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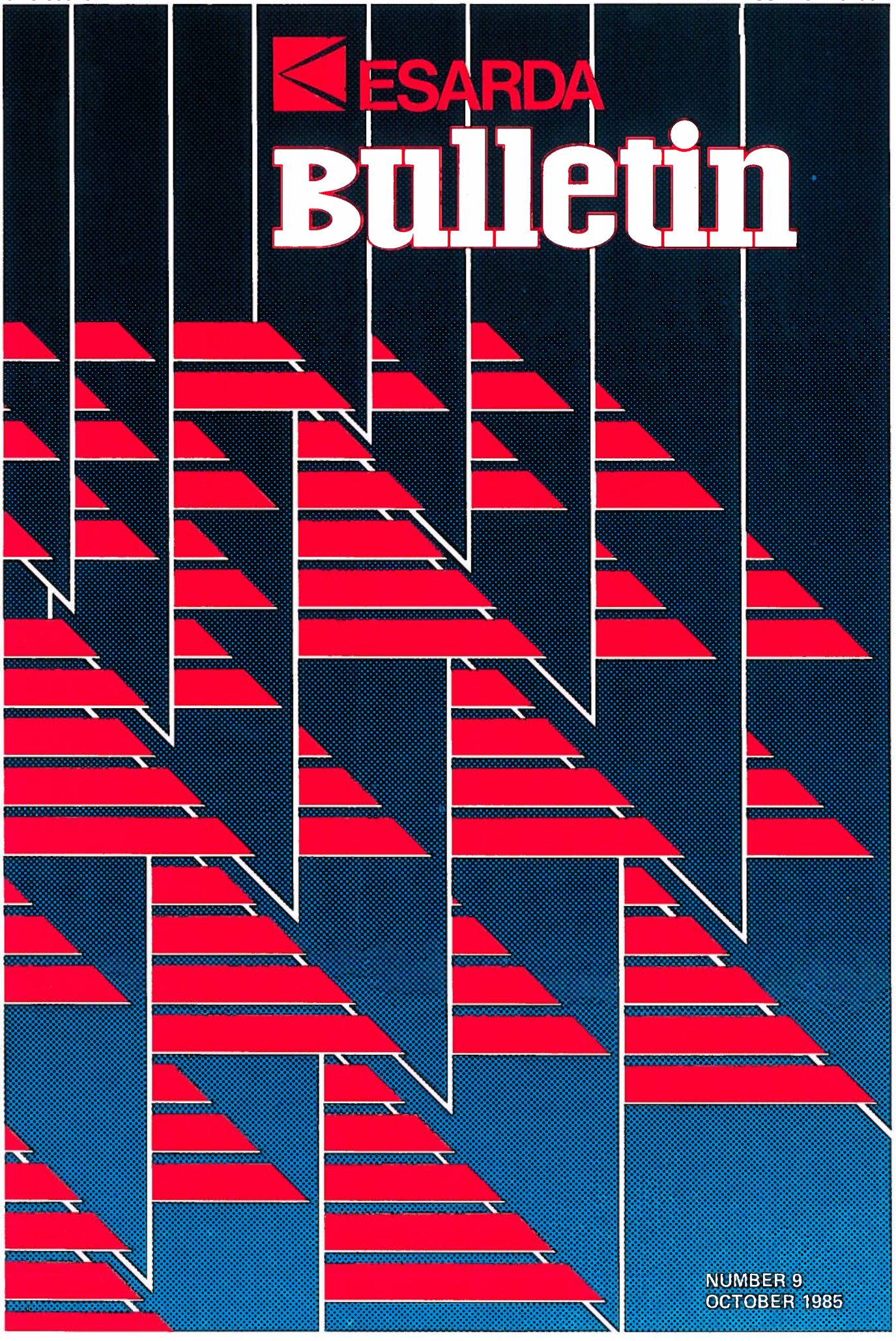


Analysis of the Liège Symposium 1985

B. Lerouge
Commissariat à l'Energie Atomique

The seventh ESARDA Symposium was held in Liège, Belgium, from 21st to 23rd May. The Congress Hall was perfectly appropriate for both presentation of communications at the podium and posters in the hall and all attendants appreciated the excellent organization of the Symposium and the Belgian welcome. The number of presentations (78) was nearly 20% less than during the last two symposia of Venice (98) and Versailles (97) and this decrease was also mirrored in the number of attendants (185 instead of 215 and 227 respectively).

If we make a more detailed analysis of the origin of the communications, we may see that those coming from the IAEA, the CEC laboratories and the U.S.A. did not change in number: 42 in total, compared to 41 and 44. They represented more than half of the presentations this year. In contrast, communications from Belgium, Denmark, Italy, FRG, France and the United Kingdom were only 22, instead of 37 in 1983 and 1984. (These statistics do not take joint presentations into account.)

I leave these data for the reader's reflection. Personally, I hope that a larger proportion of presentations will come from member countries of ESARDA in the future. One step which should help in going in that direction is the movement towards a biennial (and no longer annual) symposium. In 1986 (also because of the organization of the IAEA symposium on safeguards technology) only a restricted meeting will be organized, in Copenhagen. In 1987, a normal ESARDA symposium is expected, in a place still to be determined.

This year, the theme of the symposium was "The Impact of Advances in Data Processing on Safeguards Practices". This theme inspired few authors with general and philosophical views on international safeguards and there was no debate on such considerations. Those presented, in particular during the opening session, were all the more appreciated.

Mr. Ernemann, Belgian member of the IAEA Board of Governors presented the first paper, entitled "Reflections on Non-Proliferation and Safeguards". He expressed in a direct way views on problems related to the duality of IAEA and Euratom inspections and differences in competence between these two organizations, on differences between states inside the Community and on safeguards cost sharing.

Mr. Gmelin pursued the presentation of Euratom inspection goals made in Venice, extending them to other types of installations and discussing some practical implementation questions (synchronization of operator programmes with the inspection schedules, problems of the transport of instrumentation and safeguards irradiated samples, etc.). These questions and others are constantly discussed with the IAEA. Mr. Gmelin also stressed the importance in Euratom's view of the reverification of the Basic Technical Characteristics.

Mr. Finzi showed interactions between JRC R & D Programme and the ESARDA activities and emphasised the benefits which have been gained from this interaction, by showing examples drawn from the analysis of the activity of the seven ESARDA working groups.

Mr. von Baeckmann's paper on "The Impact of Advances in Data Processing on IAEA Safeguards" was a good preamble to the following session 2 on Nuclear Management Information Systems during which 8 papers were presented at the podium and 11 during the Poster Session.

Of the 20 papers quoted, 7 came from the U.S.A., 6 from the CEC, 4 from the FRG, 2 from the IAEA, and 1 from Denmark. General or specific needs, systems implemented in different organizations, and general trends were described. Software and hardware developments are necessary, and procedures have still to be improved. Artificial intelligence techniques may be helpful in order to render some systems usable to people who are unfamiliar with them (and also in my opinion to everyone). New materials accounting systems, such as the K2 used at Karlsruhe, also have a beneficial effect on the operating and maintenance costs which must be taken into account. Several papers stressed the point that utilization of computer systems may have a very general interest going far beyond the field of safeguards.

Session 3A opened the field of measurement techniques and was devoted to gamma measurement. The first three papers discussed techniques which are more or less oriented towards in-field routine measurements of either enrichments or plutonium mass and isotopic composition and where the measurement times were limited and equal to those times normally employed by safeguards inspectors. It is good to see that paper 3.4 discussed not only error models useful in understanding the overall uncertainties but also employed the U_3O_8 NBS-CBNM-ESARDA reference samples to identify these error estimates.

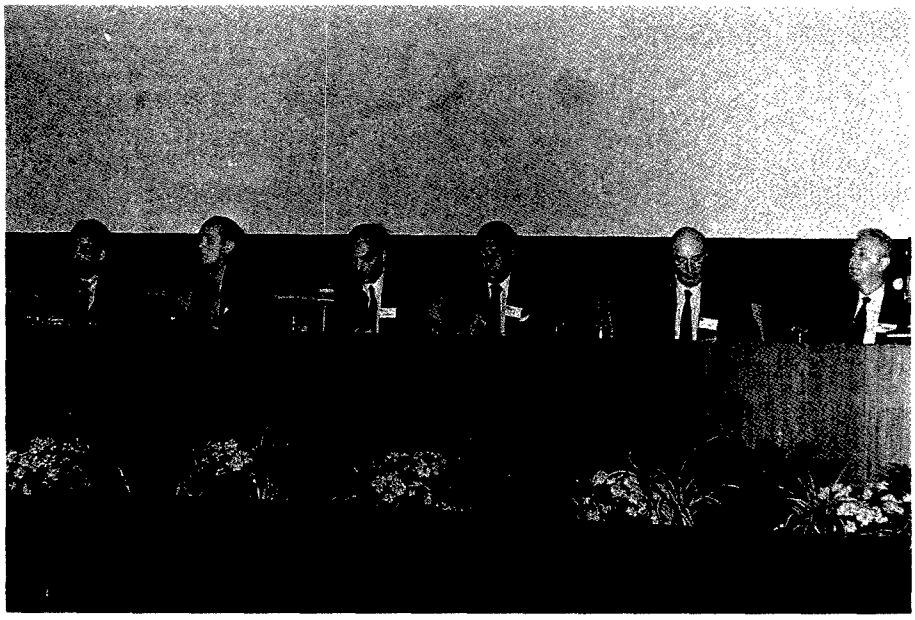
Session 3B was devoted to neutron measurement techniques. Neutron measurements (both active and passive) are commonly used by many laboratories mainly for bulk material assay because of the good penetrability of neutrons. Some advantages and disadvantages are already apparent; neutron detectors can be relatively simple and rugged; on the other hand, interpretation of results can be somewhat difficult due to sample self-multiplication and self-absorption.

