◀ ESARDA
Bulletin

1983 Target Values for Uncertainty Components in Fissile Element and Isotope Assay

Achievable Uncertainties in Destructive Assay of Nuclear Material

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endorsed by the Working Group on Techniques and Standards for Destructive Analysis of the European Safeguards Research and Development Association (ESARDA) on 5 October 1983 and written on behalf of this group.

Abstract

A list of "1983 Target Values" for achievable uncertainties components in fissile element and isotope assay, has evolved gradually from discussions within the ESARDA Working Group on Techniques and Standards for Destructive Analysis over several years. The list is given with a careful wording of what it means and of what it does not contain.

It is explained how it could be useful in safeguards accountancy verification by destructive assay.

Introduction

The Working Group on Techniques and Standards for Destructive Analysis (WGDA) of the European Safeguards Research and Development Association (ESARDA), which at present groups the representatives of 37 nuclear analytical laboratories, has long been concerned with defining realistic performance characteristics of destructive analysis techniques.

One of the terms of reference of the Working Group is : "To evaluate and recommend criteria for destructive analysis of nuclear materials for use by plant operators and safeguarding authorities".

Some of the most important and most badly needed criteria are those to be used for judging results of quantitative determinations of fissile element and isotope amounts. The working group has recognized and discussed this problem at several meetings and decided that it was appropriate to fix reasonable levels of performance as "goals" for nuclear analytical laboratories.

In 1979, the group presented a list of so-called "Target Values" for the uncertainty components in destructive analytical methods to the safeguards authorities of Euratom and IAEA. This list was intended to provide estimates of the capability which could reasonably and realistically be expected at

that time from analytical laboratories. The analytical performance was characterized by two parameters:

a) a value (s) as the estimated standard deviation of a single determination; this value gives an estimate of the reproducibility or precision of a measurement;

b) a value (e) as the estimated standard deviation of the correction factor for systematic errors, resulting in particular from the calibration for such errors.

This list was dated (in this case: 1979), because the parameters leading to it are believed to vary with time as methods are improved or developed.

Uncertainties in analytical measurements: the concept of "Target Values"

Nuclear material safeguards of nuclear facilities involves quantitative verification of the accountancy of fissile materials by independent measurements. Clearly the quality of the measurements, performed by both the operator of a nuclear facility and the inspector, is one of the main parameters determining how effective the safeguards verification can be. The International Atomic Energy Agency (IAEA) expects, therefore, that the relevant measurement systems meet "the most recent internationally accepted standards or their equivalent" /1/. The question now arises as to what are "internationally accepted standards" ? The WGDA has made an attempt to answer this question by introducing the concept of "Target Values" which intend to describe the level of performance which many laboratories achieve on a routine basis. More experienced laboratories would be expected to perform considerably better. Laboratories with less experience or entering the field should aim at achieving this "target" performance level. In other words,

it is attempted to describe a reasonable "state-of-the-practice".

It is proposed that these "Target Values" be used by the safeguards authorities as a guidance to judge differences between the facility declaration and the verification measurement. The IAEA has accepted, in fact, the concept in its present approach for the evaluation of safeguards data /2/.

An updating of the "1979 Target Values" has now been carried out jointly by the working group and the IAEA. The updated estimates of achievable uncertainties, which we shall examine below, will be referred to as the "1983 Target Values".

This paper reflects the present views of the working group and of the IAEA on this topic.

Needs and utilization of "Target Values"

What the "Target Values" should mean, cannot be made clear without considering why and where such data are needed and how they would be utilized in safeguards verifications.

Let us therefore examine briefly the Agency's approach as it was described at the ESARDA Specialists' Meeting in Petten in April 1982 /2/. Figure 1 outlines a typical IAEA procedure of the evaluation of operator-inspector (X-Y) differences. The random and systematic uncertainties are estimated after exclusion of outliers to ensure that the differences are not unnecessarily inflated.

These estimates describe the actual performance of the measurement system and include all sources of normal clerical, measurement and sampling errors actually encountered in real verifications. This actual performance can then be compared to design information provided by the operator to safeguards authorities : it contains statements by each facility operator on the

magnitude of its measurement errors. It was learned that some independent reference is needed to judge the adequacy of both design and performance values of measurement uncertainties.

Initially the Agency for example achieved this by defining capability values based on careful analytical and statistical evaluation of the measurement data involved in order to establish the present capability of the measurement system under normal industrial and verification conditions. Chemical and statistical considerations were used to establish capability values that are not influenced by outliers, unusual biases or underestimates of precision. The capability data provide the basis on which the size of the sampling effort should be based and on which the significance of the observed difference is to be tested in order to detect possible diversions of quantities of safeguards significance. All participants in this Working Group agree that "Target Values" as such are desirable and that they should be defined to include all **uncertainty contributions of the measurement systems,** i.e. not only the uncertainties in the final measurement step, but also all sample treatment uncertainties prior to the measurement as well as the sampling and the sampling plan uncertainties. It is important to accurately describe what the present list (the "1983 Target Values") does contain and does not contain.

Content and meaning of the list of 1983 Target Values

Uncertainties

An effort has been made with the "1983 Target Values" to consider the total analytical uncertainty from both measurement and sample treatment errors. It does not contain estimates for possible sampling errors because at this stage it has not yet been possible to evaluate them properly. The objective was to give a global estimate of all uncertainties occurring after the taking of the sample, i.e. in its treatment and packing before shipment, its possible modifications during transport, the uncertainties in correcting for such effects, its analytical treatment in the laboratory and all other laboratory measurement uncertainties, giving due account to potential interferences and uncertainties of calibration (s) (e).

Clearly, target values for the presently intended purpose can and should not represent the ultimate performance of a measurement method that can be obtained under exceptional or ideal laboratory conditions ("state of the art"). Target values must be achievable in normal industrial laboratory conditions and verification practices ("state of the practice"). However, they must be
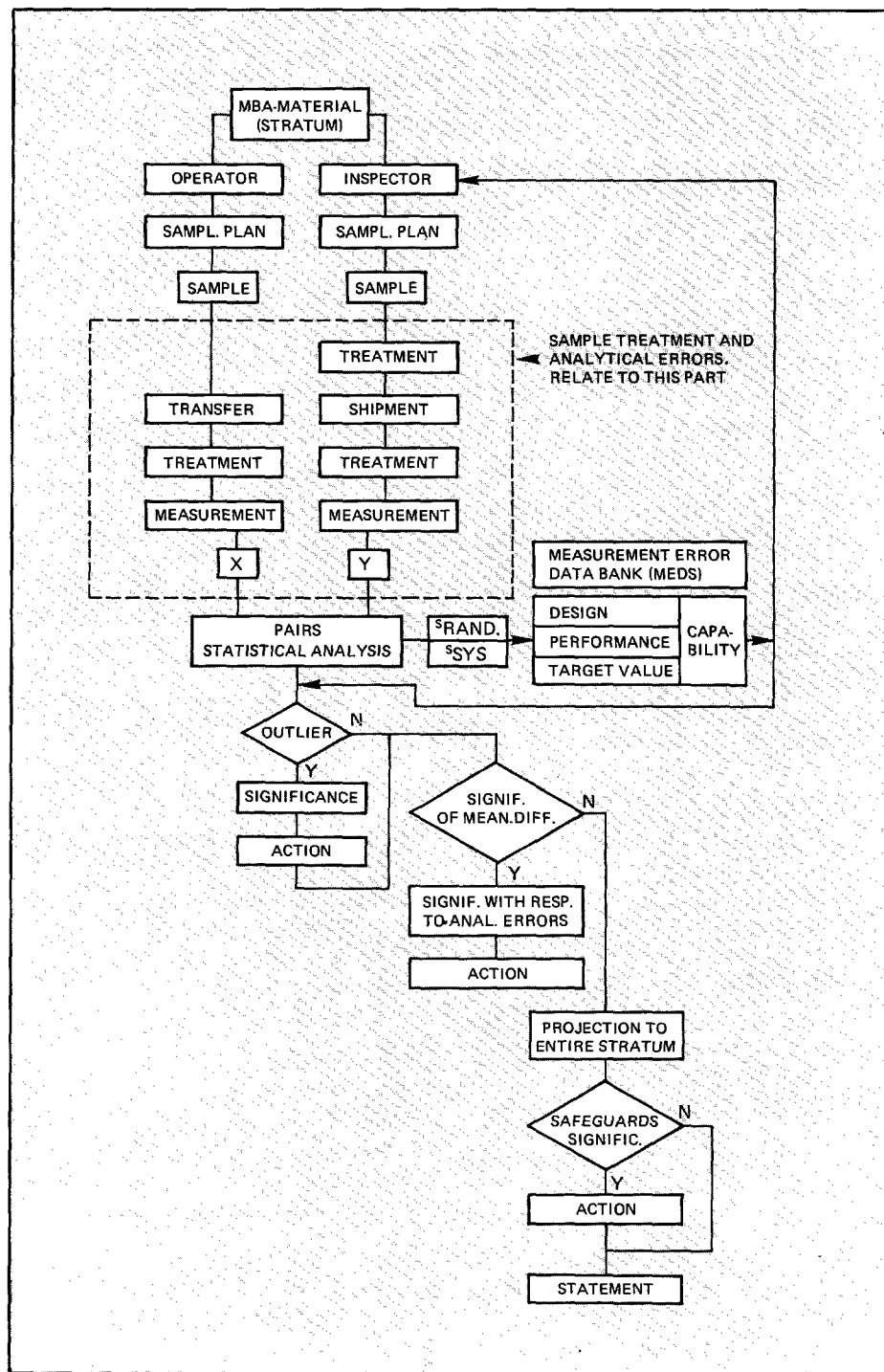

Fig. 1 - Evaluation of Operator - Inspector Differences

sufficiently strict to enable meaningful and effective safeguards.

Materials

It is clear that the safeguards requirements vary with the nature and the type of nuclear materials e.g. plutonium dioxide is a more sensitive material while natural and depleted uranium have much lower significance. Accountancy and verification procedures and practices therefore differ according to materials. Consequently it is appropriate to examine the expected performance of methods in conjunction with the nature of the material under safeguards.

The materials considered cover the feeds, intermediate compounds and products of all nuclear installations under safeguards. Research or prototype materials like U-Ta, U-Nb alloys have not been discussed in the present context.

Three grades of material quality are distinguished, using currently used classification:

- nuclear grade (NG) material:
  fulfils all NG industrial specifications or is equivalent to these in chemical purity.
- scrap materials:
  are considered separately if they contain 0.2 to 2% of possible interferences (the fissile elements remain the major constituent).
- waste materials:
  cover materials of lower purity, particularly when the fissile element is very diluted and only constitutes a minor fraction of the total.

Sample amount and analytical methods

The present paper is concerned only with methods which are of significance in material accountancy. It is thereby realized that the performance of an analytical procedure is frequently limited by the amount and the representativity of the original sample. It was impossible at this stage to define the minimum amount of material which must be taken to obtain a representative sample. Short of this, it was however felt useful to indicate the amount of the sample aliquot used in the final measurement to achieve the indicated performance. Also, rather than reviewing all methods individually, it was thought possible to group the methods in categories of similar characteristics and performance.

Eight categories of methods for chemical assay and four categories of methods for isotopic assay were selected. Table 1 lists the typical amount of element required for one measurement by each category of method. In general sample preparation is excluded except in the case of isotope dilution mass spectrometry. In this method, indeed the isotope dilution and the chemical treatment of the spiked sample cannot be dissociated from the final mass spectrometric measurement.

Titrimetry can contain the NBL Davies and Gray titration of uranium, AgO, CuCl or TiCl, redox titrations of plutonium, and complexometric titration of thorium.

