densitometry
	MOX	(0.2)		(2-4)	densitometry
Pu-238 ab(*)	PuO_2	0.5	1-3	5	gamma spec
Pu-239 ab	PuO_2	0.5	0.5-1	2	gamma spec
Pu-240 ab	PuO_2	0.5-1	2-3	5	gamma spec
Pu-241 ab	PuO_2	1	1-2	3	gamma spec
Pu-242 ab	PuO_2	(3-5)		(5-20)	isotopic correl.
Am-241 ab	PuO_2	1		5	gamma spec
$Pu\text{-}240_{eq}$	PuO_2	0.5	0.5	5-10	NCC + HRGS
U-235 ab	UO_2 (powder pellets)	0.2	0.5	1-3	gamma spec
U-235 mass	UO_2 (LEU) (powder pellets)	0.5	1	1-2	active neutrons total counting
	UO_2(HEU) (metals powders or MTR)	0.5	1	1-3	active neutrons total counting or NCC
	THTR (pebbles)		0.2	0.4	active delayed neutrons

(*) ab: abundance

Table 10: Required overall accuracies for primary NDA standards (%)

Parameter	Calorimetry	N-active	N-passive	HRGS-U	HRGS-Pu
Pu conc.	0.1				
Pu mass	0.1				
Pu-238	0.2				(0.2)
Pu-239					(0.1)
Pu-240			0.2		(0.1)
Pu-241					(0.2)
Pu-242			0.3		
Am-241	0.3				(0.3)
U conc.		0.1			
U-235				0.05	
U-235 mass		0.1			

breakdown is more detailed and must be continously updated.

- The discrepancy between "claimed and achieved performances" as far as NDA is concerned is somehow fictitious: the feeling that the results obtained in the field are of low quality is misleading and it has slowed down the process of understanding.

In fact, there are at least two reasons that explain the discrepancies between laboratory and field performances (they also explain the structure of the tables, as mentioned above):

a) the first is the measuring time: the inspector in the field does not have "infinite" time to perform his measurements and this has an impact on two error components:

a.1) the random component on the item measurements, which is almost at a negligible level in the laboratory;

a.2) the statistical error component in the calibration curve (see Table 1).

b) the second is that in the real conditions ot the field operations it is almost impossible to have a calibration curve for any material type. Slight differences between the item inspected and the calibration curve material that do not justify the creation of another curve (with new standards), expand the systematic errors, generate bias and explain the difference between laboratory uncertainties and the systematic error + bias in the field.

- In this sense the systematic error column should be splitted further: the two components (systematic error and bias) have, for the moment, been merged to fit in with the most frequently presented tables.
- In the random error column the measuring times should appear: one must read the column as generated accounting for typical "field" measuring times.

6. Required Accuracy for NDA Primary (Laboratory) Standards

On the basis of the analysis made in the previous chapter, the required accuracies for primary NDA standards are reported in Table 10. The material types are those already reported in Table 9.

The structure of the table is the following: for each parameter of interest, the required overall accuracy value is referred to the most accurate technique. For instance, for the Pu concentration of total Pu mass in PuO_2 cans, the most demanding technique is Pu-238 and Am-241 again calorimetry is the technique that imposes the lowest overall uncertainty on NDA standards. The values of Pu isotopics by HRGS are in brackets because, whilst other techniques use standards for calibration purposes, in most applications HRGS on Pu does not need external calibration.

7. DA Capabilities

The last step in defining accuracies for NDA standards is to compare NDA requirements with DA performances. It is not the scope of this paper to make a review or give any statement on the performances of DA techniques for Safeguards. Many papers have been published to assess the DA performances, particularly concerning the so-called "Target Values" representing the state of the practice /71-78/.

In this paper we will mainly focus our attention on the PERLA experience on Pu bearing samples /19,20/. A summary of required and achieved accuracies in Pu-bearing PERLA standards is given in Table 11.

Some remarks on Table 11:

- Columns are ordered following the logic sequence for standard preparation already mentioned:
  - expected NDA performances,
  - required PS accuracies,
  - expected DA performances,
  - achieved DA accuracies,
  - "state of the practice" of DA
- The expected NDA uncertainty and, as a consequence, the required PERLA standard (PS) accuracy are dated back

to 1986-87. The values in Table 11 may therefore, be slightly different from those reported in Tables 9 and 10 which are now updated.

- The values are not structured in random and systematic components. As the scope of this paper is limited to assessing NDA performances for primary NDA standard definition, this approach is justified, as was mentioned above.
- Again Table 11 is a summary: a very detailed break-down is necessary for a complete review of the PERLA standard achievements. In particular, the column "expected DA values" is detailed in Table 12, where the expected DA performances for the three laboratories participating in the PS characterization are given for each measurement method employed.
- For the column "achieved PS uncertainties" a more detailed pattern is given in Ref. 19.