The PHONID device (3.5) has been intensively tested on a very large variety of uranium and plutonium samples. This is a good example of how to develop an instrument systematically.

The neutron die-away technique (3.6) is a quite interesting and challenging method which still requires improvements, but which is capable of providing unambiguous results when applied, for example, to the determination of small quantities of fissile materials as in wastes.

Papers 3.7 and 3.8 on passive neutron coincidence counting reflect a well known technique which still requires improvement of the existing data correction criteria as well as an assessment of the overall error. Both papers contribute significantly to fulfilling these needs.

Five papers were presented in Session 3C "Other measurement techniques". They dealt with D_2O assay using ultrasonics (measurement uncertainties and tamper resistance were given special attention), studies of the differential pressure bubbling technique of liquid levels and densities in tanks, at the input of nuclear reprocessing plants (the result shows that a relative measurement precision of 2×10^{-3} can be obtained), demonstration experiments with the ION-1 FORK n,γ-measurement device performed on LWR fuel assemblies,



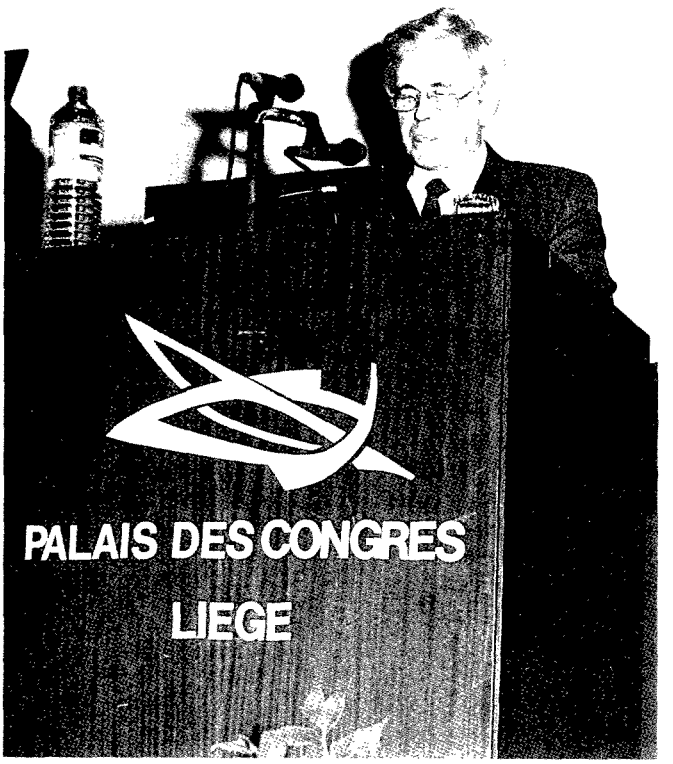
The opening of the Symposium by Mr. B. Lerouge. From left to right: W. Gmelin, A. Ernemann, B. Lerouge, J. Ley, S. Finzi, L. Stanchi

Prof. L. Stanchi, scientific secretary of ESARDA Symposium, introducing Mr. B. Lerouge, present chairman of ESARDA

Mr. A. Ernemann, Belgian Ambassador to Austria and Belgian member of the Board of Governors of IAEA giving his opening speech

A view of the congress hall

Dr. S. Finzi, Commission of the European Communities, DG XII, Brussels



Dr. A. von Baeckmann, IAEA, Vienna

Dr. W. Gmelin, CEC Safeguards Directorate, Luxembourg

The organizing secretariat

experimental studies on deposits in the header piping of a gas centrifuge plant (long term deposition growths and isotopic exchange were studied), adaptive assay system and its application to nuclear safeguards.

The papers of session 4A emphasized the amount of research which still needs to be done to identify global indicators for the quality control of near real time accountancy systems. Recognizing the vast amount of data that is required to support such a system, it is clear that operator data must form the basis for calculation of short-term material balances. It has not yet been identified how the inspector data can be used efficiently in conjunction with those provided by the operator in the near real time context. Development in this field could make a positive contribution to future methods of data capture and analysis, and this in turn has implications for safeguards systems design.

The four papers in session 4B were related to the quality of measurements. Presentations were made of pragmatic testing and acceptance guidelines for new safeguards instrumentation, of the final results of the IDA-80 programme performed under the ESARDA's auspices and with the support of the IAEA, of quality control of balances of Springfields Nuclear Power Development Laboratories and of the various parameters affecting the quality of the reprocessing input measurements.

Only three papers were presented in session 5 related to containment and surveillance. The first describes two different seals under development and tests (an optical fibre bundle seal and a thermal gradient system well adapted to heat emitter surveillance). The second presented a laser surveillance system (LASSY) tested in Vienna and the third displayed experience gained from the use of twin film camera units.

Two papers on Safeguards Performance Evaluation were presented in the last session. The first, an analysis on diversion detection capability and treatment of anomalies comparing conventional materials accounting, containment and surveillance and n.r.t.a. concluded that n.r.t.a. has a higher capability than the other two systems for the detection, the follow-up and the resolution of anomalies. The second presented a structured approach for the assessment of inspection activities carried out compared to those activities which had been formally agreed as necessary and sufficient elements in the Facility Attachment. The use of the computer program which has been developed was demonstrated in a poster session.

In parallel to the 6 oral sessions the 4 poster sessions on nuclear material management information systems, measurement techniques, error propagation and quality assurance, containment and surveillance, were as popular and efficient as the oral sessions, and were particularly useful for producing the maximum impact on the interested specialists in allowing direct dialogue with the authors. This procedure permitting non-overlapping time with oral sessions, should be continued.

Performance Assessment Method for NDA A Challenge for PERLA

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Introduction

For some years it has been pointed out and it can increasingly be seen, on the basis of experience built up over the years, that the main problem in transferring safeguards instruments and methods from the developing laboratory to the field, is that of producing reliable safeguards instruments.

It is in fact evident that most of the techniques, when applied, have a different (lower) accuracy in the field than they do when used in the laboratory.

It has only recently been realized that this is due to objective reasons rather than to, generally speaking, incorrect application of the method.

The lack of extensive structured measurement data bases and appropriate error models delayed the process of understanding.

Some objective reasons accounting for the different levels of accuracy are:

1. **Lack of time** to perform measurements, which obliges the inspectors to reduce the counting time. In the absence of an analytical model which quantifies the importance of the statistical counting variance on the overall uncertainty, subjective judgement could lead to contradictory results : sometimes the counting time reduction leads to an obvious degradation of the results, sometimes not.
   This result is certainly qualitatively true because, as is shown in the next chapters, the same device in different experimental situations can have a different sensitivity with respect to the counting time uncertainty propagation.
2. **Extreme variability of items to be measured**
   Again an error model which assigns the same overall error to almost all different combinations of instrument and measured items cannot represent the complexity of the error phenomenology which generally exists when measuring a complex population. To give an example, in an inventory for some families the calibration curve component (long term systematic error) may be dominant. In other families (small items) the counting uncertainty (random) may be the largest component; in other families geometrical or matrix effects (never taken into account in the error models) may be the strongest source of uncertainty.
   If the model does not essentially take into account this complexity of the real world, it is destined to fail.
3. **Problems in measuring large quantities**
   Again in measuring large quantities of fissile material a mismatch may exist between models which have been designed in the laboratory (generally on small samples) and the real practice where more and more bulk samples are encountered in the fuel cycle.
   Corrections are there applied to the results (e.g. NCC) or to the geometrical set up (HRGS), but the principal obstacle frequently resides in the fact that most physical models are more or less explicitly **point source** models. And 3 kg of Pu is not a point source for many of the current γ or neutron experimental setups.

It also becomes more and more evident that the automation of instruments will solve only one aspect (very important) of the problem : i.e. the part which is connected with a correct and easy and rational collection, recording and transmission of data, following agreed inspection procedures. This aspect, together with the setting up of tailored structured data banks will certainly also help in understanding where the measuring problems are.

The fundamental reason for field-laboratory discrepancies resides however in the fact that frequently the physico-statistical error models were lacking or were borrowed from applications which were different from the ones used in safeguards.

From what has been said so far one may see the need to have specific error models for any safeguards techniques applied in the field. The error model must explicitly contain all major error sources of that particular instrument when applied to any item family (or stratum). Furthermore, the model must be connected with the final use of the data, which is the comparison with operator declarations (which is quite a unique situation in the scientific world).

The set up, testing and updating of these field models are amongst the main objectives of the PERLA laboratory which is being set up at the JRC-Ispra.

The general layout of the PERLA laboratory has been given in /1/. Its main objectives have been identified as :

1. Performance tests of instruments and methods for NDA, DA, C/S
2. Recalibration of instruments
3. Training
4. New developments.