Only controlled potential coulometry is considered under coulometry.

The 1983 Target Values of uncertainty components of analytical determinations

For all material-method combinations of practical interest to safeguards, separate estimates have been defined for the coefficients of variations of the random uncertainty as well as of the correction factor for systematic errors, which are to be expected in the material, analytical treatment and measurement.

Table 1: Analytical methods and typical amounts of sample need to perform one measurement

Code	Method	Typical amount of element required for a single measurement (excluding sample preparation) (b)			Unit
		U	Th	Pu	
01	Titrimetry	20 - 100	50 - 100	5 - 50	mg
02	Coulometry	2 - 20	-	2 - 10	mg
03	Gravimetry	2 - 20	0.5 - 1	0.2 - 3	g
04	X-ray fluorescence	0.1 - 30	0.1 - 30	0.1 - 30	mg
05	Isotope Dilution Mass Spectrometry	10 - 2000	-	1 - 20	μg
06	Spectrophotometry	20 - 500	5 - 250	(a)	μg
07	Fluorometry	2 - 500	-	-	ng
08	Alpha Counting	2 - 250	-	0.1 - 1	μg
21	Gas Mass Spectrometry	20	-	-	mg
22	Thermal Ionization Mass Spectrometry	1 - 50	-	0.1 - 5	μg
23	Gamma Spectrometry	0.1 - 1	-	0.01 - 0.1	g
24	Alpha Spectrometry	-	-	0.1 - 1	μg

(a) Spectrophotometry - Direct measurement at 830 nm of Pu (VI) 0.1 - 10 mg
    - Colorimetry 1 - 100 μg

(b) The sample preparation is not considered except for the spiking and chemical treatment in isotope dilution analysis (Method 05)

The random component (s) indicates the short term reproducibility of one complete assay, estimated from the independent sample treatment and measurement of a series of replicate samples of a homogeneous material.

The systematic component (e) is an estimate of the magnitude of the uncertainty in calibrations for known errors as well as estimates for unknown errors. It refers again to the complete analytical process and not only to the final measurement step.

Random (s) and systematic (e) estimates of coefficients of variation are given only for the material-method combinations which were felt to be normally in effective use for elemental assay (see Table 2). For isotopic measurements only those isotopes are listed which are considered of major importance (see Table 3).

The results of interlaboratory measurement evaluation programmes /3 , 4 , 5 , 6/, the experience of plant laboratories /7/ and of Safeguards Authorities /2/ provided the experimental evidence necessary to define the estimates. Due account has been given to the fact that interlaboratory evaluations are run in privileged conditions compared to routine situations. Also, both interlaboratory evaluations and plant procedures rarely involve the limitations which are to be faced in safeguards verifications, e.g. the problems with shipments and instability of samples. On

the other side the evaluation of safeguards data shows that in several instances the measurement system does not yet meet a desirable standard. Thus existing experimental evidences alone are not sufficient at present. Consequently the estimates, listed in Tables 2 and 3, actually evolved from a confrontation of existing experimental evidence with safeguards requirements. A couple of examples will illustrate this point.

A first example is taken from thermal ionization mass spectrometry of the U-235 isotope (Table 3). The targets set for uncertainty contributions from systematic errors in measuring depleted, natural and low enriched uranium, may not appear very impressive in terms of modern measurement capability (<0.1 to 0.2% relative in these cases). Yet, the targets proposed for the determination of low abundances are compatible with safeguards needs and should be achievable with a minimum of precautions, even when analysing with the same instrument a large number of samples of varying natures and abundances. In view of the above arguments one may question why a value as low as 0.1% has been retained as target uncertainty contribution from systematic errors in the gas mass spectrometry of low abundant U-235 isotope. The reason is that here such a performance is specifically and currently achieved on

Table 2: 1983 Target Values for Uncertainty Components in Element Assay (estimated relative standard deviations in percent)

Method		Titrimet.		Coulom.		Gravim.		X-Ray Fl.		IDMS		Spectroph.		Fluorim.		α-count.		Remarks
Code		**01**		**02**		**03**		**04**		**05**		**06**		**07**		**08**		
Material	El	s	e	s	e	s	e	s	e	s	e	s	e	s	e	s	e	
UF_6	U	0.3	0.3	0.3	0.3	0.15	0.1	—	—	—	—	—	—	—	—	—	—	
U-solutions, pure	U	0.15	0.15	0.15	0.15	0.1	0.15	0.5	0.5	—	—	—	—	—	—	—	—	
U-solutions, scrap	U	0.3	0.5	—	—	—	—	3.0	0.5	—	—	3.0	5.0	10	10	—	—	
U-oxide powder, NG	U	0.15	0.15	0.15	0.15	0.1	0.1	—	—	—	—	—	—	—	—	—	—	★
U-oxide powder, scrap	U	0.3	0.5	0.3	0.5	—	—	1.0	1.0	—	—	—	—	—	—	—	—	
U-oxide, sintered mat.	U	0.15	0.1	0.15	0.15	0.05	0.1	—	—	—	—	—	—	—	—	—	—	
UF_4 powder NG	U	0.15	0.3	0.15	0.3	0.15	0.15	—	—	—	—	—	—	—	—	—	—	★
U metal, NG	U	0.15	0.1	0.15	0.1	—	—	—	—	—	—	—	—	—	—	—	—	
U-Al based mat., NG	U	0.2	0.3	0.2	0.3	—	—	—	—	—	—	—	—	—	—	—	—	
U-Al based scrap	U	0.3	0.5	0.3	0.5	—	—	—	—	—	—	—	—	—	—	—	—	
Zr-U materials	U	0.5	0.3	—	—	—	—	1.0	1.0	—	—	—	—	—	—	—	—	
U carbides	U	0.3	0.5	0.3	0.5	0.2	0.5	—	—	—	—	—	—	—	—	—	—	
(Th, U) 0_2, Kernels & BISO	U	0.15	0.2	—	—	—	—	1.0	0.5	—	—	—	—	—	—	—	—	
	Th	0.2	0.2	—	—	0.3	0.2	1.0	0.5	—	—	—	—	—	—	—	—	
(Th, U) 0_2 or $U0_2$, TRISO	U	0.3	0.2	—	—	—	—	1.0	1.0	—	—	—	—	—	—	—	—	
	Th	0.3	0.3	—	—	0.5	0.3	1.0	1.0	—	—	—	—	—	—	—	—	
U scraps, dirty & diluted	U	0.5	2.0	—	—	—	—	3.0	5.0	—	—	5.0	10	10	15	—	—	
Spent fuel inputs, HWR &	U	0.5	0.5	—	—	—	—	0.5	0.5	0.5	0.5	—	—	—	—	—	—	
LWR	Pu	—	—	—	—	—	—	1.0	1.0	0.5	0.5	1.0	1.0	—	—	—	—	
Spent fuel inputs, FBR	U	0.3	0.2	—	—	—	—	0.5	0.5	0.5	0.5	—	—	—	—	—	—	
	Pu	0.3	0.2	—	—	—	—	1.0	1.0	0.5	0.5	1.0	1.0	—	—	—	—	
Liquid Waste, HAW	Pu	—	—	—	—	—	—	5.0	10	—	—	—	—	—	—	10	10	
MAW	Pu	—	—	—	—	—	—	—	—	—	—	—	—	—	—	5	5	
LAW	Pu	—	—	—	—	—	—	—	—	—	—	—	—	—	—	5	5	
Pu nitrate, pure	Pu	0.2	0.2	0.2	0.2	0.1	0.15	0.5	0.5	—	—	—	—	—	—	—	—	
U/Pu nitrate sol. pure	U	0.2	0.2	0.2	0.2	—	—	0.5	0.5	—	—	—	—	—	—	—	—	★ ★
	Pu	0.2	0.2	0.2	0.2	—	—	0.5	0.5	—	—	—	—	—	—	—	—	
U/Pu solutions, waste	U	—	—	—	—	—	—	3.0	5.0	1.0	1.0	5.0	5.0	—	—	—	—	
	Pu	—	—	—	—	—	—	3.0	5.0	1.0	1.0	2.0	2.0	—	—	5	5	
Pu 0_2 powders	Pu	0.2	0.2	0.2	0.2	0.2	0.2	—	—	—	—	—	—	—	—	—	—	★
(U, Pu) 0_2 - MOX, LWR	U	0.3	0.2	0.3	0.2	—	—	—	—	—	—	—	—	—	—	—	—	
	Pu	1.0	0.5	0.2	0.2	—	—	—	—	—	—	—	—	—	—	—	—	★ ★ ★ ★
(U, Pu) 0_2 - MOX, FBR	U	0.3	0.2	0.3	0.2	—	—	0.5	0.5	—	—	—	—	—	—	—	—	
	Pu	0.3	0.2	0.2	0.2	—	—	0.5	0.5	—	—	—	—	—	—	—	—	★ ★ ★
(U, Pu) 0_2 - MOX, scrap	U	0.5	0.5	—	—	—	—	3.0	5.0	—	—	—	—	—	—	—	—	
	Pu	1.0	0.5	0.2	0.5	—	—	3.0	5.0	—	—	1.0	1.0	—	—	—	—	

★   *Control of oxidation and/or moisture pick-up required*
★ ★   *U/Pu ratio = 3*
★ ★ ★   *1 to 4% Pu*

Table 3: Target Values for Uncertainty Components in Isotope Assay (estimated relative standard deviations in percent)

Method		Gas MS*		Thi MS		γ-Spec.		α-Spec.		Remarks
Code	**Abund.**	**21**		**22**		**23**		**24**		
Isotope	**%**	s	e	s	e	s	e	s	e	
^{235}U	0.2	0.1	0.1	1	1	5	3	—	—	All Uranium bearing mat.
	0.7	0.05	0.1	0.75	1	2	1	—	—	All Uranium bearing mat.
	3	0.03	0.1	0.5	0.5	1	0.5	—	—	All Uranium bearing mat.
	20	0.02	0.1	0.2	0.1	0.5	0.5	—	—	Pure Uranium compounds
	90	0.01	0.03	0.05	0.03	0.2	0.5	—	—	Pure Uranium compounds
	20	—	—	0.2	0.2	0.5	0.5	—	—	Alloys and mixed fuels
	90	—	—	0.1	0.2	0.2	0.5	—	—	Alloys and mixed fuels
^{238}Pu	0.3	—	—	2	2	—	—	2	2	For pure Plutonium materials
	1.5	—	—	1	1	—	—	3	2	For pure Plutonium materials
^{239}Pu	80-50	—	—	0.15	0.1	—	—	—	—	For pure Plutonium materials
^{240}Pu	10-30	—	—	0.3	0.3	—	—	—	—	For pure Plutonium materials
^{241}Pu	3	—	—	0.5	1	—	—	—	—	For pure Plutonium materials
	15	—	—	0.5	0.3	—	—	—	—	For pure Plutonium materials
^{242}Pu	1-5	—	—	0.5	0.5	—	—	—	—	For pure Plutonium materials

uranium hexafluoride in gas diffusion plants where several instruments are always available and where each one may be dedicated to the assay of a narrow range of isotope abundances.

A second example is taken from the isotope dilution of plutonium (Table 2) in solutions of spent fuels : a target systematic error contribution of 0.5% still represents a real challenge which is yet not commonly achieved in interlaboratory evaluations /3 , 4/ nor in safeguards verifications /2/. Basically the measurement technique is capable of achieving an accuracy of 0.1%, but in practice the uncertainties of corrections for systematic errors have coefficients of variation of 0.7 to 1.5%. Performance is probably still limited both by calibration and treatment errors.

Conclusions

1. The "1983 Target Values" for uncertainty components in element and isotope assay result from a difficult but hopefully wise examination of all sources of uncertainties occurring after the taking of the samples, tempered by a genuine consideration of reasonable safeguards needs.