From Table 11 the following conclusions or comments can be drawn:

- For Pu-238 and Am-241 it can be seen that, as often mentioned in this paper, the required accuracy for the characterization of NDA standards is close to the capabilities of analytical laboratories: for Pu-238 in PuO_2 of low burn-up, the uncertainty reached (0.7%) does not represent a problem for calorimetry in PERLA; with such a low Pu-238 content (0.2%) the transmitted uncertainty is negligible.
- Concerning Am-241 the same conclusions can be drawn: a maximum of 0.85% declared uncertainty is not likely to propagate a substantial error component in calorimetric measurements, the Am-241 specific power being around 1/5 of the Pu-238 specific power. Nevertheless, in Pu lots with low Pu-238 content and relatively high Am-241, such a level of uncertainty may represent the prominent error component, and analytical certifying techniques could prove to be inadequate.

A more general conclusion that can be drawn from the table is that (in general) the achieved accuracies as evaluated by a two ways analysis of variance /79/ are generally lower (sometimes much lower) than both "expected DA" and DA target values. This is true for the element concentration determination and for isotopic ratio measurements

The values quoted for the performances achieved contain a component representing the instrument variability (18 results per parameter), a component linked with the sample variability (six samples) and a component representing the inter-laboratory variability (three laboratories). This last component is frequently the highest. On purpose we left out the calibration component (uncertainty on the DA of calibration standards) which is given separately in Table 13 and which might represent a sort of "state of the art" as far as DA performances are concerned.

Also the weighing error component was not propagated, since it was evaluated to be negligible /20/.

The good results obtained in PERLA standards, are certainly due to the rigorous characterization procedures and the quality control programme adopted, to minimize uncertainty sources like sampling, dishomogeneity, water content, impurities, weighing, conditioning and transport of the samples.

Experience again shows that an NDA characterization of bulk standard samples could find a limitation in the certified basic reference materials, if no or only a few new reference samples are prepared for plutonium at a high level (like those for instance of EC 201 standards). A lower certification level, may not be satisfactory. In fact, Tables 11 and 13 show that for some isotopes (Pu-240, Pu-241, Pu-242) the international certification uncertainties (and then the systematic components) are much larger than the statistical components generated by a well organized interlaboratory exercise.

8. Needs for Standards for NDA

After having discussed the accuracy levels of NDA standards, the authors feel the need to summarize their point of view as far as the need for NDA standards is concerned.

Merging the definition of standards given by the IAEA Advisory Committee (Ref. 1) and the distinction Laboratory-field made in this paper, we obtain a scheme like the one shown below:

Primary standards: for laboratory use
for field use

Secondary standards: for laboratory use
for field use

As far as primary standards are concerned both for laboratory and field use authors feel that they are now less urgently needed because on the one hand PERLA and other Laboratories are now well equipped and on the other hand field primary standards are costly, their preparation is time consuming and in any case some exist and procurement schemes and preparation procedures have been worked out (Ref. 39,40,41). The same is true for secondary standards for laboratory use.

Where Inspection Authorities have an urgent need of standards is for secondary field (facility dependent) standards, which are less costly, relatively quick to prepare, but represent a fundamental link in assuring traceability to the inspector measurements, to certified reference materials, as well as assuring the reduction of the most important source of uncertainty and bias today in Safeguards accountancy.

Table 11: Summary of Expected, Required and Achieved Uncertainties (%) in Pu-bearing PERLA standards (PS) compared with Destructive Assay Target Values.

Parameter measured	Material	Expected NDA Perform.	Required PS Uncert.	Expected DA Perform.	Achieved DA (PS) Uncert.	Target Values DA (%) (*)	
Pu conc	PuO_2	(0.2)	0.2	0.1-0.2	0.03-0.11	0.22-0.36	
Pu conc	MOX LWR	(0.2)	0.2	0.1-0.3	0.063-0.13	0.58-0.86	
	MOX FBR	(0.2)	0.2	0.1-0.3	0.045	0.36-0.45	
U conc	MOX	(0.2)	0.1	0.1	0.024-0.059	0.22	
						Target Values DA (%)(**)	
						s	e
Pu-238 0.3%	PuO_2	1.0	0.5	0.5-1.0	0.69	2.0	2.0
1.5%	PuO_2	1.0	0.5	0.5-1.0	0.17-0.21	0.7	0.7
Pu-239	PuO_2	0.5	0.1	0.03-0.06	0.005-0.31	0.1	0.1
Pu-240	PuO_2	0.5-1.0	0.1	0.07-0.1	0.006-0.22	0.2	0.2
Pu-241	PuO_2	1.0	0.2	0.2-0.3	0.04-0.1	0.3	0.3
Pu-242	PuO_2	5.0	0.3	0.3	0.06-0.15	0.3	0.3
U-235 Nat.	MOX			0.1-0.2	0.043-0.066	0.5	0.5
Enr.	MOX			0.03-0.1	0.018	0.03	0.03
Am-241	PuO_2	1.0	0.5	1.0-2.0	0.68-0.83		
	MOX				0.46-1.23		

(*) 1988 Target Values: Total random uncertainty (Ref. 73)
(**) 1987 Target Values: s = random uncertainty
e = systematic uncertainty (Ref. 72)

We want to stress here that considering their absolute importance in NDA, not only they are physically lacking, but even procedures for their preparation are neither defined nor agreed upon.