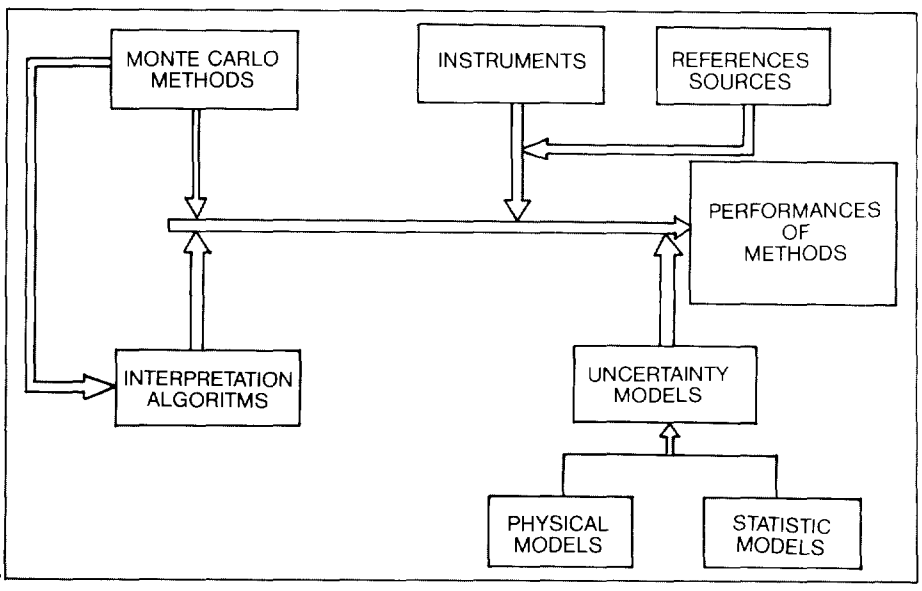

Fig. 1 - PERLA: structure of the calibration laboratory

It is the aim of this paper to describe in detail the logical approach and the actions to be taken to fulfil the objectives 1 and 2 mainly for NDA, leaving topics 3 and 4 and other disciplines for other presentations.

A very schematic logical layout of the main tools to be provided to PERLA in order to assess the experimental uncertainties is given in Fig. 1.

In the following sections of this paper more details will be given on why and how these tools will be used. Such field topics as :
- error modelling
- sensitivity studies
- fissile material procurement and characterization
- Montecarlo methods

will be dealt with.

Summarizing in a few words what was said in the introduction and trying to illustrate in one sentence PERLA's main objective, one might say that PERLA is a research laboratory where research will be carried out on the measurement uncertainty, i.e. where the target of the research is not only to provide accurate estimates of some physical parameters relevant to safeguards, but also to provide accurate error estimates of such parameters when measured in (near) real field conditions.

This kind of research and experiments will be called PERLA Type Experiments (PTE).

Measurement Error Modelling

The first step in a PERLA Type Experiment (PTE) is to derive an error model. As was said in the introduction and reported in /2,3,4/, the JRC approach to error modelling is to derive specific models for any technique or instrument: the models must take into account the physical and statistical behaviour of that particular instrument and the measurement procedures employed.

The same model will be used (and is designed for) in the further accountancy steps, i.e. operator-inspector differences, D, measurement quality control, etc.

Now, starting from the relationship connecting the raw measured data (e.g. the gross area count rates) to the quantity evaluated, it is necessary to define the analytical model describing the overall uncertainty of the parameter.

One may illustrate this principle on the basis of an example. Schematically one can say that, as an example, the isotopic ratio $^{239}Pu/^{241}Pu$ is evaluated as a function of a number of measured parameters a, b, c,... as

$$^{239}Pu / ^{241}Pu = f(a,b,c,...) \qquad (1)$$

Its overall uncertainty E(9/1) may be (again schematically) described as follows:

$$E^2(9/1) = (g_a \epsilon_a)^2 + (g_b \epsilon_b)^2 + g_c \epsilon_c)^2 + ... + g_a g_b \, cov(a,b) + g_b g_c \, cov(b,c) + g_a g_c \, cov(a,c) + ...$$

where:

ϵ_i are the individual uncertainties of the parameters a,b,c,...

g_i are the transmission factors of the individual uncertainties derived by Taylor expansion of (1) and the terms with covariances take into account the correlated components of (1). Typically the use of calibration curves (e.g. intrinsic calibration for HRGS) gives rise to correlations (covariance terms) amongst measured parameters.

Based on the above expression one may list the objectives of a PERLA Type Experiment (PTE) as:

1. Deriving the value of E_j for a variety of techniques and instruments, based on an error model specifically designed for the instruments.
2. Deriving the values of individual ϵ_{ij} if they are not defined theoretically; for example a counting has a simple theoretical variance (when poissonian) and has only to be checked, but calibration parameter variances must be determined on a separate set of measurements.

But then, after the E_j and ϵ_{ij} values have been modelled and assessed, a phase of thorough checking of the models will take place. This phase should consider the following points:

a. Check with specific experiments that E_j and ϵ_{ij} have been correctly modelled and evaluated in many typical experimental conditions (see next section);
b. Discover the most influential parameters in the error propagation structure and indicate the work to be done to minimize their influence;
c. Work out sensitivity tables to be used to predict the measurement error in conditions which are slightly different from that under study (e.g., if the inspector has to measure one new family with a slightly different α -ratio or isotopic composition);
d. Verify explicit or implicit assumptions or approximations used in the models or in the physical algorithms (see next section);
e. Verify the validity of the models used with the aid of very well characterized fissile materials (and sometimes of international standards) used in "ad hoc" experiments.

The discussion, with some examples, of the points just mentioned will follow in the following sections.

Checking the Models

It has been mentioned in the previous section that one of the most important tasks of the PERLA laboratory and PTEs will be to check the models and the assumed or evaluated error component values.

Two typical PTEs have been reported in /4,5,6/ and are only summarized here.

Uranium enrichment measurements

In refs. /4,5/ an error propagation model has been developed for ^{235}U abundance measurements in LEU powders and pellets. The uncertainty model has been applied in two different experiments : in high accuracy laboratory measurement campaign on U_3O_8 NBS-CBNM-ESARDA reference materials /7/ (see Fig. 2), and then in validating the results obtained in field conditions by the JRC enrichment meter. The conclusions reached in the experiments were the following:

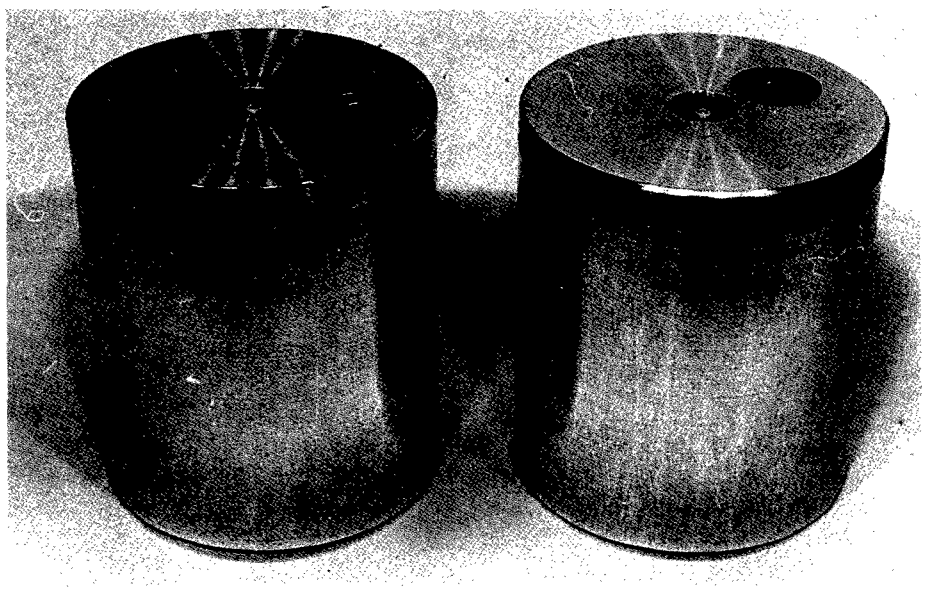

Fig. 2 - NBS-CBNM U_3O_8 reference sample

- As far as high precision laboratory measurements of ^{235}U abundance are concerned, the results given in the paper show that the technique has now reached a very high accuracy, provided that high level reference materials are available. Since the accuracy in net areas measurements can reach the order of 0.1% for LEU powder or pellet samples, high accuracy reference materials (uncertainty not higher than 0.05%) are required.
- Counting times ranging from 1 to 5 hours are required to reach the above uncertainty, for ^{235}U enrichments going from 4.5% down to 2.5% respectively.
- The predicted uncertainty evaluated by the error model presented is in very good agreement with the evaluated SD of the results, in any counting situation.
- Concerning the performance of the JRC Enrichment Meter, ref. /5/ shows that the instrument is working properly and that in many different conditions of calibration, enrichment, counting times, etc., the statistical parameters are always consistent with the theoretical ones, as derived again from the above mentioned error propagation model. This is finally the main requirement for an instrument: it gives the performance which is theoretically to be expected from it under all measuring conditions.

PHONID

PHONID is a photoneutron active interrogation device developed at Ispra for ^{235}U content determination in unknown samples.

Some examples of the device have been employed for many years in monitoring in field conditions a large variety of families going from U_3O_8 powders, metals, U-Al platelets, U-Th particles, from kgs size samples down to grams of waste. Both LEU and HEU samples have been successfully monitored and in the last measuring campaign /6/ PuO_2 cans up to 2.5 kg were measured.