2. They should be achievable or need to be achieved in 1983 under routine laboratory conditions.

3. By no means do they represent the ultimate capability ("state of the art") of the analytical techniques considered.

Safeguards authorities should utilize these data in their evaluations in order to decide whether the measurement systems are satisfactory or whether the analysts must be asked to improve their performance. It is hoped that safeguards inspectorates may be in a position to publish comparisons between the "TARGET" and "PERFORMANCE" values. The aim thereby should be to identify where the "PERFORMANCE" is satisfactory or lagging, and whether, in the latter case, this may be due to significant sampling errors.

The ESARDA-WGDA is prepared to discuss such comparisons in general and to examine selected cases with appropriate sampling experts at the request of the safeguards authorities in order to improve sampling procedures when needed. In particular, one may in this way define progressively the minimum amount of sample to be taken and the "Target Values" for the sampling errors.

Ultimately it should be possible to provide estimates of the total uncertainty, including sampling, which may be achieved under normal industrial and verification conditions. These data will need periodical updating in view of changes in performance and/or in the criteria for safeguarding nuclear installations.

Acknowledgments

References

1. INFCIRC/153, par. 55, (IAEA) (Wien)
2. G. Hough, E. Kuhn, J. Merrill, P. Siwy, W. Bush, (IAEA). Qualification of measured data for safeguards use, Proc. ESARDA Specialists' Meeting, Petten (April 1982), p. 13
3. Safeguards Analytical Laboratory Evaluation (SALE) 1980 Annual Report, NBL-300 (June 1981)
4. W. Beyrich, W. Golly, G. Spannagel (KFK), P. De Bièvre, W. Wolters, M. Gallet (CBNM), L.A. Machlan, J.W. Gramlich, J.D. Fassett (NBS). The IDA-80 measurement evaluation programme on mass-spectrometric isotope dilution analysis of uranium and plutonium. Some preliminary results. International Symposium on Recent Advances in Nuclear Material Safeguards, Vienna (November 1982), (IAEA-SM-260/33), p. 261
5. The AS-76 interlaboratory experiment, KFK 2860 - EUR 6400e, KFK 2861 - EUR 6401e, KFK 2862 - EUR 6402e (December 1979). W. Beyrich, G. Spannagel (KFK). Part 1: Performance and results.
   G. Spannagel, W. Beyrich (KFK), G. Bortels (CBNM). Part 2: Collection and evaluation of representative spectra.
   G. Bortels, J. Broothaerts, P. De Bièvre, M. Gallet, W. Wolters (CBNM), K.M. Glover, M. King, R.A.P. Wiltshire (Harwell), E.L. Garner, L.E. Machlan, I.L. Barnes (NBS). Part 3: Preparation and characterization of samples
6. P. De Bièvre, W.H. Wolters, G. Müschenborn +, W. De Bolle (CBNM), W. Beyrich, W. Golly (KFK). The 1982 state of the practice in low enriched UF_6 Isotopic measurements in Western Europe, Proc. 5th ESARDA Symposium, Versailles (April 1983), p. 177
7. K.A. Swinburn. Pu accountancy in commercial transactions involving PuO_2, BNFL Report 83(W)(1974)

Note : Tables 1, 2 and 3 will be displayed as a Poster at the 6th ESARDA Symposium in Venice, Italy (14-18 May 1984) and at the INMM Symposium in Columbus, U.S.A. (15-18 July 1984) for comments, discussion and suggestions by Symposium participants.

In the past year, the ESARDA Working Group for Reprocessing Input Verification (RIV) has dedicated particular attention to the problems connected with the measurement and verification of the volume of input solutions. Amongst the various activities carried out, two are worth a special mention.

In October 1983, a workshop was organised at the Ispra Establishment of the Joint Research Centre on the subject "Volume Determination of Reprocessing Input Solutions by Tracer Techniques". The coordinator and Chairman of the workshop, Mr. Guzzi, has summarized the issues which were discussed and the conclusions which were drawn in the three days meeting, which convened 22 specialists from R & D organizations and from Safeguards Authorities.

The WG is also taking part in the Reprocessing Input Tank Calibration Exercise (RITCEX), carried out at the EUROCHEMIC plant, with the sponsorship of the Belgian Government and under the coordination of the CEN/SCK Mol. Mr. Franssen, coordinator of the experiment and an active participant in our WG since its inception, has kindly agreed to describe the aims of the exercise and the progress so far achieved for the ESARDA Bulletin.

The WG is indebted to the authors for these two stimulating reports which are published hereinafter.

C. Foggi, Convenor, ESARDA RIV WG

Report on the Workshop on

Determination of the Volume of Reprocessing Input Solutions by Tracer Techniques

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As announced in the ESARDA Bulletin No. 5, October 1983, a Workshop on "Determination of the Volume of Reprocessing Input Solutions by Tracer Techniques" was held at Ispra from 8th to 9th November 1983.

The Workshop was organised by the ESARDA working group for "Reprocessing Input Verification", and conducted according to the following agenda:

Tuesday, 8th November 1983

1. Welcoming address
2. Approval of the agenda
3. Reports and discussion on:
   - Review of practical experiments
   - Selection of tracers
   - Spiking, homogenization of input solutions and sampling procedures

Wednesday, 9th November 1983

4. Reports and discussion on:
   - Chemical separations
   - Mass spectrometry measurements
5. Future trends, perspectives and recommendations
6. Conclusions

The workshop convened experts from Safeguards Authorities (IAEA, Euratom), Research Organisations (ENEA, CEA, ORNL, UKAEA and JRC) and Industrial Plant Operators (ITREC, EUREX, BNFL).

The list of participants is the following:

Safeguards Authorities
H. Shimojima, IAEA, Vienna
J. Toussaint, Euratom, Luxembourg

Research Organisations
G. Bardone, B. Mattia, ENEA, Casaccia
M. Lucas, CEA, CEN - Saclay
M. Neuilly, CEA, CEN - Cadarache
R. Platzer, CEA, CEN - Fontenay-aux-Roses
J.A. Carter, ORNL, Oak Ridge
A.G. Wain, UKAEA, Harwell
C. Foggi, G. Guzzi, P.R. Trincherini, JRC-Ispra
M. Cuypers, H. Dworschak, M. Franklin, F. Mousty, JRC-Ispra (part time)

Plant Operators
G. Arcuri, S. Miglietta, V. Morano, ENEA-ITREC, Trisaia
V. Calì, C. Guidotti, ENEA-EUREX, Saluggia
R. Howsley, BNFL, Risley

The accounting of the input in reprocessing plants is based on the measurement of three quantities: volume, density of the solution and U and Pu concentrations. The first two are measured by the plant operators, while the third is measured either by the operators or by the inspectors.

The measurement of the volume of the solution is an indirect, very complex and cumbersome technique which presents many problems. The inspector in general can not repeat it: he is limited to observing what the operator does. In some facilities, nevertheless, the inspector has already at his disposal electromanometers to perform partially independent measurements of volumes.

The tracer technique, if correctly developed, could give the inspector an independent verification method for indirectly measuring the masses of U and

The volume of the accountability tank in a reprocessing plant, or better the volume of the hot solution transferred to the plant, is a fundamental parameter for the determination of the amount of U and Pu contained in the tank.

The measurement of the volume is performed by the plant operators using different techniques: an independent verification method could be of benefit both to the operator and to the inspector.

The "tracer technique" could provide a means to reach this goal.

It is based on the addition to the hot solution of a known amount of a specific element (tracer) and on the measurement of its concentration in an aliquot of the solution drawn from the tank after homogenization. The ratio between the concentration of the tracer added and the concentration of the tracer found after mixing gives the volume of the solution in the tank.

The measurement of the tracer concentration in the drawn sample is performed by isotopic dilution mass spectrometry with the aid of a well calibrated spike and after a very good clean up procedure of the tracer element is executed.

Pu contained in the tank also.

Having the above concepts in mind, the main topics of the workshop were:

a. to review the "state of the art" of the tracer technique, to try to identify the problems which it presents and to put forward suggestions for dealing with and solving them,

b. to discuss possible developments and future programmes to make a common and concerted effort, avoiding as far as possible duplication of work and maintaining ever closer contacts between the people working in this field.

Past and ongoing experiments carried out world-wide were reviewed and the following up-dated list was established (see Table 1). The list is probably not complete as some results are not in the literature. Participants were then asked to supply new information in order to gain as complete a picture as possible of what has been achieved in the field and to improve the future work in the light of results which are already available.

Only the EUREX and TASTEX experiments have been performed in operating conditions, while the others have only been carried out in very small scale conditions: some of them in highly controlled laboratory conditions. Quoted reproducibilities for volume determination by using U tracer, range between 0.1% in the best conditions (Marcoule) to 0.5% in the WAK experiment. In regard to this experiment it has been cleared up that the "best values" in the calibration of the 800 litres circular slab (WAK Experiment 1) are those presented here in Figure 1 and not the ones reported in JEOPAG / 10.82-PRG-56 Fig.6, p.19.

Reports on practical experiments carried out at Marcoule, EUREX, Oak Ridge and JRC-Ispra were presented and the results obtained were discussed.

Two applications of the tracer technique appeared to be:

- the determination of the volume of the tanks,
- the determination of the masses of U and Pu in the solution.

In both cases the technique provides results which are independent from the present measurement methods.

According to the final goal of the experiments there are two different procedures:

- to add the tracer in the accountability tank and to sample from the accountability tank,
- to add the tracer in the dissolver and to sample from the dissolver.

One should also investigate the possibility of introducing the tracer - in solid or soluble flask form - directly in the dissolver basket and of sampling after having transferred the

Table 1

Year of the experiment	Site	Volume of the tank (litres)	Tracer used	Laboratory involved
1973	EUROCHEMIC	2,000	Li	Eurochemic
	IDAHO FALLS [1]		Sr	Idaho Nat. Engineer. Lab.
1975	TARAPUR	1,500	Mg	Bhabha Atomic Res. Centre
1975	WAK	800	D_2O	Zentral Büro Geel
1976	TARAPUR	1,500	Pb	Bhabha Atomic. Res. Centre
1979	LA HAGUE [1]	3,000	Mg	CEN Saclay
1980	WAK	800	U, Li, Mg	IAEA-FRG Support Progr.
1982	WAK	800	D_2O	Karlsruhe Nucl. Res. Centr.
1982	MARCOULE	500	U, Mg	CEN Saclay
1982	OAK RIDGE	200	Lu	OAK Ridge Nat. Lab.
1982 (*)	EUREX	300	Mg, Lu	ENEA EUREX-JRC ISPRA
1983	ALKEM	300	Mg, U	IAEA-FRG Support Progr.
1983 (*)	ITREC	100	Lu	ENEA ITREC-JRC ISPRA
1983 (*)	EUROCHEMIC [2]	4,000	Pb, Lu, Nd	IAEA-EURATOM-Belgium Sup. Progr.
1983 (*)	SELLAFIELD		Gd	British Nuclear Fuel Ltd.