The authors strongly recommend that the IAEA and EURATOM support a campaign aimed to:

- identify needs for secondary field standards;
- define and evaluate field NDA performance values, with the aim of preparing field standards;
- define procedures and procurement schemes;
- prepare secondary field standards.

9. An Interlaboratory Standards Framework: the NDA Link.

There is another experience from the first two years of exercise of PERLA which might be worth mentioning.

When inspectors come to PERLA to follow training courses or to prepare calibration or intercomparison exercises, they find it very useful to the quality of their measurements, to calibrate instruments against standards which they know well, because they have the same at their Headquarters or they encounter them in-field. This is the case, for instance, for U_3O_8 standards already referred to /32/ or the CBNM pilot samples /80/ or some NBS standards for NDA /81/.

On the other hand Safeguards inspectors are daily and worldwide confronted with the calibration of NDA instruments and with the definition of the range of applicability of calibration curves to specific nuclear material encountered in the fuel cycle.

In order to assist the inspectors in this difficult and important task and to contribute to the improvement of the quality of measurements performed in different facilities, one has to make available to the inspectorates Reference Materials, as we have seen in the previous chapters and well calibrated measurement systems to be used following standard measurement procedures.

They should be located in well equipped laboratories (as well as in-field) in different parts of the world where important training and calibration exercises are required. The standards and well calibrated measurement systems should be identical or very carefully intercalibrated so that an NDA instrument can be calibrated independently in any of the above sites.

The standards similar to the family to be measured in a facility can then be characterized by DA techniques or by DA/NDA to agreed upon conditions described above. Probably even a limited number of these standards could be available in several laboratories (e.g. MTR plates, PuO_2 powder, pellets of LEU and MOX).

The specific material in a facility will finally be measured using calibration curves closely related to plant material using working standards and, if needed, some correction factors must be applied.

The procedure mentioned above creating a sort of link of standards between different laboratories and the field should be able on the one hand to improve NDA performances and on the other to create gradually an improved traceability of NDA measurements to primary standards or International Reference Materials.

Table 12: Typical expected uncertainties (RSD %) for DA measurement methods used for PERLA standards characterization.

Parameter	Material	Method	Lab. A	Lab. B	Lab. C
Pu conc.	PuO_2	AgO or Mc Donald	0.2	0.1	0.15
		Gravimetry	0.1		
	MOX	AgO or Mc Donald	0.2-0.3	0.1	0.15
	LWR Type	AgO	0.3		0.3
	FBR Type	AgO	0.2		
U conc.	MOX	Gravimetry	0.1		
		Davies & Gray		0.1	0.1
Pu-238	PuO_2	Mass spectrometry	0.5	1.0	0.5
	Low burnup	Mass spectrometry	1.0		
	Low enrich	Mass spectrometry			1.0
	MOX	Alpha spectrometry	2.0	2.0	2.0
Pu-239	All	Mass spectrometry	0.06	0.05	0.03
Pu-240	All	Mass spectrometry	0.1	0.1	0.07
Pu-241	All	Mass spectrometry	0.3	0.3	0.2
Pu-242	All	Mass spectrometry	0.3	0.3	0.3
U-235	Low enrich	Mass spectrometry	0.2	0.1	0.2
	High enrich	Mass spectrometry	0.1	0.03	0.1
Am-241	All	Gamma spectrometry	2.0	1.0	1.0

Table 13: Uncertainties on the International Reference Materials used in Pu-bearing PERLA standards characterization.

Parameter	Standard	Percentage Uncertainty	Date of preparation
Pu conc	CBNM EC-201	0.052	May 1975
	NBS 949f	0.08	October 1980
U conc	CBNM EC-110	0.015	February 1984
	NBS 950a	0.02	December 1961
	NBS 960	0.017	May 1972
Pu-238	CBNM SMS 6766	0.2	April 1982
	NBS 947	2.03	October 1971
Pu-239	CBNM SMS 6766	0.13	April 1982
	NBS 947	0.03	October 1971
Pu-240	CBNM SMS 6766	0.2	April 1982
	NBS 947	0.12	October 1971
Pu-241	CBNM SMS 6766	0.22	April 1982
	NBS 947	0.13	October 1971
Pu-242	CBNM SMS 6766	0.24	April 1982
	NBS 947	0.34	October 1971
U-235	NBS U010	0.1	April 1969
	NBS U500	0.1	May 1966
	NBS U750	0.03	February 1966
Am-241	CBNM ST4	0.25	January 1977
	ORIS EL4	0.8	June 1986
	PTB 5678-5679-5694	1.5	April 1988

10. Conclusions

10.1 General Conclusions

- The first aim of this paper was to stress the fact that a complete traceability to Certified Reference Materials must be assured to all inspector NDA measurements.