Following the scheme of a PTE a new error model has been prepared and old and new measurement campaigns have been interpreted. The conclusions were as follows:

- PHONID measures HEU und LEU samples from grams to kgs size with a typical uncertainty of 1 - 2%. The error model correctly reproduces the overall measurement uncertainty.
- Some advantages may be presented by active/passive plutonium interrogation with PHONID with respect to passive interrogation alone. In particular, a lower error transmission to the global uncertainty of Pu total is to be expected from active interrogation, because the $^{239}Pu_{eq}$ uncertainty is transmitted instead of that of $^{240}Pu_{eq}$ (see below).

The two experiments described above should now be repeated on a larger variety of fissile materials when PERLA laboratories (or Pre-PERLA) are operative to confirm that the models adopted are correct.

We will again treat in the following the two experiments mentioned above to give examples of the logical approach used in PTEs.

Another important aspect of modelling is shown in points b) and c) mentioned in the previous section and must be taken up again now here, with more details : they refer to the importance of performing parametric error studies in order to

b. discover the most influential error components
c. make sensitivity studies.

Concerning the enrichment measurements the results of some similar parametric studies are shown in Table I and Fig. 3.
A ^{235}U abundance measurement consists in the determination of the net area A_x under the 185.7 keV peak of the unknown sample and in the net area determinations (A_1, A_2) of two (or more) reference standards of known enrichment (E_1, E_2). Approximating the measurement Standard Deviation (SD) with the variance square root and assuming, for example, a 0.1% SD on all the parameters, we obtain (being E_x = 2.9843%, E_1 = 1.9658%, E_2 = 4.5167%) the results shown in Table I.

As can be seen from the table, the largest contribution to the overall uncertainty of E_x(assumed as SDE_x) for the given assumptions comes from the determination of A_x. But an appreciable contribution also comes from the E_1 and E_2 parameters.

TABLE I Variance components in E_x determination

Source of uncertainty		Variance component
from	A_x	7.95 - 6
	A_1	2.02 - 6
	A_2	2.4 - 6
	E_1	2.2 - 6
	E_2	2.2 - 6
Var E_x =		16.97 - 6
$SD_{E_x} = \sqrt{Var\ E_x}$ =		4.1 - 3 = 0.14%

It has been shown in /5/, based on such parametric studies, that the two standard enrichments must be known with a factor at least 2-3 better than the net area A_x, if one wants to keep their contribution to the overall uncertainty negligible.

Similar studies were then conducted on the expression giving the net area: an errror sensitivity study was made giving the results summarized in Fig. 3, where it is shown that assuming the theoretical random uncertainty as the only source of variance, one can obtain an uncertainty of 0.1% on A_{net} for a 4.5% enrichment sample with about 1 hour of counting.

The same level of uncertainty for a depleted (0.32%) sample can be obtained in about 100 hours counting /5/.

A similar analysis has been carried out for the PHONID device: some results are given in Figs. 4 and 5 where the error components and the overall uncertainty are plotted versus the ^{235}U mass, as resulting from a parametrization of the uncertainty relationship.

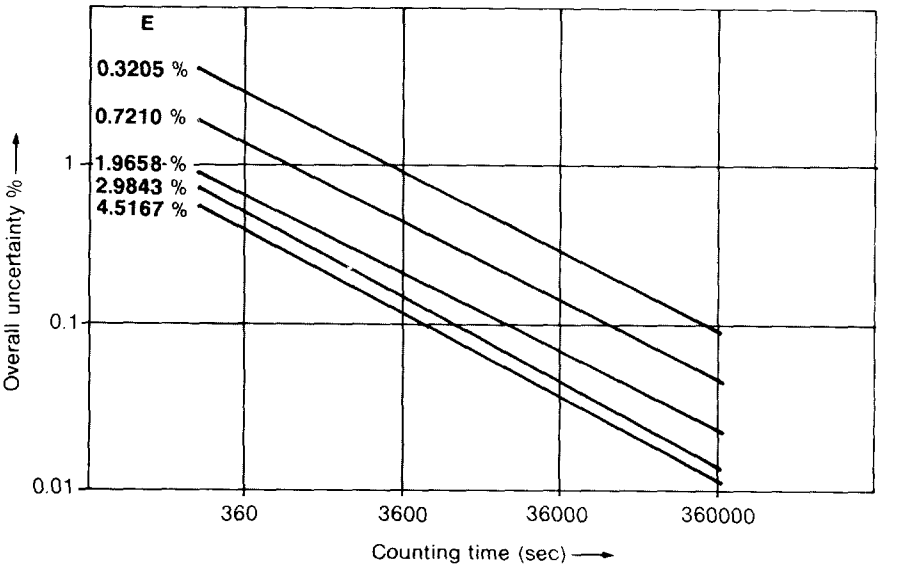

Fig. 3 - Uranium Enrichment: predicted A_{net} uncertainty as a function of counting time

One can see the different role played by the counting statistics and the calibration curve error component at different ^{235}U weight and different counting times. The effect of such a curve set is to quantify the obvious consideration that in certain experimental situations (low masses) the counting statistics can be a source of uncertainty if insufficiently long counting times are employed.

Similar curves or tables (like Table I) are extremely useful and should be prepared, based on the error propagation models, for other experimental situations and for other techniques and/or instruments.

These tables should then be checked with suitable experimental measurements.

Another very interesting outcome of these parametric studies was found in /6/ and concerned the overall Pu_{tot} uncertainty. It is well known that the total plutonium content of unknown samples is frequently determined on the basis of two complementary measurements: an isotopic composition determination and a $^{240}Pu_{eq}$ measurement. A preliminary model of such a procedure has been developed in /6/ and the overall typical uncertainty derived. The same procedure has been followed for active interrogation where the so called $^{239}Pu_{eq}$ is determined with PHONID.

The comparison showed two interesting features:

1. The overall Pu_{tot} uncertainty in passive determination is typically of the order of 5%, while from active interrogation one can have a total plutonium content with almost 2.5% uncertainty.
2. The error structure in the two approaches is quite different: in the passive determination (see Fig. 6) the uncertainty of the ^{240}Pu abundance dominates the overall Pu_{tot} uncertainty and the neutron measurement uncertainty (assumed as 2%) is negligible (the ^{242}Pu abundance uncertainty only becomes relevant at higher burnup).
   In the active interrogation on the other hand (Fig. 7) the uncertainty in the neutron measurement (also assumed as 2%) represents the most important contribution. Only if the active neutron detection uncertainty is lowered to 1% (which is not an unrealistic task) does the isotopic composition uncertainty begin to be noticeable.

The above considerations are summarized in Table II and III. In Table III the components of the overall variances for active and passive emissions are given with the assumptions made in Table II.

Before leaving this section it is worth mentioning point d) as referred to in the previous section, i.e.:

verify explicit or implicit assumptions or approximations of the model

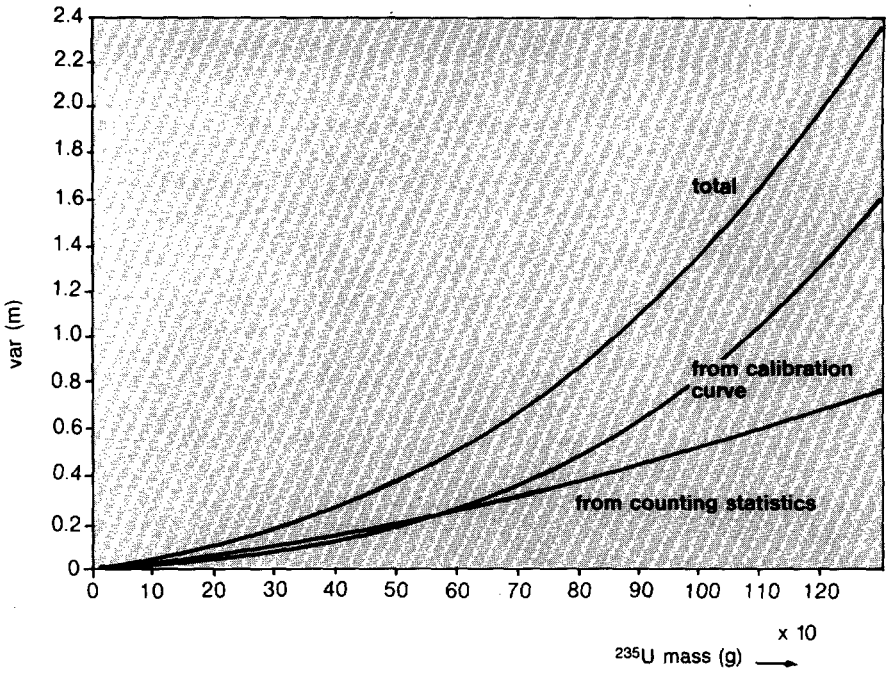

Fig. 4 - PHONID error structure with 100 sec counting time

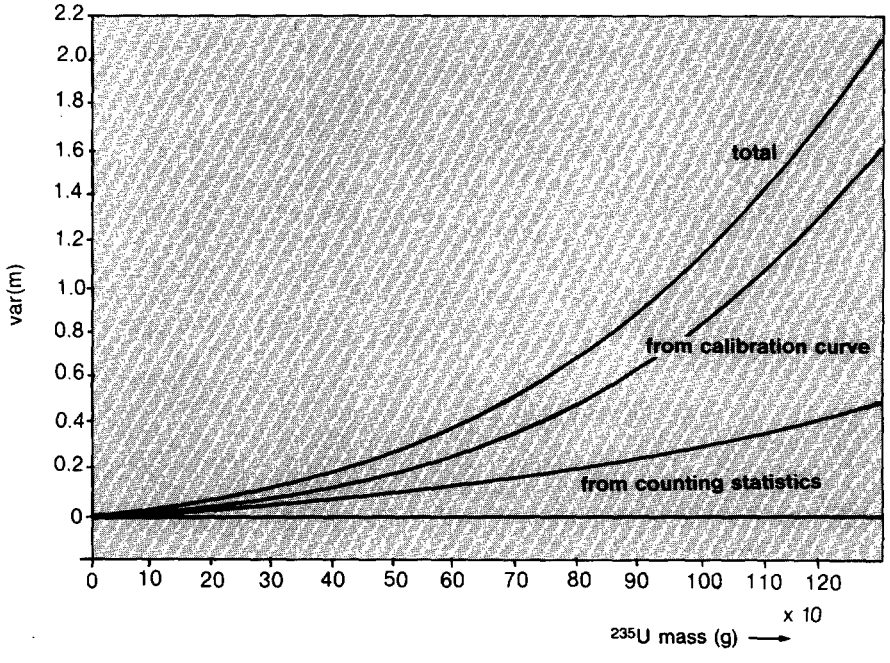

Fig. 5 - PHONID error structure with 400 sec counting time

This is a very important point because frequently assumptions are made implictly (i.e. without referring openly to them), but they remain at the basis of the technique. This is the case for instance with the assumption that certain events are Poisson distributed, which is not always so obvious (at least in principle) especially when correlated events are monitored.

Another example of a more or less implicit assumption is frequently found in measuring large U or Pu samples : as already said almost all theories assume implicitly that the sample is a point sample, which is certainly not true for the majority of detection heads currently used.

As an example of the explicit type of assumption we refer to the enrichment meter model, where we assumed that net area and background were not correlated: an experimental test was then carried out to verify this assumption. The exercise con-

Table II: Assumed uncertainties for Pu_{tot} error analysis

parameter	uncertainty	comments
e_8	5%	isotopic composition uncertainty from HRGS
e_9	1%	
e_0	5%	
e_1	1%	
e_2	10%	
$^{240}Pu_{eq}$	2%	typical uncertainty from passive or net active measurements
$^{239}Pu_{eq}$	2%	

sisted in determining the net area with two different background subtraction algorithms: a linear subtraction and a stepwise one. In both cases a correlation coefficient of the background and the net areas was determined.

The results /5,8/ of the measurements can be summarized as follows:

1) The two algorithms provide net area results which are slightly different. A systematic difference of around 0.5% was found as compared to a statistical uncertainty of 0.1%.
2) The consequent variation of the measured enrichment is negligible in the limit of experimental uncertainty (0.1-0.2%).
3) The correlation between net area and background is shown in Fig. 8 in terms of the correlation coefficient:

   $r = s_{xy}/s_x s_y$

   where s_x, s_y, x_{xy} are the standard deviations of x, y and their covariance, $(-1 \leqslant r \leqslant 1)$ respectively.
   It can be seen from Fig. 8 that there is not a strong correlation between the two parameters. The correlation is slightly stronger in the case of the linear subtraction. There is no obvious dependence of the correlation index on the enrichment: the points at 2.9843% enrichment look slightly anomalous without any obvious explanation.

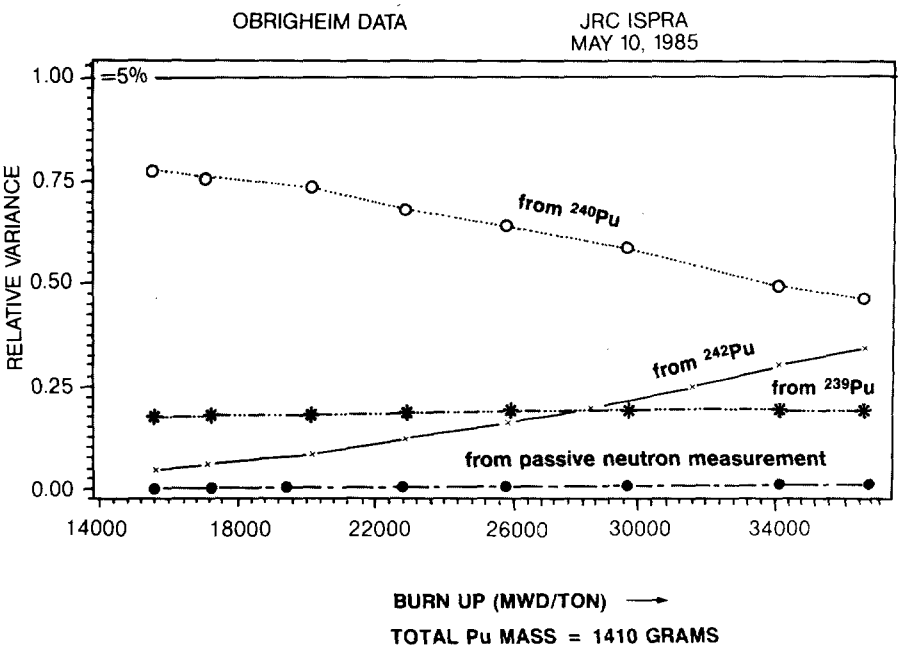

Fig. 6 - Passive assay: plutonium error propagation analysis

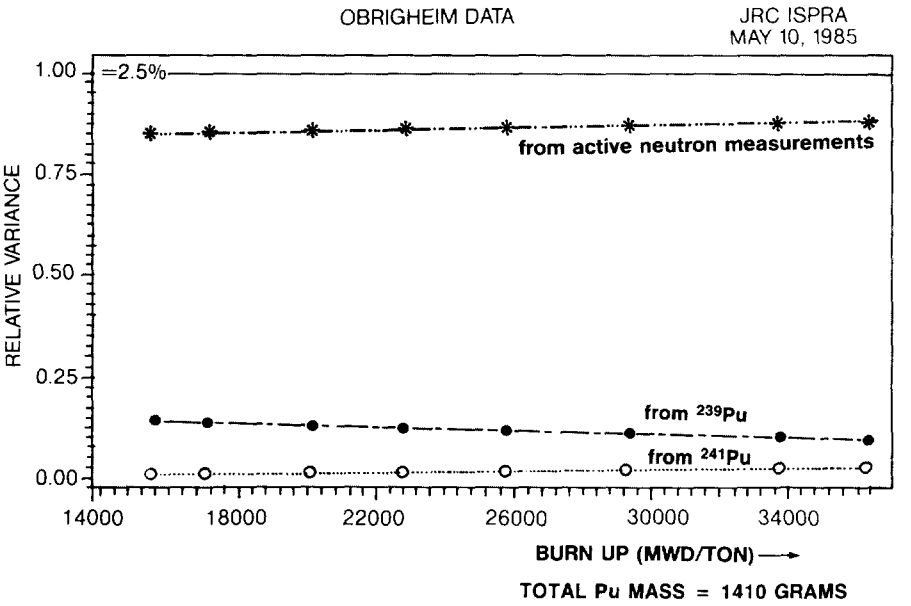

Fig. 7 - Active assay: plutonium error propagation analysis

Application of Monte Carlo Codes

Two Monte Carlo codes are presently running at JRC-Ispra for safeguards purposes: TIMOC /9/ and MERCURE /10/. TIMOC is employed for neutron detection head studies. MERCURE is currently employed for γ-transport studies. The nuclear data libraries of both codes have been recently modified and updated to adapt their use to the purposes required in safeguards.

Their use in safeguards has different aspects:

1. They are employed primarily in the design of new (or modified) instruments. This topic refers mainly to the fourth aim of PERLA (see introduction): **new developments,** and will not be treated here.
2. The second use of the codes is to calculate correction terms of algorithms, which are difficult to evaluate or measure: a typical example is the calculation of the multiplication factor of bulk Pu samples. The calculated values are then introduced directly into accurate algorithms or are used to check approximate corrections used in in-field algorithms. This aspect is not covered here in more detail either /11/.
3. Finally, Monte Carlo codes will be used for PERLA purposes for analysing those sources of variability (i.e. source of uncertainty) which are not accounted for in the uncertainty analytical expression. A typical example is represented by the effect of matrix and geometry on neutron counting (both passive and active). These characteristics of the sample matrix (specially when containing moderating materials) and of the geometry have the effect of modifying the neutron spectrum in the sample, with a consequent variation of the average neutron cross sections. In other words the neutron output (neutron/g. fissile) is

TABLE III Plutonium error propagation analysis

COMPONENTS OF OVERALL VARIANCES IN PASSIVE AND ACTIVE EMISSIONS UNDER THE ASSUMPTIONS OF TABLE II

BURNUP	ASSAY	f_{240eq} · var ($240Pu_{eq}$)	f_{239eq} · var ($239Pu_{eq}$)	f_8 · var (e_8)	f_9 · var (e_9)	f_0 · var (e_0)	f_1 · var (e_1)	f_2 · var (e_2)	SD (%)
5000 MWD/ton	passive	$4 \cdot 10^{-4}$		$5.42 \cdot 10^{-7}$		$2.34 \cdot 10^{-3}$		$3.18 \cdot 10^{-6}$	5.24
	active		$4 \cdot 10^{-4}$		$9.71 \cdot 10^{-5}$		$2.13 \cdot 10^{-8}$		2.23
36260 MWd/ton	passive	$4 \cdot 10^{-4}$		$2.81 \cdot 0^{-5}$		$9.78 \cdot 10^{-4}$		$7.21 \cdot 10^{-4}$	4.61
	active		$4 \cdot 10^{-4}$		$4.47 \cdot 10^{-5}$		$1.10 \cdot 10^{-5}$		2.13

different if the geometry and/or the matrix vary from item to item.