(1) Experiments carried as part of TASTEX
    (Tokai Advanced Safeguards Technology Exercise)

(2) Experiment carried as part of RITCEX
    (Reprocessing Input Tank Calibration Exercise)
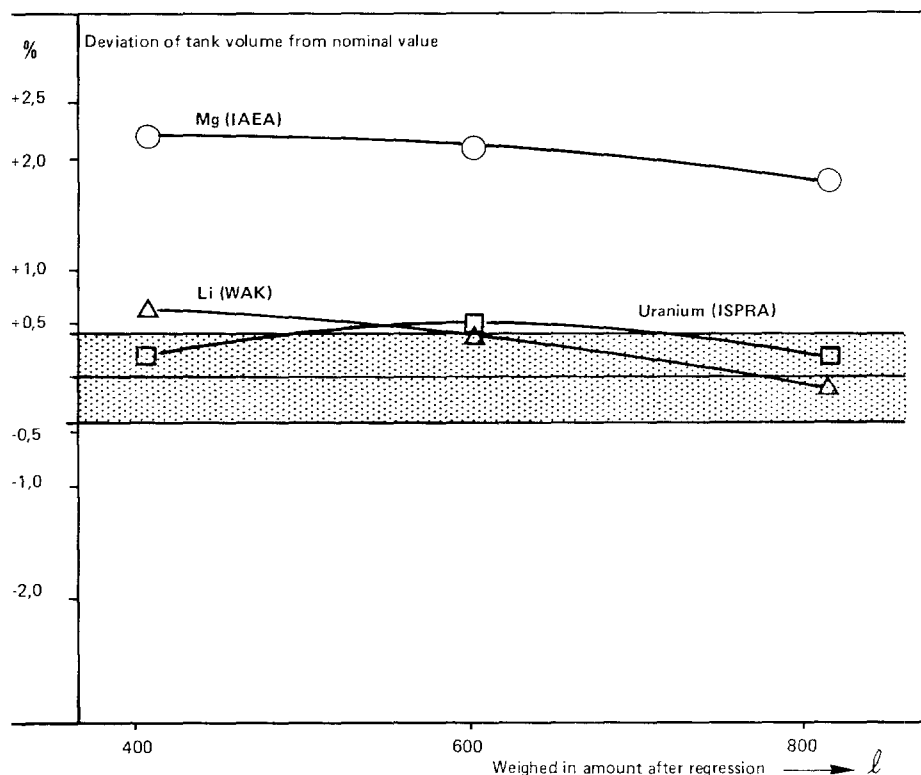
(*) Ongoing experiments.

Fig. 1 - Best values in WAK Experiment 1

homogenized solution into the accountability tank.

The main criteria for the selection of the tracer are the following:

- compatibility with the process
- ease and accuracy of measurement
- minimum risk of contamination in handling
- availability of tracer and spike for isotopic dilution analysis
- absence of isobaric interferences
- low blank in process solution.

These criteria were extensively discussed by the Meeting. The tracers used (or proposed) were: Li, Mg, Sr, Gd, Lu, U (Nd, Pb, Er, Yb), for all of which some experience has been collected in different conditions. There was not a common preference for one of these elements since the choice is also dependent on the final purpose of application, on the facility design, on the long term availability of the tracer and on economic considerations. There was a tendency to prefer elements with high masses, because of the easier measurement conditions, although some doubts were raised concerning the use of Pb.

It was agreed that one of the most critical point of the whole technique is to obtain representative samples for mass spectrometric analysis. This implies high level homogenization procedures and measurements of the density and of the temperature of the solutions. Systematic investigation in these fields are still needed, although studies of both technical and scientific nature are already in progress by CEA.

It was suggested that a study on the hydraulics and on the temperature dependence of the homogenization of the solution could be carried out during the forthcoming RITCEX experiment at Mol. It should be very well prepared in order to avoid the variety of results which occurred in the WAK experiment due to the problems which arose in the various steps of procedure.

After the completion of the RITCEX experiment, another meeting would be desirable to exchange impressions in the light of the results obtained. Much attention should be paid to a correct design of new tanks (especially the larger ones) and to the related equipment, in order to obtain good homogenization of the solution and consequent representativity of the samples.

Procedures for chemical separation of the tracers (Mg, Lu and Gd) from hot input solutions have only been performed at the EUREX plant, in the TASTEX experiment and at BNFL.

There is consequently little information on this subject the principal points of which are:

- to achieve a good separation of the

tracer from the fission products in order to be able to carry out mass spectrometric measurements in the presence of low activity

- to search for simple separation methods in order to reduce the number of steps and render the separations easy to perform remotely.

The chemical separation procedures used until now seem to be satisfactory, but more extended experience is needed to attain standard purification methods for the various tracers.

The precision of volume determination is highly dependent on the mass spectrometry measurements of the elements used as tracers. Heavy elements such as Lu or Gd look a better choice than light elements such as Mg or Li, because of their isotopic fractionation which strongly affects the overall accuracy. A good policy would be to promote in the future joint initiatives for measurements and comparisons in order to learn more about the results and their significance. The advantages of the use of the resin bead technique in the deposition of samples onto filaments were also indicated.

Results have been presented on mass spectrometry measurements performed with various equipment and techniques, at the JRC-Ispra, in France and in the United States. The techniques appear to be at the operational stage without insurmountable major problems.

The following conclusions were drawn by the Meeting:

the "tracer technique" has two possible applications:

- the determination of the volume of the tanks,
- the determination of the masses of U and Pu in the solution.

In both cases the technique provides results independent of the present measurements methods and can be of benefit to the Operators and to the Inspectors. Further research is needed to assess the accuracy and precision of the method in operational plant conditions. It must be noted that each experiment must be designed taking into account the features of the facility, and that quality of the results is likely to be plant specific.

The ongoing experiments (EUREX, ITREC, RITCEX) will help in understanding some of the existing problems; however, a systematic collaboration is desirable among Operators, Research Centres and Safeguards Authorities, leading to in-field exercises in order to demonstrate the practical applicability of the technique.

The following Bibliography is representative of major works relating to the use of the "tracer technique" in both the above mentioned applications.

References

H. Bokelund: Investigation of reprocessing input measurement using tracer technique. Eurochemic Report ETR-266 (final report on IAEA Research Contract 876/RB) (1970)

R. Berg: Verification on reprocessing input measurements. Eurochemic Report ETR-277 (1973)

P. De Bièvre, J. Van Audenhove: An accurate procedure to safeguard the fissile material content of input and output solutions of reprocessing plants. IAEA-SM-201/108, Proc. Symp. Safeguarding Nuclear Materials, Vienna, October 1975

C.K. Mathews, H.C. Jain, S.A. Chitamber et al.: Independent method for input accountability in reprocessing plants: Magnesium tracer technique for the accountability of plutonium (MAGTRAP). BARC-809 (1975)

C.K. Mathews, H.C. Jain, V.C. Kavimandan, S.K. Aggarwal: An independent method for input accountability in reprocessing plants (MAGTRAP). IAEA-SM-201/87, Proc. Symp. Safeguarding Nucl. Material, Vienna, October 1975

P. De Bièvre, J. Broothaerts, M. Gallet, A. Loopmans, E. Sattler, J. Van Audenhove: Performing accurate measurements of fissionable material for safeguards purposes in reprocessing plants in Europe. Proc. 17th Ann. Meeting INMM, June 1976

C.K. Mathews, H.C. Jain, V.C. Kavimandan, S.K. Aggarwal: Tracer techniques for the input accountability of plutonium in reprocessing plants: MAGTRAP and LEADTRAP. Nucl. Techn. 42, 297 (March 1979)

W. Lycke, M. Gallet, F. Peetermans, R. Damen, E. Bouwmeester, P. De Bièvre, J. Van Audenhove: Accurate in-situ assay of total fissile isotopes in inputs or reprocessing plants: a reference method or a definitive method ? NBS Spec. Publ. 582 (1979)

TASTEX Draft Final Report ISPO/03, June 1980

TASTEX Draft Final Report CEA-CdAn No. 80-94, June 1980

P.R. Trincherini, S. Facchetti, H. Hullah: Input measurement in reprocessing plants. EUR 6473 EN (1980)

E. Michiels, P. De Bièvre: A precise method for the mass spectrometric determination of lithium isotope ratios. Proc. 29th Conf. on Mass Spec. Allied Topics, ASMS, p. 240 (1981)

H.J. Hein, J. Lausch, W. Schwerter: Volumetric assay in a 800 L ring slab using a Li-Mg-U tracer. JEOPAG/10.82-PRG-56

H.J. Hein, H. Breton: Photometrische Kalibrierung zur Verifizierung von Behältnisfüllständen mittels Tracertechnik. Atomwirtschaft XXVII, 5 (Mai 1982)

H. Breton, H.J. Hein, G. Krieg: Verification of filling levels in tanks by means of the D_2O tracer technique and use of a spectran 677 IR process photometer. ESARDA Bull. No. 3, Nov. 1982

D.H. Smith, R.L. Walker, C.A. Pritchard, J.A. Carter: Evaluation of lutetium for volumetric calibration by isotope dilution mass spectrometry. Anal. Chem. 55, 578 (1983)

P.R. Trincherini, F. Mousty, W. Pfeiffer: Precise determination of Mg and Lu in input accountability tank solutions of nuclear reprocessing plants by IDMS. Report EUR (in press)

E. Michiels, P. De Bièvre: Absolute isotopic composition and the atomic weight of a natural sample of lithium. Int. J. Mass. Spec. Ion Phys. 49, 265 (1983)

Reprocessing Input Tank Calibration Exercise (RITCEX)

F. Franssen
CEN/SCK Mol, Belgium

Introduction

The ESARDA Working Group on Reprocessing Input Verification stated in its May 1983 meeting at Versailles that "significant improvements in precision and accuracy of uranium and plutonium destructive measurements have been observed during the last few years".

In the Reprocessing Plant, where the Nuclear Material Accountancy is based on the "volume times concentration" method, the need exists therefore to re-assess the precision and accuracy of the volume determinations.

A first move has been made in this direction with the Reprocessing Input Tank Calibration EXercise, RITCEX.

The Belgian Nuclear Research Centre, SCK/CEN at Mol, Belgium, conducted this exercise during the month of January 1984, at the facility of EUROCHEMIC's Reprocessing Plant at Mol, Belgium, with the assistance of the staff members of the plant.

The exercise is one of the tasks of the Belgian Support Programme to IAEA-Vienna.

Preliminary activities

RITCEX was decided upon during a meeting held at SCK/CEN, Mol on 28th July 1983. Mr. Foggi, JRC-Ispra, attended as the convenor of the ESARDA Working Group on Reprocessing Input Verification, the group on which the problem of volume calibration was raised.

Messrs. H. Arenz and J. Toussaint represented the Inspectorate of Euratom, Luxembourg, while Messrs. C. Beets and F. Franssen, both members of the Safeguards Project of SCK/CEN, Mol, acted respectively as the representative of the Belgian Support Programme and as the mandatory of EUROCHEMIC.

The aims of the exercise were discussed and, in view of the world-wide interest on the subject, it was decided to inform all other representatives of National Support Programmes to the IAEA about the exercise. Any possible contribution to RITCEX from any institute or country would be gladly accepted.

The response to this invitation was overwhelming as can be judged from the list of participants given in table 1.

The general information meeting, held at SCK/CEN, Mol from 13th to 15th October 1983, was attended by 37 interested persons.

The aims of the exercise and the time schedule were thoroughly discussed. In order to proceed in detail with all subjects, three groups of special interest were formed :

- the group of classical incremental weight calibration procedure.
- the group involved with tracer techniques of calibration and finally
- the group dealing with the evaluations of calibration data.

The experiment was first scheduled during the month of November 1983, but after consultation within the groups and with representatives of EUROCHEMIC the month of January was agreed upon. This permitted the utilization of the RUSKA electromano-meter of the IAEA and also of the Time Domain Reflectometer, TDR, of ENEA (Italy) and allowed EUROCHEMIC more time to thoroughly prepare the exercise.

Aims of the exercise

The exercise was intended to provide ultimately a contribution to the improvement of the input volume determination in a reprocessing plant.

The tank calibration is of prime importance for the achievements of this goal. Several individual steps influence the uncertainty of the tank calibration. Each of these steps must be evaluated thoroughly in order to limit or at least to estimate its contribution to the total uncertainty of the calibration.