- The existence of well-characterized primary and secondary standards is an essential point in assuring that possibility.
- This paper has reported the existence of primary standards both for laboratory use and for in-field use, but it has also stressed the importance of Safeguards Authorities promoting campaigns for the preparation and the certification of secondary standards. They have the advantage that they can be prepared using DA and NDA, in a relatively short time. They are representative of the present production and can be given back to the plant operator when production changes and are thus much cheaper than primary standards. The lower accuracy level that secondary standards generally have, with respect to primary ones (remember definitions given in chapter 2), does not represent a problem, provided that it does not introduce a major error component.
- This paper also pointed out the role of NDA standards in:
  - normalizing and calibrating measurements;
  - correcting bias;
  - resolving discrepancies;
  - authenticating instruments;
  - assessing NDA performance.
- The paper was focused particularly on the role of NDA performances assessment in the preparation of the standards. But well structured tables of NDA performance values are essential and recommended in other fields also, such as:
  - in planning inspections by Safeguards Authorities;
  - in analysing operator-inspector differences in Safeguards verification and accountancy;
  - in any other cases in which NDA is applied.

10.2 Preparation and Characterization of NDA Standards

The experience of PERLA in preparing small and bulk NDA standards was used to recommend the following logic sequence in planning, preparing and characterizing standards:

- define NDA performances, structured for preparation of standards;
- define consequently NDA standard accuracy requirements;
- compare NDA requirements with DA performances;
- define preparation and characterization schemes suitable for attaining the above requirements.
- set up a Quality Control programme to ensure that the above requirements are attained.

10.3 NDA Performance Values

- A table with NDA performance values was given in chapter 5 to define characterization levels of NDA standards. The discussion pointed out that it is necessary, for NDA performance tables oriented for other purposes, like safeguards accountancy, to be more structured, to represent the error behaviour as a function of the measurement procedure.
- In particular it was pointed out that as far as the NDA performances are concerned, the model where in the laboratory "good" measurements are claimed and actually performed, and in-field lower accuracies are "achieved", is somehow misleading. There are in fact objective reasons for determining that discrepancy, reasons that are not always present in DA techniques.

10.4 DA Capabilities

- PERLA results were presented, where it was shown that for some isotopes (mainly Pu-238 and Am-241) analytical techniques are at the limit of offering performances suitable for NDA requirements. So preparing bulk standard samples for calorimetry may represent an objective difficulty, since accuracies better than those provided by routine DA are required for the above isotopes.
- It has also been shown that when sampling and control procedures are applied accurately, an interlaboratory average value can present (for some parameters) a variance lower than some evaluated uncertainties in the standard certificates. This aspect also says that the present high level of NDA performances requires standards with always higher accuracies in NDA laboratories and in the field, and that also in International Reference Laboratories (NBS, CBNM) there is a need for Certified Reference Materials for DA laboratories, for preparing NDA standards, the scheme of Fig. 7:

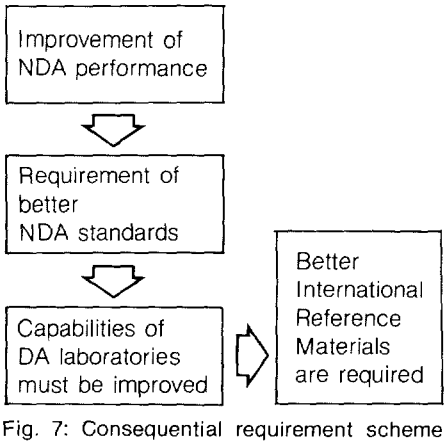


Fig. 7: Consequential requirement scheme caused by the improvement of NDA performance.

10.5 The NDA Link

A last conclusion, also drawn from the PERLA experience, was that it would be very useful, to improve NDA performances and traceability, to have a framework of common well-characterized standards Safeguards Laboratories.

11. References

/1/ Advisory Group Meeting on the Use of Physical Standards in Inspection and Measurements of Nuclear Material by Non Destructive Techniques, Vienna, IAEA doc. AG-112, 22-26 August 1979.

/2/ ISO REMCO 7, ISO Committee on Reference Materials, February 1976.

/3/ M. CUYPERS, M. FRANKLIN, S. GUARDINI, Methodology for NDA performance assessment, Proc. of the 27th INMM Annual Meeting, New Orleans, 22-25 June 1986.

/4/ M. FRANKLIN, Data evaluation for operator-inspector differences for a specific NDA instrument, Report EUR 9230 EN (1984).

/5/ M. FRANKLIN, S. GUARDINI, Information requirements and options for implementation of an integrated scheme for data evaluation for specific NDA instruments, EUR Technical Note No. I.05.C3.84.50 (1984).

/6/ G. DE GRANDI, M. FRANKLIN, S. GUARDINI, G. CORTELLAZZI, M. MONTAGNANI, Software, data structures, and data evaluation algorithms for a Verification Data Management System, Proc. of the 6th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Venice, 14-18 May 1984.