Normally these effects are accounted for in the calibration curve.

Frequently, however, it is not possible or practical to have calibration curves representing all the possible families and items in one inventory. For that reason frequently more "close" families are represented with one calibration curve and this increases the response variability. This source of variability is implicitly accounted for in the calibration curve parameter uncertainty (var a, var b in the case of a power function $N = a M^b$ as for instance in the PHONID case).

But still the situation is not always satisfactory, because frequently the inspector is confronted with items which are difficult to assign to one or another calibration curve.

It is then important to have quantitative evaluations of the variation of the parameter under measurement (e.g. $^{240}Pu_{eq}$, when the geometry varies or the moderator content varies.

These sensitivity studies cannot give an immediate evaluation of the overall uncertainty variation (remember that these parameters are only implicitly accounted for through the calibration curve). It is, however, important to have such parametric studies, for instance to judge whether a new calibration curve is necessary for a new family (e.g. cans with a different geometry or rodlets with a different isotopic composition).

An example of this type application of the TIMOC code is given in /6/: the active neutron emission has been evaluated by TIMOC for different isotopic compositions (burnup values from 5 GWd/ton up to 35 GWd/ton) obtaining the results shown in Fig. 9. One can see how close the active curves are when compared with passive curves, as evaluated from nuclear data and shown in Fig. 10. This is again a possible advantage of active interrogation since it gives the possibility of using a smaller number of calibration curves.

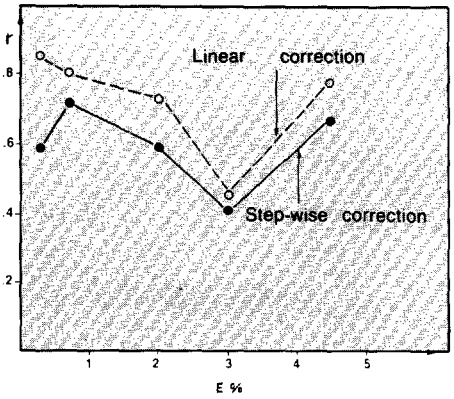

Fig. 8 - Uranium Enrichment: correlation between net area and background

A clearer picture of the burnup dependence of active and passive emission is shown in Fig. 11, where, for a Pu mass of 1410 g, the normalized emissions are shown as a function of the burnup.

Fissile Material Procurement and Characterization

One of the most important points in the PERLA laboratory set up concerns the procurement of fissile materials and their characterization. The whole concept of testing the models of calibration and recalibration finally is based on the condition of having a well characterized PERLA standard. An extensive description of the fissile material for PERLA is given in /1/. We give a summary here of the main items.

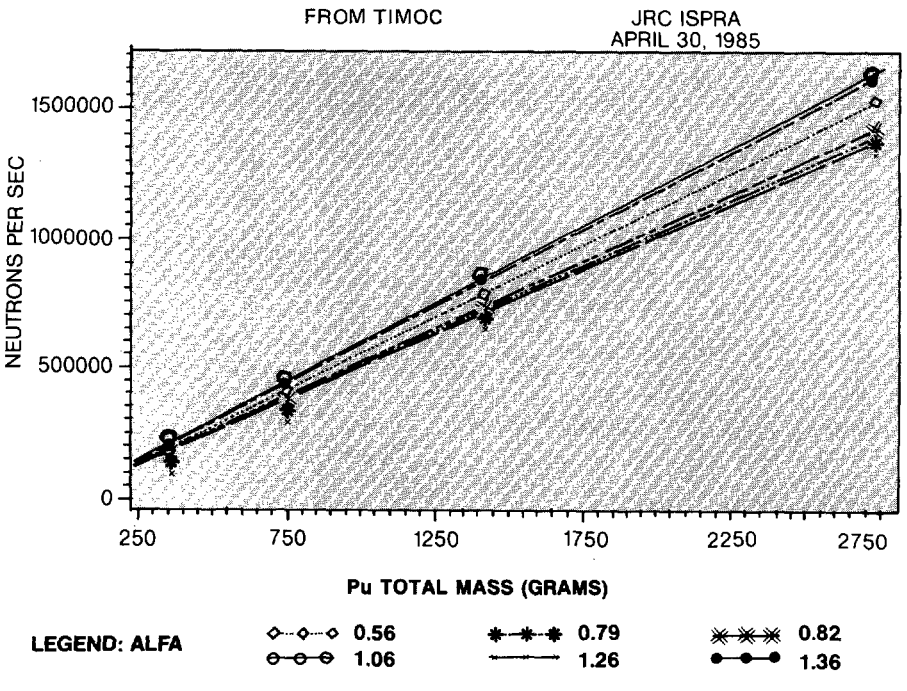

Fig. 9 - Pu active emission at different isotopic compositions vs Pu mass

PuO_2 or MOX	powders
highly enriched U	powders plates, pellets pebbles
low enriched U	powders pellets pins assembly (mock-up)
spent fuels (for the storage pool)	MTR BWR PWR

It may be worthwhile to develop in slightly more detail the aspect of the role and the importance of the procurement and characterization of fissile materials for PERLA.

Procurement: The variety of plant specific materials procured will ensure the role of PERLA as a bridge towards the field application; this is one of the main reasons for the establishment of the laboratory. In effect we are talking about a near real field laboratory, because obviously we cannot cover the whole spectrum of plant production, but we try to cover the most important items encountered in fabrication plants.

Concerning the spent fuels, the pool storage existing in ESSOR will allow us to carry out NDA measurements of spent fuels of LWR and MTR type and to assess instruments and methods in this important field of safeguards.

Regarding reprocessing plants the JRC management is concerned to cover that important field also. Some new activities are now being launched in connection with the PETRA facility in construction at Ispra.

PETRA is a laboratory-scale reprocessing facility for waste management studies, on which some safeguards activities will be carried out.

Characterization of fissile materials: All the points developed in the preceding sections concerning the assessment of uncertainties of NDA methods are based on one common assumption: the fissile materials used as reference must be known with an uncertainty that must be significantly lower than that of the NDA methods under study.
If, e.g., the uncertainty under assessment is that of HRGS for Pu isotopic composition (say 0.5% for ^{239}Pu) the fissile materials used as a reference in the experiments must be known at worst with 0.2% in order not to introduce an error component in the overall NDA experiment.

If the NDA method under study is very precise and accurate (e.g., calorimetry) the required knowledge of the fissile materials used must be substantially better (say less than 0.1%), compatible with technical and economical restrictions. This, in any case, requires a very careful characterization of the individual fissile material families, tailored to the specific item (or items) and to their future foreseen uses.

A detailed procurement scheme will be prepared for any fissile material family with the following main objectives (see also /12,13/):

- identify the uncertainty level required by any fissile material type based on its future NDA use;
- describe the preparation and characterization procedures which can guarantee this uncertainty level;
- prepare statistical sampling and homogeneization procedures for DA analysis;
- prepare an analytical scheme for DA analysis;
- apply statistical analysis to the results to work out the final overall uncertainties of the fissile material content of all families and single items;
- guarantee traceability to primary international standards.