The primary goal, however, was to detect and subsequently to eliminate any systematic error.

During the exercise, several instruments and procedures were compared under real process conditions.

General view in the transmitter galery

Two scale fabricators, TOLEDO and WIDRA, installed their weighing instruments with digital readers. They were used for weighing the increments to be added for calibrating the tank. The most suitable approach for calibration of these scales, and for estimation of their accuracy and precision was sought.

Several measurement instruments were installed to evaluate the liquid depth in the tank. A comparison will be made of the response of the original EUROCHEMIC U-tube manometers, to ENEA's TDR, to SCK/CEN's acoustic system and finally to the RUSKA electromanometer system of the IAEA, already proven in TASTEX. In addition, two out of four classical incremental calibration runs will be compared with the tracer techniques, applied in this experiment, using different tracers.

The tank used for this exercise was decontaminated so that it was accessible for insertion of direct feed lines. Therefore the four runs carried out can be considered as direct calibrations. Some tests, however, were done to simulate remote calibrations. All available data and the experience of the participants will be employed to describe the best suited procedures for calibration or recalibration, of a vessel and to determine the best methods to evaluate the collected data.

The last aim of the exercise is to combine all ideas and experience, resulting in the design of the ideal input tank, including its form, instrumentation and operation procedures, from the point of view of safeguards.

Activities performed until end January 1984

The minutes of the general information meeting, including the finalized time schedule, were mailed to the interested persons on 27th October 1983.
The extended international co-operation to the exercise requires strict adherence to that time schedule.

The month of November 1983 was almost entirely devoted to the calibration of the TOLEDO and WIDRA scales. Besides calibrations with the standard weights provided by the scale fabricators, standard weights owned by Franco-Belge de Fabrication de Combustibles (FBFC), were also used. These weights were calibrated by the Belgian Ministry of Economic Affais. An agent of this Ministry assisted at one of these scale calibrations and stated that the requirements implemented by RITCEX greatly exceeded those of commercial applications of the scales. Additional standard weights were provided by JRC-Ispra . These weights were calibrated by the Eichamt of Bremen (FRG), and are used in an interlaboratory experiment all over

Table 1 - List of Institutions and Companies Interested in RITCEX and Participating People

Berkel, Brussels, Belgium:
. Janssens
J. Nobels

BNFL, Risley, U.K.:
R. Howsley
R. Marsh

Brookhaven Nat. Lab., New York, U.S.A.:
L. Green
L. Solem

CEA, CEN Cadarache, France:
M. Neuilly

CEC, CBNM, Geel, Belgium:
P. De Bièvre

CEC, DCS, Luxembourg:
H. Arenz
G. Busca
G. Meyers
J. Toussaint
E. Van Der Stijl

CEC, JRC-Ispra, Italy:
C. Foggi
G. Guzzi
H. Muntau
R. Trincherini

ECN, Petten, The Netherlands :
W. Zijp

ENEA, Casaccia, Italy:
M. Aparo
G. Bardone
F. Bellisario

ENEA, EUREX, Saluggia, Italy:
S. Ilardi
W. Marsotto
B. Mattia
V. Pagliai

ENEA, ITREC, Policoro, Italy:
G. Arcuri
M. Bianchi
S. Miglietta
V. Morano

EUROCHEMIC, Dessel, Belgium:
M. Ascani
A. De Bie
L. Geens
C. Gevers
H. Van Bijlen
I. Van De Ven
F. Van Es
E. Detilleux
F. Dobbels
W. Hild
J. Mertens
J. Van Geel

FBFC, Dessel, Belgium:
P. Boermans

EXXON Idaho, Idaho Falls, U.S.A.:
F. Cartan
W. Harris

IAEA, Vienna, Austria:
W. Franzel
K. Gharwal
G. Hough
A. Ramalho
H. Shimojima
S. Suda
L. Thorne
D. Thurman
A. Von Baeckmann

KfK, Karlsruhe, F.R. Germany:
U. Knapp
D. Sellinschegg

Min. Econ. Aff., Brussels, Belgium:
. Bairy
E. Befahy
. France

Min. of Foreign Affairs, Brussels, Belgium
S. Herpels

UKAEA, NMACT, Harwell, U.K.
A. Wain

PNC, Tokai, Japan:
K. Iwasaki
Y. Murakami
T. Uchida
O. Yamamura

SCK/CEN, Mol, Belgium:
C. Beets
P. Bemelmans
R. Carchon
J. Challe
J. De Backer
M. De Bie
P. De Regge
F. Franssen
G. Stiennon
F. Ven

THORP Repr. Techn., Sellafied, U.K.:
J. Hall

TOLEDO, Beersel, Belgium:
L. Baartmans
E. De Koninck

TOLEDO-Europe, Köln, F.R. Germany:
L. Richter
K. Rieck
G. Wienecke

WAK, Karlsruhe, F.R. Germany:
R. Berg
H. Hein
B. Stojanik

WIDRA, Eupen, Belgium:
F. Dalimier

Table 2

NR	EVENT - OPERATION	JANUARY 1984	
		1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31	
0	SAT SUN HOLYDAYS		
1	INSTALLATION RUSKA/U-TUBES		W. HARRIS-F. CARTAN-K. GHARWAL - EUROCH.
2	ADAPTATION SOFTWARE		W. HARRIS-F. CARTAN-K. GHARWAL
3	F.B.F.C. WEIGHTS TO EUROCHEM.		VAN HOOF-BOONS
4	CALIBRATION TOLEDO		VAN HOOF-BOONS-SELLINSCHEGG
5	INTER INSTRUMENTS TEST		GHARWAL-CARTAN - EUROCHEM.
6	INSTALLATION FEED TUBES		VAN DAEL-FRANSSEN
7	INSTALLATION ACOUSTIC SYS.		VAN DAEL-DE REGGE
8	HUMIDIFICATION FEED TUBES		VAN LOO-FRANSSEN
9	CALIBRATION RUN # 11		SEE PARTICIPANTS LIST (10 PERSONS)
10	CALIBRATION TOLEDO		VAN HOOF-BOONS-ARCURI-BIANCHI
11	EVALUATION RUN # 11 (PROVIS.)		SELLINSCHEGG-ARENZ
12	ARRIVAL TDR PROBE		TRANSPORT FIRM
13	ARRIVAL TDR HARDWARE		BELLISARIO-SCKCAR - (CUSTOMS)
14	CALIBRATION RUN # 12		SEE PARTICIPANTS LIST (12 PERSONS)
15	ADAPT MEASUREMENT PROGRAM		GHARWAL-CARTAN
16	CALIBRATION TOLEDO		VAN HOOF-HUYSMANS-FRANSSEN
17	EVALUATION RUN # 12 (PROVIS.)		SHIMOJIMA-UCHIDA-MURAKAMI
18	INSTALLATION TDR		VAN DAEL-APARO-BELLISARIO
19	INSTALLATION SAMPLING/TEMP.		VAN DAEL-DE REGGE-VAN ES
20	CALIBRATION RUN # 13		SEE PARTICIPANTS LIST (14 PERSONS)
21	ADAPT SOFTWARE (70 INCR.TS)		SHIMOJIMA-CARTAN
22	TESTING TDR		BARDONE-APARO-BELLISARIO-MATTIA
23	CALIBRATION RUN # 14		SEE PARTICIPANTS LIST (15 PERSONS)
24	ADDITIONAL TESTS		CARTAN-TOUSSAINT-BARDONE
25	DATA COLLECTION DISTRIBUT.		FRANSSEN
26	EVALUATIONS ALL RUNS		SHIMOJIMA
27	CALIBRATION TOLEDO		VAN HOOF-FRANSSEN
28	F.B.F.C. WEIGHTS TO F.B.F.C.		VAN HOOF
29	EVALUATION SCALE CALIBRAT.		FRANSSEN
30			
31			
32			
33			
34			
35	SAMPLING TRACER TECHN.		DE REGGE

EUROCHEMIC STAFF : (P.O. DI VANDEVEN - ASCANI - VAN DAEL - VAN LOO (INSTR) VAN ES - DE BIE - GEVERS - BENNICK

Europe, in order to get an estimate of the precision and accuracy of the scales used in LEU fabrication plants.

Due to the holiday periods in the month of December 1983, only some administrative work on the organization of the exercise could be accomplished. However, some important tests were done, such as tests on the humidification of the feed lines and more scale calibrations. A first test on the RUSKA electromanometer was performed by the IAEA in the presence of the RITCEX co-ordinator.

On the 20th and 21st December 1983, a meeting was held at SCK/CEN, Mol, on the evaluation of historical data of six calibration runs of the tank concerned.

Some important decisions were taken during the meeting, attended by eight persons including three specialists in statistics and members of the ESARDA Working Group on Mathematics and Statistics :

- the number and the size of the increments for future calibrations were fixed,
- the distribution of the raw data was discussed and the methods for correction of these data were decided.

Finally, during the month of January 1984, four calibration runs were performed on the former input tank of EUROCHEMIC according to the time schedule presented in table 2.

A total of 31 persons, representing 6 nationalities and 12 different institutes, contributed actively to the exercise.

The raw data from all calibration runs were distributed among the participants and the evaluation of these data has been started. Provisional results are expected before the middle of March 1984 and a general meeting to discuss these results will be held at Mol in the week of the 7th to 11th May 1984. After finalising the results, a report will be prepared.

At that time, the final phase of the exercise, namely the design of the ideal input tank will be initiated. Final reporting of the whole exercise is scheduled to be finished by Autumn 1984.

Aberration diagnosis for a weighing scale system

F. Brunelli, L. Olivi
CEC, JRC-Ispra (Italy)

Introduction

In the context of the LEU ESARDA Working Group, it has been originated the idea to perform a comparative exercise in order to establish general and easy procedures of weighing. Such procedures should enable the users to control the accuracy and the precision of their scale system, without making use of expensive calibration procedures. Following this idea two estimation procedures have been proposed.

In a previous work /1/, Argentesi and Canali proposed the use of constrained regression to estimate both the accuracy and the precision of scale systems. The proposed estimator is applied to a simple model to estimate casual environment disturbances and systematic effects of rounding errors and biases on scales.

Residual analysis provides quite a good criterion for singling out aberrant instruments. However, the authors remark that the method is adequate even with random disturbances and systematic deviances affecting various scales, if "important violations of the model can be excluded".

A second more recent work /2/ has proposed an iterative procedure in order to remove such deficiencies. It enables us to exploit the information obtained from the comparison of the estimates of the variances obtained in both the unconstrained and constrained procedures. This improves the robustness of the estimator with respect to the unadequacy of the modelling and provides a more effective tool for the diagnosis of aberrations.

This paper provides a simplified description of such a method, without going through the complex algebra needed for its development, mainly to illustrate its use for safeguards purposes.

The model

The basic assumption for modelling scale behaviour is that the random disturbances, rounding errors and systematic distortions contribute as additive terms to the weighing measures. The corresponding statistical model is, therefore, given by:

$$y_{ij} = \mu_i + b_j + u_{ij} \tag{1}$$

If j indicates the j-th scale and i indicates the i-th measure referred to it, y_{ij} represents the reading of a weighing measure, μ_i the true unknown value of the weight, u_{ij} a random cumulative disturbance term due to both the background and the random contribution of the rounding error and b_j the systematic cumulative contribution of both the rounding error and the inaccuracy bias.

The u_{ij} will be assumed as independent and identically distributed (i.i.d.) with a normal distribution $N(O_j, \sigma^2_j)$.