/7/ D.I. PIKE, A.I. WOODS, D.T. DUNN, The effect of the calibration history of a measuring system on the variance-covariance matrix of a sequence of measurements, Proc. of the 5th Annual Symposium on Safeguards and Nucl. Mat. Management, Versailles, 19-21 April 1983.

/8/ M. CUYPERS, G. DE GRANDI, M. FRANKLIN, K. MUELLER, S. GUARDINI, An integrated scheme for NDA data evaluation, Proc. of the 5th Annual Symposium on Safeguards and Nucl. Mat. Management, Versailles, 19-21 April 1983.

/9/ T.N. DRAGNEV, Intrinsic self-calibration of non-destructive gamma spectrometric measurements, IAEA report STR-60.

/10/ R.J.S. HARRY, J.K. AALDIJK, J.P. BRAAK, Gamma spectrometric determination of isotopic composition without use of standards, Proc. of the IAEA Symp. on Safeguards Nucl. Mat., Vienna, 20-24 October 1975.

/11/ R. GUNNINK, J.E. EVANS, In-line measurement of total and isotopic plutonium concentrations by gamma ray spectrometry, Lawrence Livermore Laboratory report, UCRL-52220 (February 1977).

/12/ T.N. DRAGNEV, M. FRANKLIN, The propagation of errors in the measurement of plutonium isotopic composition by gamma spectrometry when using intrinsic calibration, Proc. of the 26th Annual Meeting INMM, Albuquerque, 21-24 July 1985.

/13/ IAEA Safeguards Technical Manual, Part F: Statistical concepts and techniques, Vol. 3, IAEA-TECDOC-261, 1982.

/14/ J.L. JAECH, Statistical methods in nuclear material control, Technical Information Centre, Office of Information Service, TID 26298 (1973).

/15/ W. ZIJP, R. AVENHAUS, H. FRICK, G. HATMANN, Guidelines for the evaluation of data in nuclear material accountability and Safeguards statistical methods and examples, Proc. of the Symp. on Practical aspects of R & D in the field of Safeguards, Rome, 7-8 March 1974.

/16/ M. CUYPERS, T. DOYLE, S. GUARDINI, J. LEY, PERLA, a laboratory for training and performance assessment of Safeguards techniques, Proc. of the 27th Annual Meeting INMM, New Orleans, 22-24 June 1986.

/17/ S. GUARDINI, Progressi in PERLA, ESARDA Bulletin No. 14, May 1988.

/18/ M. CUYPERS, T. DOYLE, S. GUARDINI, G. GUZZI, J. LEY, S. ZANELLA, Procurement and characterization of nuclear material for the performance assessment of measurement techniques in PERLA, Proc. of the Int. Symp. on Nuclear Material Safeguards, Vienna, 10-14 November 1986.

/19/ C. BIGLIOCCA, S. GUARDINI, G. GUZZI, L. HAEMERS, F. MOUSTY, E. KUHN, N. DOUBEK, R. FIEDLER, A. ZOIGNER, S. BAUMANN, K.H. NELGES, G. HESBACHER, P. DE REGGE, L. VANDEVELDE, R. BODEN, D. HUYS, Characterization of Pu-bearing PERLA standards, Proc. of the 29th INMM Meeting, Las Vegas, 26-29 June 1988.

/20/ S. GUARDINI, G. GUZZI, F. MOUSTY, C. BIGLIOCCA, E. KUHN, N. DOUBEK, R. FIEDLER, A. ZOIGNER, S. BAUMANN, K.H. NELGES, G. HESBACHER, P. DE REGGE, L. VANDEVELDE, R. BODEN, D. HUYS, Pu PERLA Standards: quality control provisions during preparation and analyses of the samples, Proc. of the 11th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Luxembourg, 30 May - 1 June 1989.

/21/ J. MOREL, B. CHAUVENET, Preliminary results on PIDIE intercomparison exercise for the measurement of Pu isotopic composition, Proc. of the 11th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Luxembourg, 30 May - 1 June 1989.

/22/ T. DRAGNEV, B. BARNES, S. FIARMAN, Gamma spectrometric and absorption measurements on special nuclear materials, Proc. of the Int. Symp. on Nuclear Material Safeguards, Vienna, 10-14 November 1986.

/23/ R. CARCHON, G. SMAERS, R. COLOMBO, P. DELL'ORO, S. GUARDINI, A. PROSDOCIMI, Active interrogation of Pu and U bulk samples with PHONID devices, Proc. of the 7th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Liège, 21-23 May 1985.

/24/ S. GUARDINI, G. GUZZI, P. PERONI, Bench Mark report relevant to Trino Vercellese and Garigliano reactors, Report EUR COM 3609 (1978).

/25/ S. GUARDINI, G. GUZZI, Bench Mark report relevant to Gundremmingen and Obrigheim reactors, Report EUR COM 3703 (1979).