In effect it is worth mentioning that the fissile

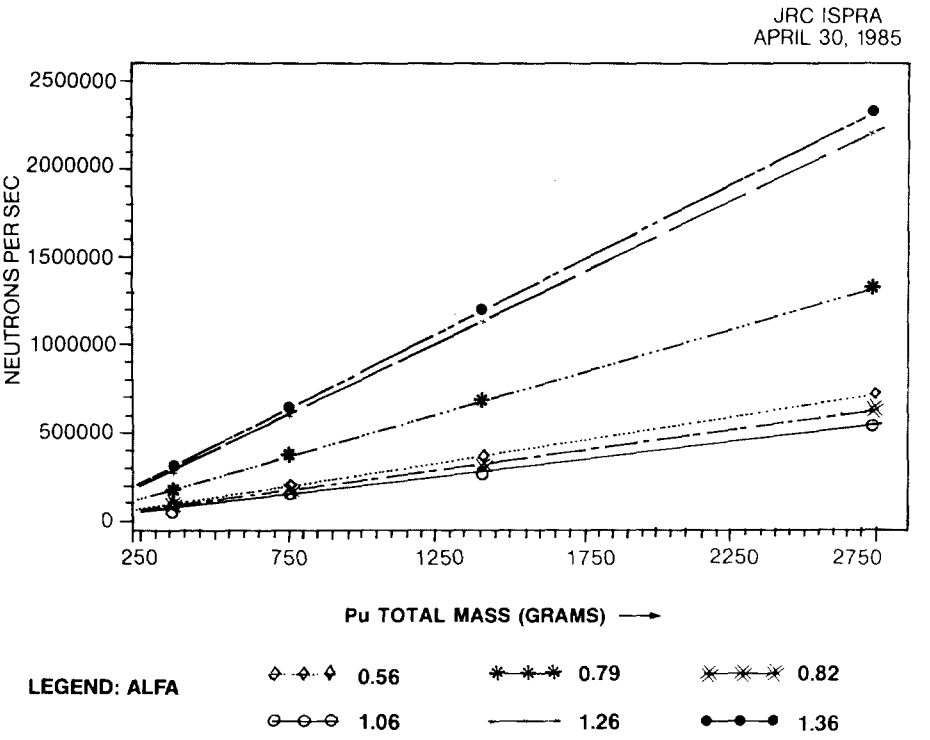

Fig. 10 - Pu passive emission at different isotopic compositions vs Pu mass

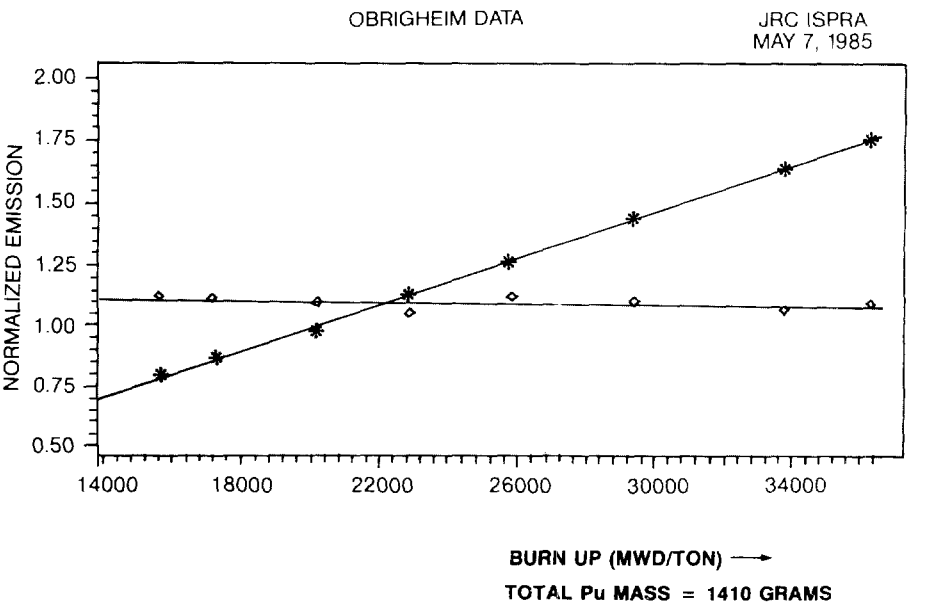

Fig. 11 - Plutonium emission analysis: normalized passive and active emissions as functions of burnup

materials used in PERLA will not be (in general) certified standards. Apart from a few exceptions (e.g., U_3O_8 powder standards certified by NBS-CBNM under ESARDA NDA working group management /7/), the majority of the fissile materials used in PERLA will be only very well characterized samples but not certified ones being rather internal working standards; for this reason they are called PERLA Standards (PS).

Some external laboratories will be asked to participate in the various steps of the preparation of the fissile materials, but no formal certification will be issued.

A detailed procurement scheme, following the lines just described, has already been prepared for a plutonium bearing fuel family. A family is a set of standards covering, for instance, a range from PuO_2 powder through MOX powder, pellets down to rods and possibly a fuel assembly mock-up, all derived from the same original PuO_2 production batch.

Conclusions

A functional layout of the PERLA laboratory has been presented in the paper showing the hardware-software tools which will be employed in a typical PERLA Type Experiment (PTE) conducted to fulfil objective 1 of PERLA.

A definition of PTE has been tried as being research on the measurement uncertainty of instruments and methods, mainly applied in (near) real field conditions.

A schematic description of a PTE has been given as follows:

- set up the error model by Taylor expansion of the expressions leading from raw measured data to the quantity required,
- identify all significant error sources and quantify them,
- derive the overall uncertainty of the method or instrument under evaluation in different experimental conditions,
- parametrize with the possible help of Monte Carlo codes to cover large ranges of application (preparation of sensitivity tables and curves),
- discover the most important parameters in the error propagation structure and describe the working plan to minimize their influence in detail,
- check the model with tailored experiences using well-characterized fissile materials (PERLA standard) and validate the assumptions made in describing the model.

Two examples of PTE have been given in the paper, namely for ^{235}Pu abundance measurements and for PHONID active measurements on ^{235}U and Pu determination. The examples showed the validity of the approach chosen.

References

/1/ M. CUYPERS et al. — PERLA, safeguards performance, calibration and training labratory at JRC-Ispra. Proc. 6th ESARDA Annual Symp., Venice (Italy), 14-18 May 1984

/2/ M. CUYPERS, G. DE GRANDI, M. FRANKLIN, K. MÜLLER, S. GUARDINI, M. MONTAGNI — An integrated scheme for NDA data evaluation. Proc. 5th ESARDA Annual Symp., Versailles (France), 19-21 April 1984

/3/ G. DE GRANDI, M. FRANKLIN, S. GUARDINI, G. CORTELLAZZI, M. MONTAGNI — Software, data structures and data evaluation algorithms for a verification data management system. Proc. 6th ESARDA Annual Symp., Venice (Italy) 14-18 May 1984

/4/ G. DE GRANDI, M. FRANKLIN, S. GUARDINI — A project for NDA data treatment and the evaluation of instrument performance at the JRC-Ispra. Advisory Group Meeting on the Evaluation of the Quality of Safeguards NDA Measurement Data. Vienna, 19-23 Nov. 1984

/5/ G. CONTI, M. CORBELLINI, S. GUARDINI — Gamma spectrometry measurements for the determination of ^{235}U abundance with laboratory and field instruments. Proc. 7th ESARDA Annual Symp., Liège (Belgium), 21-23 May 1985

/6/ 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. 7th ESARDA Annual Symp., Liège (Belgium), 21-23 May 1985

/7/ P. DE BIEVRE, B.S. CARPENTER et al. — Progress report on the establishment of uranium isotope abundance reference materials for NDA. Proc. 4th ESARDA Annual Symp., Petten (Netherlands), 27-29 April 1982

/8/ M. CORBELLINI, CEC, JRC-Ispra. Personal communication

/9/ R. JAARSMA, H. RIEF — TIMOC 72 code manual. EUR 5016.EN (1973)

/10/ C. DEVILLERS, C. DUPONT — Mercure IV: Un programme Montecarlo à trois dimensions pour l'intégration des noyaux punctuelles d'atténuation en ligne droite. CEA-N-1726 (1974)

/11/ F. BOGGIANI, A. SALLUSTIO — Metodo di correlazione neutronica per l'analisi non distruttiva di materie fissili a base di plutonio. Tesi di laurea, Politecnico di Milano, CESNEF-Istituto di ingegneria nucleare, 1983

/12/ G. BUSCA, M. CUYPERS, S. GUARDINI — General criteria for the procurement of plant specific reference materials. Proc. 4th ESARDA Annual Symp., Petten (Netherlands), 27-29 April 1982

/13/ R. ABEDIN-ZADEH et al. — Preparation of a plant specific standard for MOX pins. Proc. 4th ESARDA Annual Symp., Petten (Netherlands), 27-29 April 1982

International Enrichment CRMs for Gamma Spectrometry and Safeguards

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Introduction

During the summer of 1985 a really unique international project came to its conclusion. To this project many persons and institutions have contributed. It is unique for three reasons:

1. It is the first time that an international Certified Reference Material (CRM) for Non-Destructive Analysis (NDA) has been made.
2. The new CRMs allow a determination of the $^{235}U/U$ abundance by gamma spectrometry, with an accuracy comparable to the results of mass spectrometry.
3. These CRMs will facilitate the safeguards acceptance of the calibrations, thanks to the ultrasonic seals applied on the CRMs.

The specifications are:

EC CNRM 171 NBS SRM 969

^{235}U Isotope Abundance Certified Reference Material (U_3O_8) for Gamma Spectrometry

$^{235}U/U$ abundances	
Mass per cent	Uncertainties
0.3166	0.0002
0.7119	0.0005
1.9420	0.0014
2.9492	0.0021
4.4623	0.0032

Confidence level 95%

Each sample contains 200 gram of uranium oxide in an aluminium can with a well specified bottom window, which allows accurate gamma spectrometric measurements of 186 keV gamma-rays in the "infinite thickness" geometry.
The CRMs can be obtained from the Commission of the European Communities, Joint Research Centre, Geel Establishment (CBNM), or in the United States of America from the US Department of Energy, New Brunswick Laboratory.

Progress in NDA Measurement of Enrichment

Twenty five years ago the sodium iodide scintillation detectors were introduced for enrichment measurements. Use of "standards" led to very good accuracy claims for the very precise counting results. The limiting factor was the accuracy of the mass spectrometry, applied to the "standards". Today the same boundary condition still holds. The relative precision of 0.001 has to be supported by CRMs which allow an equally accurate calibration.

The introduction of the semiconductors detectors for gamma spectrometry improved energy resolution, and similar reproducibilities have been obtained again. The calibration always required destructive sampling and mass spectrometry for the highest accuracies. It is even today impossible to base accurate measurements on calibrations of the counting equipment with accurately specified radiation sources. The two fundamental problems encountered by that approach are:

1. the accurately defined gamma sources normally are not better defined than 0.5% relative accuracy;
2. because accurate nuclear data on photon emission probabilities and on the photon mass attenuation coefficients are lacking, it is impossible to calculate accurately the 186 keV emission rate per surface area of the sample.