Measurement procedure and observation campaign

Optimality conditions suggest that the observations should be performed in correspondence with all the possible combinations of a limited number of weights, where at least one is known as standard, and to perform the same observation set on all the scales. In addition, the proposed procedure enables one to profit from the information on the equality among the true values of the weights corresponding to the same observation performed by different scales and of the knowledge of the true values of the standards.

This is obtained by expressing the model (1) in the form of a multiple linear regression, where the equality of the weights and the values of the standards are taken into account as constraints on the unknown regression parameters. In this condition there exists a classical solution /3/, which provides the estimates of such unknown parameters (weights estimates), of their variances and the variance of the error. This estimation procedure is called constrained regression and can be applied as long as the model /1/ holds.

Departure from the model of additive error

The model (1) describes a scale behaviour as if every possible error, either of a systematic or of a random nature, which affects the reading of the weighing measure, contributes only as an additive term. In this case one can assume that for every j-th scale the true value of the weight for the i-th measure does not change; i.e. μ_i is equal for every scale and at least one of them has a defined value. In reality, the true behaviour of a scale is unknown and in a more general way can be described as:

$$y_{ij} = \mu_i + \sum_{k=0}^{\infty} b_{jk}\, \mu_i^k + \mu_{ij} \tag{2}$$

This model considers as purely additive the random disturbance alone, whilst it describes the systematic bias as a function of the true weight, depending upon the single scale.

Hence, viewed fron the point of view of constrained linear regression modelling, the contributions of the true weight and of the bias become indistinguishable. Therefore the larger the contribution of sum terms with $k > 0$, the less correct are the estimates. Nevertheless, residual analysis can show whether there are heavy aberrations or not, but it cannot identify which scales are aberrant. This is because **the same systematic violation of the model (1), by several scales introduces a definite level of distortion which, because of the constraints, affects the estimates for all the scales of the set.**

The iterative procedure

If one can assume that bias terms of order larger than one are negligible ($b_{jk} = 0$, for $k > 1$), a significant correction can be introduced in the estimation procedure and also the aberration of the first order evaluated (multiplicative bias). In this case, the constrained estimates are still biased, while the estimates of variances obtained for

each scale by the usual linear multiple regression (i.e. without profiting from the knowledge of the equality constraints and of the standards) are unbiased. Therefore the difference between the variances obtained in the two procedures provides an index of the accordance of the behaviour of a single scale to the model (1).

This quantity is used for an interative estimation procedure, like the weighted least square, in such a way as to identify those scales which present a significant aberration. This method can also be applied during an ordinary measurement campaign, if a known standard weight can be added to the set to be measured. This enables the operators to verify periodically the efficiency of their instruments, without the need for separate recalibration campaigns.

Application in simulation tests

Two simulation exercises have been performed where observation data have been generated according to a linear varying bias model:

$$y_{ij} = (1+b_j)\mu_j + a_j + u_{ij} \qquad (3)$$

where y_{ij} is the simulated reading of a weighing measure, b_j is the multiplicative term of the bias, a_j is the constant bias contribution, μ_j is the true weight value, u_{ij} is an atificial random disturbance with a distribution $N(O, \sigma_j^2)$.

Table 1 shows the comparison of the estimates with the true values, for the first estimation step and for the last step.

The five scales at the end of the iteration are grouped into two classes of different behaviour; the application of the iterative estimator causes the estimates to approach the real simulation values remarkably. The most meaningful result is a precision gain in the estimate of the slope of the residuals, which is a consequence of the whitening of the residuals of the scales conforming the constant bias model.

As is shown in Table 1, scale 3, which has a "true" slope equal to 1, presents an apparent anomaly in the first estimation step, whilst in the last its residuals are practically whitened. Such behaviour could induce us to divide the scales into groups; the first

three scales present a slope of a value which approaches the true value, the others have a slope with a small but not negligible distorsion as in the reality.

One can also remark that the error variance estimate provided by the constrained estimation is increased for the aberrant scales, as expected, because of the overload due to the non constant bias.

Final comments

The proposed method is now object of a circular experiment within the framework of the LEU ESARDA working group.

All the experimenters arrange the experimental design (i.e. how to assemble and distribute the k available weights in the $2^k - 1$ weighing measures) in a common way, and use one known standard weight as well as in the simulated tests.

It is expected to verify the effectiveness of the system of scales up to appreciate systematic errors.

References

1. F. Argentesi, S. Canali: L'uso di regressioni vincolate per la stima dell'accuratezza e della precisione delle bilance. Rivista di statistica applicata, Vol. 15, N° 3 (1982)

2. F. Brunelli, L. Olivi: Weight estimation and aberration diagnosis for a weighing scale system. EUR 8831 EN (1983)

3. D.S.G. Pollock: The Algebra of Econometrics, p. 150, Whiley, London (1979)

TAB. 1

SIMULATION VALUES		ESTIMATES		
		FIRST		**AFTER 3 ITERATIONS**
SCALE	BIAS AVERAGE	BIAS AVERAGE		BIAS AVERAGE
1	— 15	— 21.2		— 17.9
2	31	27.3		30.5
3	— 30	— 38.2		— 34.9
4	35	20.9		24.1
5	10	1.4		4.6
	SQM	SQM	SQM NOT CONSTRAINED	SQM
1	22	22.9	22.3	22.5
2	33	34.5	33.8	34.3
3	25	27.9	26.4	27.4
4	28	31.7	31.0	32.4
5	26	26.9	24.5	28.1
	SLOPE $(1+b_j)$	SLOPE $(1+b_j)$		SLOPE $(1+b_j)$
1	1.0	0.99988		0.99998
2	1.0	0.99985		0.99996
3	1.0	0.99967		0.99977
4	1.0003	1.00019		1.00029
5	1.0005	1.00041		1.00051

ESARDA Meeting on Research Centers' Accounting

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At an ad hoch ESARDA meeting on research Centers' accounting, Karlsruhe, 1-2 December 1983, experts presented papers on accounting and reporting aspects, followed by a demonstration of the Karlsruhe computer accounting system.

Various topics were then selected and discussed. Among these were relations to Euratom in Luxembourg, inspectors, external MBAs, Euratom Supply Agency, and authorities. As regards the safeguards administration at the centers the group discussed centralised and decentralised preparation of reports, use of computers, physical inventory takings, rounding adjustments, consolidation, decimals, obligations, waste accountancy, criticality control, etc.

With respect to reporting to Euratom H. Kschwendt, Euratom, encouraged reporting in computer readable form. This would lead to advantages not only for Euratom, but also for the operators. He recommended that installations planning such reporting should contact Euratom in order to ensure that the development would proceed in the right direction. (See also the article : Advantages of Reporting in Computer Readable Form, by H. Kschwendt, here) The group identified possibilities ranging from a general safeguards reporting system operated on a word processor and reporting on diskettes to comprehensive multipurpose computer accounting systems reporting on tape.

H. Kschwendt recommended that the discussion should not be limited to research centers. A similar discussion on aspects at other types of nuclear installations could be useful if not already covered in plant specific working groups. The participants will evaluate the possibilities for future co-operation within the group.

Advantages of Reporting in Computer Readable Form

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Out of over 500 installations within the European Communities which deal with nuclear materials, approximately 220 have a sufficient stock of material and a sufficient number of movements to warrant a computerized internal nuclear materials accounting system. European Community Regulation 3227/76, which requires the operators of installations to send regular accounting reports (ICRs, PILs, MBRs) to the Euratom Safeguards Directorate, allows for these reports to be in computer-readable form (note 4 on page 40 of the Regulation) and the corresponding report forms (annexes II, III and IV) were designed to facilitate the production of reports by the operators' own computer system. To date, 90 installations already send their reports in computer-readable form on tape or diskette (floppy disk) and another 30 installations will do so in the near future.

The advantages which have resulted from this development should encourage other operators to follow this trend. It has been observed that :
- The cost, effort and time needed to produce the reports are all significantly reduced;
- Reporting deadlines can more easily be adhered to. This permits the inspectors to have more up to date information available when carrying out accounting verifications, which is in everyone's interest;
- Communication on magnetic tape or diskette is less error prone (avoidance of transcription errors on the part of the operators and avoidance of key-punching errors on the part of the Euratom Safeguards Directorate).

The following transmission media may be used at present : 9 track (1/2") tapes with a density of 1600, 6250 or 800 bytes per inch and EBCDIC or ASCII coding, as well as 8" and 5 1/4" diskettes. It is, however, emphasized that reports should not be made in form of computer printouts as data entry is more difficult (and therefore more error prone) than when made from the report forms of the Regulation. From the software development point of view, the operator will also normally find it simpler to produce reports on tape or diskette than as a printout.

The detailed provisions concerning the production and dispatch of computer readable reports are contained in the Reference mentioned below. Apart from the accompanying letter, no additional papers, listings, etc. are required. The Reference also discusses related issues such as the definition of the book inventory, the adjustment of the book inventory to the physical reality, the calculation of the MBR and the correction mechanism. A further paper which will be presented at the 6th ESARDA Symposium in May 1984 in Venice describes other features of a computerized internal accounting system which are related to the carrying out of inspections.

All operators who are interested in computerizing their accountancy systems are invited to consult the Reference and to contact the author of this article for further discussions and clarifications. It is essential that the operators are familiar with the details of the requirements **before** any development work is begun. In these discussions, practical arrangements may be made for the return of the tapes or diskettes and for the period of transition which may involve parallel reporting for a few months in both manual and computer readable form. This gives the opportunity to the Euratom Safeguards Directorate whose agreement is necessary for reports to be made in computer readable form to verify the Safeguards parts of the computerized system and the assurance for the operator that the reports are produced in the correct manner.

Reference

H. KSCHWENDT, M. LITTLEJOHN, "Specific Questions Concerning Reporting by Computer", Safeguards and Nuclear Material Management, Proc. Symp., Versailles, 1983, Euratom-ESARDA-16 Ispra (1983), pp. 509-512

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Activities of the ESARDA Working Groups

Techniques and Standards for Non-Destructive Analysis

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Introduction

In the ESARDA Bulletin No. 1, of October 1981 a short report has been given of the activities of the ESARDA Working Group on Techniques and Standards for Non-Destructive Analysis (NDA-working goup). In 1982 the work of the NDA-working group was presented at the Fifth International Conference on Non-Destructive Evaluation in the Nuclear Fuel Industry, San Diego, 10-13 May 1982, under the title Recent Activities of the ESARDA Working Group on NDA. This report is published in the proceedings of the conference by the American Society for Metals, Ohio 44073, U.S.A. In this article the developments since that publication will be reviewed, like they were presented to the ESARDA-Board on 22 November 1983.

Objective and aims

The objective and aims of the NDA-working group that served since 1976, have been revised by the ESARDA Board (the most recent publication of the terms of reference of all ESARDA working groups can be found in ESARDA Bulletin No. 4 of April 1983, as appendix to the paper "Cooperative Development of Safeguards within the ESARDA" by W.L. Zijp, presented also at the 1982 safeguards symposium of the IAEA). The objective is now formulated as : To evaluate and recommend criteria for non-destructive analysis of nuclear materials for use by plant operators and the Safeguarding Authorities.

The aims have been replaced by the present activities which are continuously changing according to the progress made. They will be dealt with in the following part of this article.

Membership

The membership of the NDA-working group consists of 13 persons representing 10 institutions while there are 3 observers from outside ESARDA. The secretariat of the working group changed in summer 1982 from Dr. A. Adamson of UKAEA-NMACT at

Harwell to Dr. B. Chauvenet of CEA-LMRI at Saclay.

No plant operators are permanently engaged in the NDA-working group; however from time to time operator representatives participate for some specific topics, like also other specialists do. In the last Board meeting where this report was discussed, it is also proposed to have combined meetings with plant specific working groups like the MOX and LEU fuel fabrication working groups.