/26/ S. GUARDINI, G. GUZZI, BENCHMARK: reference data on post irradiation analysis of light water reactor fuel samples, Report EUR 7879 en (1982).

/27/ R. CARCHON, S. GUARDINI, Calorimetry for Safeguards and nuclear material management, ESARDA Bulletin No. 7, October 1984.

/28/ Calibration techniques for the calorimetric assay of plutonium bearing solids applied to nuclear material control, ANSI N.15-22 (1975).

/29/ C. BEETS, R. CARCHON, P. FETTWEIS, M. CORBELLINI, D. D'ADAMO, S. GUARDINI, W.W. RODENBURG, W.W. STROHM, S. FIARMAN, A. KEDDAR, Pu content verification by calorimetry, Proc. of the 6th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Venice, 14-18 May 1984.

/30/ G. CONTI, M. CORBELLINI, S. GUARDINI, Gamma spectrometry measurements for the determination of U-235 abundance with laboratory and field instruments, Proc. of the 7th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Liège, 21-23 May 1985.

/31/ P. MATUSSEK, Accurate determination of the U-235 isotope abundance by gamma spectrometry: A user's manual for the certified reference matrices, EC-NRM-171/NBS-SRM-969, Kernforschungszentrum Karlsruhe, Report KfK 3752 (May 1985).

/32/ a) R.J.S. HARRY, Enrichment Standards for Gamma-Spectrometry, Proc. of the 6th Annual ESARDA, Symposium on Safeguards and Nucl. Mat. Management, Venice, 14-18 May 1984.
b) P. DE BIEVRE, H.L. ESCHBACH, M. MEYER, J. VAN AUDENHOVE, B.S. CARPENTER, 235-uranium isotope abundance certified reference material for gamma spectrometry, EC - Nuclear Reference Material 171 Certification report, 1985, COM4153.

/33/ A. PROSDOCIMI, P. DELL'ORO, Performance and operation of a photoneutron active interrogation system for non-destructive assay of U-235, Proc. of the 2nd Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Edinburgh, 26-28 March 1980.

/34/ B.G.R. SMITH, A. VANDERGUCHT, L. BECKER, P. DELL'ORO, M. FRANKLIN, S. GUARDINI, The use of the PHONID for the routine measurement of low enriched uranium. Proc. of the 11th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Luxembourg, 30 May - 1 June 1989.

/35/ H.O. MENLOVE, Description and operation manual for the active well coincidence counter, Los Alamos National Laboratory report LA-10827 US, (ISPO-66), May 1979.

/36/ H.O. MENLOVE, J.E. STEWART, A new method of calibration and normalisation for neutron detector families, Los Alamos National Laboratory, LA-11229-US, April 1988.

/37/ M. CUYPERS, E. VAN DER STRICHT, M. BOURSIER, M. CORBELLINI, The verification of the U-235 flow at the output of the THTR fuel fabrication plant, Proc. of the IAEA Symposium on Safeguards Nucl. Mat., Vienna, October 1975.

/38/ P. AGOSTINI, L. BECKER, M. BERNEDE, L. CALDON, G. CORDANI, R. GALANTUCCI, S. GUARDINI, R. HAAS, A. JANSSENS, SIGMA-R: an improved version of the delayed neutron counting device for THTR fine element verification, Report EUR 2059 EN (1989).

/39/ R. ABEDIN-ZADEH et al., Preparation of a plant specific standard for MOX pins, Proc. of 4th Annual ESARDA Symposium Special Meeting on Harmonization and Standardization in Nuclear Safeguards, Petten, 27-29 April 1982.

/40/ R. ABEDIN-ZADEH, T. BEETLE, G. BUSCA, S. GUARDINI, E. KUHN, D. TERREY, Preparation and characterization of plant specific reference materials. results and progress, Proc. of the 5th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Versailles, 19-21 April 1983.

/41/ G. BUSCA, M. CUYPERS, S. GUARDINI, General criteria for the procurement of plant specific reference materials, Proc. of the 4th Annual ESARDA Symposium Special Meeting on Harmonization and Standardization in Nuclear Safeguards, Petten, 27-29 April 1982.

/42/ ESARDA Coordinators, Capabilities and objectives of the use of NDA-DA-C/S measures in Safeguards, Proc. of the 8th Annual ESARDA Meeting, Copenhagen 13-15 May 1986.

/43/ M. FRANKLIN, Non-detection probabilities of attribute mode sampling plans, Proc. of 11th ESARDA Annual Symposium on Safeguards and Nucl. Mat. Management, Luxembourg, 30 May - 1 June 1989.

/44/ J. MANDEL, The statistical analysis of experimental data, J. Wiley & Sons Inc., Interscience Publications, 1964.

/45/ M.S. KRICK, H.O. MENLOVE, The high level neutron coincidence counter HLMCC, Users Manual, LA-7779-US, LANL (1979).

/46/ A. RAMALHO et al., The high level neutron coincidence family of detectors, Nucl. Safeg. Techn. 1982, Vol. II, p. 81, IAEA STI/PUB/629.