In order to take advantage of the excellent performance of today's mass spectrometry for the calibration of the accurate NDA measurements, the logical solution is the use of the present CRMs, which are specially designed for the calibration of gamma spectrometric enrichment measurements in the "infinite thickness" geometry.

Publications

A full description of the historical development of the project has been given in a paper at the ESARDA Symposium in Venice /1/. It contains a complete list of references to the international project to produce the CRMs.

Fundamental aspects of the role of ESARDA in view of the existing international framework for the certification of standards were investigated and the results of those considerations were described in a paper for the ESARDA Symposium in Edinburgh in 1980, entitled: "The ESARDA Approach to International Standards" /2/.

The meticulous process of certification of these CRMs has been concluded with the publication of the official certification reports. As this is an international project, the reports have been issued by the two certifying "authorities" involved, namely CBNM and the United States of America National Bureau of Standards /3/.

In the design of the CRMs, all factors which might have influence on the measurement results were discussed and assessed. The knowledge on the measurement technique, which is essential for the execution of precision meassurements, has been compiled together in a "Users Manual" /4/. The users are asked to give their comments on the manual based on their experiences. With their support it is possible to elaborate the manual to a good procedural standard for the gamma spectrometric determination of the $^{235}U/U$ abundance.

Some Fundamental Concepts

Infinite thickness

To obtain a reasonable counting rate, even with the depleted sample, a counting geometry has been chosen of a cylindrical can on top of a cylindrical collimator with a diameter which allows counting with the full sensitive surface of the Ge-detector.

If about 200 gram of uranium oxide is put into the can, the counting rate is about 99.9% of the counting rate which can be obtained if the amount of oxide is infinite. This natural limit arises due to the self-absorption of the 186 keV photons by the uranium itself. Thus originated the concept of "infinite thickness" geometry, or the "enrichment meter principle" /5/.

Traceability

The ultimate basis of all measured values is the SI-base units of the "International System of Units". In order to have a good traceability of the measurements to this basis it is necessary to establish the shortest measurement chain between the measurement and the relevant SI-base units. The accuracies and precisions of each step have to be known, and they should be acceptably small /6/.

Hierarchy of standards

Owing to the use made of the terms primary, secondary, etc. it was necessary to agree on new terms to distinguish the different grades of the standards or reference materials. The newly used

terminology is:

1. Certified Reference Material (CRM)
   A reference material one or more of whose property values are certified by a technically valid procedure, accompanied by or traceable to a certificate or other documentation which is issued by a certifying body /7/.
2. Reference Material (RM)
   A material or substance one or more properties of which are sufficiently well established to be used for the calibration of an apparatus, the assessment of a measurement method, or for assigning values to materials /7/.
3. Normalisation Samples
   A sample selected from production material with a nominal value for the parameter of interest assigned from production data /8.b/.

Acceptance

In 1976 and 1977 two IAEA-Advisory Groups discussed the safeguards acceptance of NDA-measurement data, and the related problems of procurement of reference materials /8/. The basic philosophy recommended by those advisory groups, has been followed in the procurement scheme for these CRMs.

Another important factor was that there existed a long standing close contact between the NBS and the CBNM which included exchange of information on a possible co-operation in the project to make those CRMs for NDA.

In the definition phase of the project the ESARDA working group on techniques and standards for NDA took advantage of the existing possibility for international co-operation on the certification between the NBS and JRC. It would ensure compatibility of measurements internationally, increase the availability of identical materials, and provide for a monetary saving by the sharing of characterization results and methods.

Initially the NBS work on the project was funded by the USA Nuclear Regulatory Commission, which also will use the CRMs for its inspection purposes. Similarly, the Euratom Safeguards Directorate has been engaged in the project, and contributed to its development. The ultrasonic Euratom seals enable the Safeguards Inspectorates to verify the CRMs in the field quickly.

Also the IAEA has been an observer to the project since its start. However, before the certified values of the CRMs can be accepted by the IAEA, an independent verification remains necessary.

Strangely enough, there is little legal basis for the acceptance of measurement values obtained with these CRMs in the fuel cycle in the European Community as well as in the national legislation of the member states. Physical quantities like enrichment or isotopic abundance have not yet attracted the same legislative attention as length or mass. For commercial contracts one still has to recur to an umpire laboratory, in case of dispute.

The CRMs, and the meticulous work which underlies their characterization and certification offers a traceable basis for enrichment measurements. Due to the work of CBNM and NBS direct traceability to the SI-units is assured.

The future role of the CRMs, described here, will depend completely on the voluntary acceptance of their certified values by the users in the fuel cycle.

Conclusion

The co-operative certification by NBS and CBNM forms a unique broad basis for international acceptance. The careful design and the accurate certified enrichment values enable accurate and traceable calibration for enrichment measurements by gamma spectrometry. The manual forms a basis for a procedural standard for these measurements. The safeguards identification seals make the CRMs also a reliable reference for safeguards uses.

The high accuracy reached in these radiation sources opens the possibility for new investigations in the field of very accurate gamma spectrometry counting; for instance the comparison of detection efficiencies of germanium detectors.

The project that has produced these CRMs could be an example for a similar project in the future, for example a project for plutonium samples with certified isotopic compositions.

The present CRMs are the result of the contributions of many members of the ESARDA working group on techniques and standards for NDA, the production, characterization and certification work of CBNM and NBS, and the support from the inspectorates of IAEA, Euratom and US-NRC.

The NRM 171 / SRM 969 in its case, together with two transport containers for the single standard samples. The set is completed with an empty can, which allows comparison measurements on material with an unknown enrichment in exactly the same geometrical conditions.

References

1. R.J.S. Harry, "Enrichment Standards for Gamma-Spectrometry", Proceedings of the 6th Annual Symposium on Safeguards and Nuclear Material Management, 14-18 May 1984, Venice, Italy; JRC-Ispra (VA), Italy (1984), ESARDA-17, p. 187-194
2. R.J.S. Harry, "The ESARDA Approach to International Standards", Proceedings of the 2nd Annual Symposium on Safeguards and Nuclear Material Management, 26-28 March 1980, Edinburgh, Scotland; JRC-Ispra (VA) Italy (1980), ESARDA-11, p. 258-263
3. P. De Bièvre, B.S. Carpenter et al., "^{235}Uranium Isotope Abundance Certified Reference Material for Gamma Spectrometry, EC Nuclear Reference Material 171, Certification Report", Central Bureau for Nuclear Measurements, Commission of the European Communities, Joint Research Centre, Geel, Belgium (1985), COM 4153 (published in parallel as: B.S. Carpenter et al., "Uranium-235 Isotope Abundance Standard Reference Materials for Gamma Spectrometry Measurements", U.S. Department of Commerce, National Bureau of Standards, Washington, D.C. 20234, U.S.A., NBS Special Publication 260-96
4. P. Matussek, "Accurate Determination of ^{235}U Isotope Abundance by Gamma Spectrometry. A User's Manual for the Certified Reference Material EC-NRM-171 / NBS-SRM-969", Kernforschungszentrum Karlsruhe GmbH (1985), KfK 3752

5.a J.T. Russel, "Method and Apparatus for Nondestructive Determination of ^{235}U in Uranium", U.S. Patent 3,389,254, June 18, 1968 (to United States Atomic Energy Commission)

5.b L.A. Kull, R.O. Ginaven, "Guidelines for Gamma-Ray Spectroscopy Measurements of ^{235}U Enrichment", Brookhaven National Laboratory Associated Universities, Inc., Upton, New York 11973, U.S.A. (March 1974), BNL 50414 (Safeguards-Nuclear Materials Security TID-4500)

5.c J.L. Parker et al., "Passive Assay - Innovations and Applications", Institute of Nuclear materials Management Twelfth Annual Meeting, 29 June - 1 July 1971, INMM, Chicago, Illinois 60631, U.S.A., Vol. II, p. 515-547

6.a C.D. Bingham, H.Th. Yolken, W.P. Reed, "Nondestructive Assay Measurements can be Traceable", Nuclear Materials Management, Journal of the Institute of Nuclear Materials Management, INMM, Chicago, Illinois 60631, U.S.A., Summer 1976, Vol. V, No. II, p. 32-35

6.b G.A. Uriano, C.C. Gravatt, "The Role of Reference Materials and Reference Methods in Chemical Analysis", CRC Critical Reviews in Analytical Chemistry, October 1977, p. 361-411

6.c P. De Bièvre, J. Huré, "Reference Materials in the Nuclear Fuel Cycle", Invited Review Paper at the International Symposium on Production and Use of Reference Materials, Berlin, November 1979

7. "Terms and definitions used in connection with reference materials", ISO Guide 30, International Organization for Standardization, Geneva, Switzerland (1981), Ref. No. ISO GUIDE 30-1981 (e)

8.a "Report on the Advisory Group Meeting on Qualification of Nondestructive Analysis for Application in IAEA Safeguards Verification Activities, Vienna, 8-11 June 1976", IAEA, Vienna, Austria (1976), AG-80

8.b "Advisory Group Meeting on the Use of Physical Standard in Inspection and Measurements of Nuclear Materials by Non-Destructive Techniques, 22-26 August 1977", IAEA, Vienna, Austria (1977), AG-112

LB-AB-85-009-EN-N