Working method and meetings

The working group meets normally twice a year. Sometimes a special topic is dealt with in an ad-hoc specialist meeting.

From time to time the programmes on NDA research of the partners are reviewed. The last time this was done at the occasion of the NDA-working group meeting at Dublin. The group visited Ireland on the invitation of Dr. N.V. Nowlan of the Irish Nuclear Energy Board, in order to give also a presentation of the highlights of the programmes to the Irish scientific community members that are interested in the subject of NDA.

The following meetings have been convened :

22/24 September 1981
Luxembourg, the meeting included a combined session with the DA-working group, and an ad-hoc specialist meeting on neutron coincidence counting.

23/24 March 1982
Dublin, this meeting included a special session presenting the NDA-working group activities to the representatives of the Irish Universities.

4/5 November 1982
Cancelled.

8/10 February 1983
Paris-Saclay, this meeting included final discussions on the preparation of PIDIE (Plutonium Isotopic Determination Intercomparison Exercise).

14/15 September 1983
Mol, this meeting included a visit to the CBNM Laboratories, to see the preparation of the Certified Reference Material for gamma-spectrometric determination of the enrichment.

The working method of the group is to concentrate on a few selected tasks, which are of common interest, and to which several partners can contribute. For every task one participant assumes the responsibility to collect the information, to prepare the execution and to guide the task to the final stage of publication of the results. Sometimes it is necessary to split the task in some subtasks.

The NDA WG of ESARDA being guest of Dr. N.V. Nowlan at the University College, Dublin, 24 March 1982

Enrichment standards for gamma-ray spectrometry

This project goes beyond the limited scope of the normal NDA-working group activities, out of which it originates. The most important result of the project is that in cooperation between CBNM and the US NBS sets of Certified Reference Material (CRM) have been produced, that enable enrichment measurements with gamma-spectrometry to be calibrated with an accuracy of about 0.1% relative; which is about ten times more accurate than the 1.5% relative uncertainty in the gamma-emission probability of the 185.7 keV from 235-U, which error dominated previously the uncertainty in this kind of measurements. This performance target is also about the best that can be achieved by mass-spectrometry today. The enrichment measurements become directly traceable to the basic standards of the International System of Units (SI), which is the officially and internationally recognized basis for all measurements. The CRMs get a special importance both in the U.S.A. and in the European Community countries due to the cooperative Certification by CBNM and the US NBS. Special attention is given to the identification of the CRMs with ultrasonic seals developed at JRC Ispra, supported by EURATOM Safeguards Directorate, which makes them also suited for safeguards use by the IAEA, EURATOM, and the US NRC. The CRMs can soon be purchased from CBNM and the US NBS.

Under the aegis of the NDA-working group a detailed users manual for the CRMs has been written at KfK. If the availability of qualified manpower allows so, this work can be used in the future to make a procedural standard for gamma-spectrometric enrichment determination.

The project will be described in more detail in a paper at the ESARDA-symposium in Venice, 14/18 May 1984.

The international project for Pu-isotopic CRMs for DA and NDA

Taking the previously described project of CRMs for uranium isotopic composition as example, a similar cooperative action in the field of plutonium isotopic composition was conceived. In that international project also the Japanese organisations JAERI and PNC were invited to participate. The main impetus of the project comes from the needs for mass-spectrometry purposes, to have well characterized CRMs for isotopic abundance measurements of the different isotopic compositions, that correspond to the different burn-up levels. For the European Community the responsibility for this project lies with the CBNM, while in the U.S.A. the New-Brunswick Laboratory is acting on behalf of the NBS. The definition of the needs in the DA field lies in the hands of the DA-working group; the NDA-working group has tried to formulate its part of the problem in the group's meeting of 14 and 15 September 1983. The position of the NDA-working group can be summarized as follows :

If during inspections neutron coincidence counting or calorimetry is applied to measure the total amount of plutonium, some knowledge of the isotopic composition is necessary for the evaluation of the measurement results. Gamma-spectrometry is applied in order to verify the operator declarations on the isotopic composition, or to determine the isotopic composition independently.

For the accurate determination of the isotopic composition by gamma-spectrometry of different plutonium samples it is imperative that the samples are all brought in the same form and counting geometry. One of the geometries favoured is the infinite thickness, however for the higher photon energies which are used in the practical applications, this condition will involve also larger samples to be handled. In order to get the most reliable results, measurements are to be calibrated with carefully characterized CRMs. Identical counting geometries for the calibration samples and the samples under investigation have to be realized. This will involve a sample preparation with plutonium handling, which effort can likewise be spent on the taking of a sample for mass-spectrometry analysis and measurement by that technique, of which the results are at this moment of a better quality than those of gamma-spectrometry.

As a first conclusion it may be stated that the present status of the gamma-spectrometry does not yet justify the large effort that is needed for the preparation of isotopic CRMs of Pu. The best accuracies claimed at this moment for gamma-spectrometric determination of isotopic ratios of Pu-isotopes are 1% or worse, but these values are still under investigation.

If for the needs of mass-spectrometry batches of Pu-materials are characterized, it might be prudent to involve larger batches that can later also be used as a supply for the NDA CRMs. Such a measure could make the future NDA-CRMs traceable to the same characterisation work as the DA-CRMs. At this moment it is not possible to give a clear indication of the amount of material necessary for NDA purpose, but it will be significantly more as is needed for mass-spectrometry.

For the improvement of the gamma-spectrometry on Pu it will be useful if samples of various isotopic compositions can be measured. It seems that there are still some arbitrary elements in the spectrum evaluation procedures, and also the ingrowth of 241-Am poses some particular problems. The PIDIE experiment, which aims at the distribution of samples with seven different isotopic compositions to various laboratories, as well as the CBNM pilot sample project will help to find answers to the still unresolved questions in the near future. When these samples have been measured and the results have been evaluated, it will be more clear if it is justified to ask for Pu-isotopic CRMs for NDA.

The PIDIE project and CBNM's pilot samples

The Plutonium Isotopic Determination Intercomparison Exercise, PIDIE, started with a modest proposal to distribute sets of plutonium samples of about 0.5 g of 7 different isotopic compositions to about 15 laboratories, that would do gamma-spectrometric analysis and, if possible, also mass spectrometric determinations on the seven samples. The results would give an indication of the capabilities to determine the isotopic composition of Pu by gamma-spectrometry. In the preparation of the project it was decided to give somewhat more thinking to the data gathering and evaluation of the project. Some members of the working group have defined the questionnaire and the information to the participants, the comparison of the results will be done at the CEA-LMRI. The preparation of the samples at Harwell is finalized and the samples will be dispatched to the participating laboratories quite soon. Also the CBNM has announced that it will make available Pu-samples. These CBNM Pilot Samples are designed to offer an infinite thickness for the gamma-radiation below 200 keV, which means that the samples will contain several grams of Pu.

The experience gained with the various samples will be useful to assess the performance of the gamma-spectrometry and also it may give useful information for the design of CRMs if these are needed in the future.

Intercomparison of NDA on kg size Pu-samples

In the previous report of the working group it was mentioned that some inter-comparisons were done with gamma-spectrometry and neutron measurements at the Belgonucléaire plant in Dessel, Belgium. The reporting on this subject should be done by ECN, but due to the allocation of manpower to other subjects of more urgency to ECN this work is not yet finished. In the meantime other experiments with similar kind of samples are performed by KfK at the ALKEM facility where, just like with the sample at Belgonucléaire, similar

difficulties arose with the inhomogeneity of the isotopic composition in a large fraction of the number of samples.

At Belgonucléaire other measurements have been performed with gamma-spectrometry, that confirm the previously found indications of inhomogeneity. Further also calorimetric measurements are performed in a wide international collaboration, the results will be presented in a paper at the ESARDA symposium in Venice, 14/18 May 1984.

Special subjects

Neutron coincidence counting has been subject of a critical review, specially with regard to the treatment of the signals and the procedure to calculate the mass of fissile material from these signals. On 24 September 1981 the NDA-working group had an ad-hoc meeting with specialists to discuss the subject. Some progress has been made that justifies another ad-hoc meeting on the subject, that will be held adjacent to the NDA-working group meeting at Harwell on 8 May 1984.

In a combined meeting with the DA-working group special attention was given to the K-edge and the L-edge densitometry. The DA-working group presented their work on interlaboratory comparison evaluations and the operator / verifier differences, which subject is closely related with the list of target values (this subject is dealt with in another article in the Bulletin by the convener of the DA-working group). From time to time it is mentioned that also the NDA-working group should try to establish target performance data of NDA-techniques. The measurement procedures are however less clearly defined, which makes the situation more complicated to describe in a simple way the performance that can be obtained.

The US DOE New Brunswick Laboratory has organized an intercomparison on the NDA determination of 235-U in simulated waste samples that are sent around. Two members of the NDA-working group have expressed their intent to participate. The samples will be in Europe soon.

The ESARDA working group on Mathematical and Statistical Problems is planning to organise in cooperation with the INMM a workshop on problems related with NDA. Several members of the NDA-working group will contribute to that meeting in Ispra in autumn of this year.

Conclusion

With the cooperative production and certification by CBNM and the US NBS of the 235-U Isotope Abundance CRM for gamma spectrometry and the related work like the writing of a manual, one of the first tasks of the NDA-working group is approaching its final accomplishment. In the field of Pu isotopic Abundance CRMs the situation for NDA is not yet clear but the NDA-working group is progressing steadily to answer the still open questions. In the field of neutron coincidence counting progress is continuing, while also some in plant testing of this and other techniques is progressing. Like the ESARDA-Board emphasised in its meeting of 22 November 1983, it is important that the future work of the NDA-working group should be brought in closer relation with the plant specific working groups of ESARDA.

Achievements of the ESARDA LEU WG

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A.A. Musto
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Introduction

The ESARDA LEU WG was founded in 1978 and represented a new departure for ESARDA since it was intended that the prime input should come from plant operators. The Group met on seven occasions in the period March 1978 to March 1980 after which it was decided that they should recess to allow development work to progress. The Group reconvened at BNFL Springfields in September 1982 and adopted an action plan for a second round of meetings which reaffirmed the commitment to consider the practical problems of safeguards for both operator and safeguards inspectorate. The most recent meeting, the 10th, was hosted by F.N. at Bosco Marengo. During the current round of discussions, meetings have been attended by a total of 26 people representing 6 plant operators, 6 research centres and the Euratom Safeguards Directorate.

The Group provides a forum in which the Euratom Safeguards Directorate can explain their needs and the rasons behind those needs and in which the practical problems of meeting these requirements can be discussed. The discussions have included the possible safeguards use of systems implemented by operators for their own nuclear material accountancy. Recognising that safeguards approaches would always be to a large extent plant specific, the Group discussed approaches in a sufficiently general way to be of interest and help to all participants. Ways in which the safeguards objectives may be met at minimal cost and with minimal interference to existing systems by building on those systems may be discussed without commitment on either side. Through such free-ranging discussions and exchanges of information the Group has kept up to date with the latest advances in computerised materials accounting systems and in measurement technology. The Group has been able to identify areas where further work was needed and has organised inter-site comparison exercises to improve understanding of certain techniques.

The work of the Group has been reported on three occasions in the past: to the INMM Symposium in Albuquerque in 1979 /1/; to the ESARDA Symposium in Edinburgh in 1980 /2/; and to the ESARDA Symposium in Versailles in 1983 /3/. This paper brings together brief outlines of the major achievements of the Group (below); it is not intended to be a full report on each of the topics, some of which have been reported elsewhere.