/47/ B.W. HOOTON, F.J.G. ROGERS, Calibration of passive neutron coincidence and an assessment of overall errors, NMACT (84), p. 26, AERE Harwell, 1984.

/48/ Report of the Advisory Group Meeting on Progress in Neutron Coincidence Counting Techniques, Vienna, 7-11 October 1985, STR-206 (AG-506).

/49/ M. NEULLY, Precision of plutonium determination in dioxide powders, Proc. of the 9th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/50/ F.J.G. ROGERS, G.M. WELLS, Investigation into the performance of high level neutron coincidence counters under adverse conditions, Proc. of the 9th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/51/ H.G. WAGNER, J. GOERTEN, P. LOUIS, EURATOM experience with in-field gamma spectroscopy, Proc. of 29th Annual INMM Meeting, Las Vegas, 26-29 June 1988.

/52/ J. MOREL, B. CHAUVENET, Final results on the PIDIE intercomparison exercise for the measurement of Pu isotopic composition, to be published.

/53/ PERLA Intercomparison Exercise on Plutonium Spectra, Ispra, June-July 1989, to be published as an EUR report.

/54/ R.B. PERRY, S. FIARMAN, Recent developments in fast calorimetry, Proc. of the 29th Annual INMM Meeting, Las Vegas, 26-28 June 1988.

/55/ S. FIARMAN, W. HIGINBOTHAM, S. MUGHABGHAB, S. SANBORN, Material accountancy verification strategies for PuO_2 and MOX powders and pellets, Final Report ISPO Task C.5f, TSO-86-16, BNL, June 1986.

/56/ R.M. AUGUSTSON, W.W. STROHM, Analysis of data process simulated physical inventory verification exercise at a mixed oxide fuel fabrication facility, Report LA-UR-84-3617 (1984).

/57/ J.A. MASON, R.W. WILDE, J.C. VICKERY, B.W. HOOTON, G.M. WELLS, Development and evaluation of a plutonium assay calorimeter test-bed, Report LA-UR-84-3617 (1984).

/58/ D.T. FRASER, G.P.D. VERRECCHIA, M.S. KRICK, H.O. MENLOVE, T.R. CANADA, P. FREDERIKSEN, E. JORGENSEN, Calibration of the active well coincidence counter for cylindrical, high enriched uranium aluminium fine assemblies, Report EUR 10426 EN (1986).

/59/ G.P.D. VERRECCHIA, B.G.R. SMITH, S. GUARDINI, The calibration of four Euratom active well coincidence counters for a wide range of high enriched uranium material, Proc. of the 11th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Luxembourg, 30 May - 1 June 1989.

/60/ P. DELL'ORO, G. HUNTER, B.W. OLDFIELD, A. PROSDOCIMI, F.J.G. ROGERS, Field experience on non destructive assay of low enriched uranium by means of photoneutron interrogation, Proc. of the 6th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Venice, 14-18 May 1984.

/61/ P. SCHENKEL, B.G.R. SMITH, S. TSALAS, H.O. MENLOVE, M.S. KRICK, P. DELL'ORO, A. PROSDOCIMI, T.R. CANADA, J. FAGER, F. SCHINZER, Calibration and experimental comparison of the active well coincidence counter and PHONID II, Report EUR 10377 EN (1986).

/62/ J.L. PARKER, M. BROOKS, Accurate, wide range uranium enrichment measurements by gamma ray spectroscopy, Los Alamos National Laboratory, Report LA-11277-US, September 1988.

/63/ G.M. WELLS, F.I.G. ROGERS, B.G.R. SMITH, G.P.D. VERRECCHIA, H.G. WAGNER, Evaluation of the performance of UKAEA and Euratom gamma spectrometers in determining plutonium isotopic abundances, Proc. of the 9th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/64/ R. GUNNINK, A new core detector analysis method for rapid high-precision plutonium isotopics measurements, Proc. of the 9th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/65/ H.G. WAGNER, P. LOUIS, J. GOERTEN, R. SCHENKEL, R. GUNNINK, W. ROHTER, D. CAMP, Field experience of EURATOM with LLNL-MGA code for Pu gamma spectrometry, Proc. of the 11th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, Luxembourg, 30 May - 1 June 1989.

/66/ B.G.R. SMITH, G.P.D. VERRECCHIA, F.A.G. ROGERS, G.M. WELLS, Calibration of EURATOM and UKAEA neutron coincidence counters for PuO_2 and mixed oxide samples, Proc. of the 9th Annual Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/67/ S. FIARMAN, R.B. PERRY, Field test results of fast calorimetry using the Argonne bulk calorimeter at Rocky flats and TA-55, TSO 87-18, BNL, 1987.

/68/ B.G.R. SMITH, Proc. of the 30th Annual INMM Meeting, Orlando, 1989.

/69/ R. SCHENKEL, Recent progress in the test and implementation of measurement equipment for Safeguards applications, ANS winter Meeting, 1988.

/70/ DONALD R. ROGERS (Ed.), Handbook of Nuclear Safeguards Measurement Methods, Monsanto Research Corporation, NUREG/CR-2078, MLM-2855.

/71/ W. BEYRICH, W. GOLLY, G. SPANNAGEL, P. DE BIEVRE, W. WOLTERS, The IDA-80 measurements: evaluation programme on mass spectrometric isotope dilution analysis of uranium and plutonium, Vol.1: Design and results, KFK 3760, EUR 7990 e, December 1984.

/72/ P. DE BIEVRE, S. BAUMANN, T. GÖRGENYI, E. KUHN, S. DERON, P. DE REGGE, J. DALTON, R.E. PERRIN, C. PIETRI, 1987 Target values for uncertainty components in fissile isotopes and element assay, Proc. of the International Symposium on Nuclear Material Safeguards, IAEA-Vienna, 10-14 November 1986.

/73/ P. DE BIEVRE, S. BAUMANN, R. SCHOTT, J.C. DALTON, R. BERG, P. DE REGGE, T. GÖRGENYI, M. HÖFLICH, S. DERON, E. KUHN, E. MAINKA, R.P. BRADSHAW, C. PIETRI, 1988 Target values for random uncertainties in sampling and element assay of nuclear materials, ESARDA Bulletin No. 13, October 1987.

/74/ P. DE BIEVRE, W.H. WOLTERS, The regular European interlaboratory measurement evaluation programme REIMEP, Proc. of the 9th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/75/ G. GUZZI, A. FEDERICO, R. DEPAUS, Laboratory and in-field performance tests of the transportable thermoionic quadrupole mass spectrometer (THQ Finnigan MAT) for solid U samples isotopic composition determination, Commission of the European Communities Technical Note No. I.07.C3.86.24 FMM 116 (1986).

/76/ G. GUZZI, A. FEDERICO, R. DEPAUS, In-field performance of the "thermoquad" transportable mass spectrometer for the isotopic composition determination of solid U-bearing materials, Proc. of the 9th Annual ESARDA Symposium on Safeguards and Nucl. Mat. Management, London, 12-14 May 1987.

/77/ G. GUZZI, A. FEDERICO, Performance evaluation of the transportable VG336 mass spectrometer: measurement of U- and Lu-bearing samples, Commission of the European Communities Technical Note No. I.88.120 (1988).

/78/ F. MOUSTY, R. PASSARELLA CURCILLO, P. BOERMANS, B. MARIN, S. AURICCHIO, R. DEPAUS, In-field potentiometric determination of uranium in uranium dioxide pellets and powders, ESARDA Bull. No. 15, November 1988.

/79/ H. AIGNER et al., ANOVA: two way variance analysis for the statistical evaluation of the results obtained with a single method, Report IAEA/RL/85, September 1981.

/80/ Commission of the European Communities, CBNM nuclear reference material 271, Certificate of Analysis 1989.

/81/ National Bureau of Standards, standard reference material 947, Certificate of Analysis 1972.

News about ESARDA

We are pleased to inform that Mr. J. Regnier of COGEMA is now a representative of France in the ESARDA Steering Committee replacing Mr. A. Petit.

We also inform that Mr. J.A. Suarez González del Rey of CIEMAT is a new representative of Spain in the Steering Committee. He also replaces Mr. A. Velilla as Coordinator.

A heartly welcome to Messrs. Regnier and Suarez González del Rey with thanks to Messrs. Petit and Velilla for their activity.

In addition we inform that Mr. F. Mousty of CEC (Joint Research Centre of Ispra) is the new secretary of the LEU Working Group replacing Mr. V. Verdingh.

We also inform that Mr. M. Dionisi (ENEA Casaccia) is the new Italian coordinator and replaces Mr. M. Aparo. A heartly welcome to Mr. Dionisi with thanks to Mr. Aparo.

ESARDA News

12th ANNUAL ESARDA MEETING (Restricted participation)

Como (Italy), 15 - 17 May 1990

The twelfth Annual Meeting will be held at the Centro di Cultura Scientifica «Alessandro Volta», Villa Olmo, Como, Italy.
The attendance will be limited to the ESARDA Steering Committee members, coordinators, working group members and observers.

13th ANNUAL ESARDA SYMPOSIUM

Avignon (France), 14-16 May 1991

ESARDA is pleased to announce that the thirteenth Annual Symposium will be held in the historical town of Avignon in the south-east of France. This town hosted the popes in the 14th century. The symposium will be held in the restored palace of the popes.

This will be a general symposium on safeguards and nuclear material management and will have an open participation.

In Memoriam

The secretary of the ESARDA-WG, Mr. Vital Verdingh of the CEC, JRC-Geel, died on 8 August 1989, after about half a year of illness.

For more than 3 years Vital was secretary of the ESARDA-LEU WG, and for more than 6 years he was member of the ESARDA-DA WG.

As doctor in the chemistry, he was a talented and dedicated collaborator of our organization, and also a good friend; we miss him dearly.

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