The Specific Achievements of the Group

Experimental use of the Neutron Collar to verify LWR fuel assemblies

The Neutron Collar is a special design of neutron coincidence counter developed at Los Alamos laboratories for the verification of fresh fuel assemblies /4/. During the first round of meetings the Group discussed ways of safeguarding the assembly area of LEU plants and recognised the potential value of a fuel assembly verification measurement device. The instrument has been tested in the presence of Euratom safeguards inspectors at FBFC Dessel as part of the Belgian support programme to the IAEA /5,6/. The Neutron Collar has also been used by Euratom safeguards inspectors at a number of reactor sites and LEU fabrication plants in the Community.

The experience gained from using the instrument was discussed by the Group and, in particular, the practical problems of use for safeguards verification in an operating plant were addressed.

The main parts noted were as follows:
- to some extent the use of the instrument will be fuel and plant specific in terms of calibration and of the physical constraints imposed by the design of the fuel store;
- agreements will be required to comply with site licences, transport regulations and covering liability in the event of damage to finished fuel;
- there may be limitations of use with multi-enrichment fuel or with fuel containing burnable poisons;
- the instrument proved to be reliable and robust under plant operating conditions using a measurement which only lasts a short time.

The Group concluded that the active Neutron Collar promises to be a useful measurement technique for flow and inventory verification provided that the administrative problems of usage can be

overcome and that the instrument can be adapted for specific applications.

Examination of weighscales

The Group discussed weighing and exchanged information on measurement control procedures for weighscales. As a result one operator has reviewed and revised his weighscale purchasing and measurement control policy. During the first round of discussions, the Group concluded that the precision and accuracy of weight measurements were not sufficiently well understood. As a result the Group set up an exercise to gather comparative data from a number of weighscales and JRC Ispra initiated a research programme to define a method of analysis for such data.

The weighscale comparison exercise involved four LEU plant operators in EEC countries, and the JRC Ispra. JRC manufactured the set of seven standard weights for the exercise. These weights were then sent to each operator in turn, who carried out a sequence of 127 weighings of different combinations of the weights on one or more weighscale. The 127 combinations gave a sequence of weights evenly distributed through the range 0-90 kg.

JRC developed a new methodology for the analysis of random and systematic errors in weighing. An initial assessment of the data showed that the error structure is more complex than had previously been believed since bias is not constant. To overcome this problem JRC developed a second method which assumed a model with linear bias. The results of the exercise and the analytical methodology were reported briefly in reference /7/ and in detail in reference /8/. The algorithm for analysis of weighing data is now available for use by the operators in computer tape form.

The comparison exercise is viewed as a success by the Group since it has led to an improved understanding of weighing errors and has allowed operators to assess their position relative to other operators. Further, the exercise was an example of productive cooperation between the operators and a research centre. Members of the Group have, therefore, decided to set up a second round of the weighscale comparison exercise using the same set of standard weights and this is scheduled to start during November 1983.

Examination of Analytical Techniques

The Group has discussed a number of analytical techniques including the "Total Impurities and Stoichiometry Analysis" (TISA) method /9/ for the determination of U in uranium compounds. This method is considered to be highly accurate and suitable for use as a reference method.

Members of the Group have taken part in an inter-laboratory comparison exercise for the determination of 'U' in UO_2. JRC Geel produced a certified reference batch of 10g UO_2 pellets for this exercise and used direct uranium measurements by potentiometry and controlled potential coulometry for the certification. The TISA method was applied for verification purposes. The exercise involved analytical laboratories from all of the European LEU fuel fabricators and from certain Research Centres. The exercise served to confirm the excellence of 'U' in UO_2 determination for pure pellets using the gravimetric method at analytical laboratories throughout Europe. The part of the exercise concerned with production and certification of the pellets is reported in reference /10/.

Development of computerised nuclear materials management systems

The Group agreed upon the usefulness in LEU plants of computerised material management systems containing up-to-date information. Each of the Plant Operators has already implemented a computerised system for the accountancy and management of materials which cover part of all of their LEU activities. There was an exchange of information between the operators concerning the objectives, scope and operating methods and safeguards aspects of their respective systems.

In particular, a near real time nuclear materials management system has been developed and implemented at BNFL Springfields under a Joint Collaboration Contract between BNFL and JRC Ispra and has been reported at the ESARDA Symposia on Nuclear Materials Management in references /11/ and /12/. The project was made possible through the LEU WG and incorporated safeguards objectives within its framework from its very conception. Through this work:

- BNFL have been able to introduce a more comprehensive system for LEU nuclear materials accounting and control than would otherwise have been possible;
- JRC Ispra has gained experience in the workings of and influenced the design of a computerised materials management system for a large plant.
- It is hoped that the Euratom Safeguards Inspectorate will benefit in two ways (i) the system should be easier to audit since consideration was given to simplicity and 'transparency' during the design stages with audit requirements in mind; (ii) the system should produce the Euratom reports (ICR, PIL, MBR, LII) directly and will eventually include an ispector interface package which should help to speed-up and simplify inspections.

In the light of the success of this collaboration project it is hoped that further collaboration exercises will take place between the inspectorate, the JRC and the plant operators. One such exercise currently under discussion is an examination of the alternative techniques available for physical inventory taking which might reduce the requirement for plant shutdown and process clean out.

The application of NUMSAS to plant data

The NUMSAS package (**NU**clear **M**aterial **S**tatistical **A**ccountancy **S**ystem /13/14/), developed by JRC Ispra is a statistical tool for the analysis of materials balance information. The Group expressed interest in this package since the Euratom Safeguards Directorate had indicated that it intended to use the package on a routine basis. Following initial discussions, an exercise was arranged through the Group to apply NUMSAS to plant data from FBFC Dessel.

The Group had noted that the analysis of material balance data at such LEU plants would be difficult without the use of computerised tools because of the volume of data involved. The code is designed to give MUF, the standard deviation of the MUF and, for each primary error source component, the contribution to the overall variance of the material balance.

The Group noted the following main points arising from the experiment:

- the main effort required to implement the code is concerned with collecting the measurement uncertainty data for the primary error sources and to allocate the error paths;

- the definition of systematic error in some cases was necessarily rather arbitrary and the treatment of systematic errors was problematic;

- in the case considered with 10,000 entry lines, there were 65 distinct error paths comprising 36 primary error source components;

- the code is designed to eliminate all correlated batches from the calculation of MUF variance. In this case, it was only possible to eliminate those batches which appeared unchanged on both Physical Inventory Listings.

- the results of the run prompted a review of the error table. The merit of the code and of this exercise was that it highlighted those factors which made the greatest contribution to uncertainty in the material balance.

Evaluation of the potential use of rod-scanner for safeguards purposes

All of the European LEU fuel fabricators have, or soon will have, a rod-scanner installed in their plant for Quality Control

purposes. It has been suggested that it may be possible for safeguards inspectors to make use of the scanners to verify operators' data for finished fuel rods. This suggestion was discussed by the Group and a sub-group of expert representatives of plant operators was set up to examine such a possibility.

The initial findings of the sub-group were reported to the 9th meeting of the Group in June 1983. These were:

- usage of rod-scanners would present different problems in different plants. For example, the rod-scanner is an integral part of the production line in certain plants, hence use for safeguards purposes could, under certain circumstances, interfere with production;

- preparation of standards may present a problem to the safeguards imspectorate, although some standard rods have already been prepared and are in use at certain plants. Given adequate standards, rod-scanners provide a sufficiently accurate total U-235 measurement for verification purposes for standard fuel;

- fuels containing Gadolinium (a burnable poison) present special problems for neutron-activation rod-scanner systems. For these fuels a passive high resolution detection system may be required.

As a result of their discussions the sub-group decided to consider a comparison exercise to evaluate the performance of rod-scanners for measurement of total U-235 in fuels containing impurities (such as U-232 or U-236 which are present in recycled uranium after reprocessing). The sub-group is currently looking for suitalble standard rods for the exercise and is investigating whether the exercise is feasible given the differing geometries of different fuel types.

Other areas of discussion

Other areas which have been discussed by the Group and where there has been a useful exchange of information are as follows:

- sampling techniques

- the use of the PHONID III instrument

- non-destructive analysis of heterogeneous materials.

Conclusions

The Group has provided an excellent forum for an interchange of views between plant operators, research centres and the Euratom Safeguards Directorate. A number of detailed experiments and practical exercises have been organised through the Group giving rise to a better understanding of operational and safeguards problems.

References

1. R. Stewart et al. The ESARDA approach to facility oriented safeguards problems. Proceedings of the 20th Annual Meeting of the INMM, 1979, pp 37 - 47

2. S. Saiger. Activity of ESARDA Working Group on safeguarding low enriched uranium conversion and fabrication plants, Proceedings of the 2nd Annual ESARDA Symposium 1980, ESARDA 11 pp 27 - 34

3. R. Stewart, A.A. Musto. A survey of the interaction between Euratom, the European Research Centres and plant operators in the field of LEU, conversion and fuel fabrication, Proceedings of the 5th Annual ESARDA Symposium 1983, ESARDA 16 pp 339 - 341

4. H.O. Menlove (Los Alamos Scientific Laboratory). Description and performance characteristics for the neutron coincidence collar for the verification of reactor fuel assemblies, LA 8939-MS (ISPO-142), 1981

5. CEN, IAEA, LASL. Optimisation of NDA measurements in field conditions for safeguards purposes, BLG-543, February 1981

6. CEN, IAEA, FBFC, LASL. Optimisation of NDA measurements in field conditions for safeguards purposes, BLG-553, January 1982

7. F. Argentesi, H. Muntau, S. Canali. A standard method for the estimation of scale accuracy and precision: a case study for the evaluation of bias time evolution. Proceedings of the ESARDA Specialist Meeting 1982 (ESARDA 15 pp 113 - 114

8. F. Brunelli, L. Olivi. Weight estimation and aberration diagnosis for a weighing scale system, EUR 8831 EN, 1983

9. J.R. Entwistle. The determination of uranium in UO_2 pellets from measurements of impurities, O/U ratio and U-235, presented to LEU WG, 1982

10. V. Verdingh, H.L. Eschbach, Y. Le Duigou, W. Beyrich. Characterisation of a uranium dioxide reference material for in-plant use, Proceedings of the 5th Annual ESARDA Symposium 1983 (ESARDA 16 pp 197 - 198)

11. F. Argentesi, T. Casilli, S.M. Fensom. Implementation of an on-line material management system in a LEU fabrication plant, Proceedings of the 2nd Annual ESARDA Symposium 1980 (ESARDA 11 pp 208 - 213)

12. F. Argentesi, L. Costantini, T. Casilli, B. Burrows, S.M. Fensom. NUMIS: a real time system for nuclear materials management, Proceedings of the 5th Annual ESARDA Symposium 1983 (ESARDA 16 pp 443 - 446)

13. F. Argentesi, T. Casilli, M. Franklin. NUMSAS: a statistical nuclear materials accountancy system in the Euratom framework, Proceedings of the 1st Annual ESARDA Symposium 1979 (ESARDA 10 pp 388 - 390)

14. F. Argentesi, T. Casilli, M. Franklin. Nuclear material statistical accountancy system, CEC JRC Ispra 1979, EUR 6471 EN

Destructive Techniques and Standards

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The Group has been active in a number of items :

1. It advised frequently on the execution of the IDA-80 programme (Isotope Dilution Analysis) (1) which aims at describing the present state-of-the-practice in isotope ratio and isotope dilution measurements, especially on input samples of reprocessing plants (both U and Pu), amongst the world's nuclear analytical laboratories. More and more evaluation graphs are becoming available from the programme, from the total of minimum 120 expected. Figures 1-3 display some of them.

Figure 1 shows present capability of 30 laboratories to determine U concentration. The small graph displays the difference between calculation of results by the laboratories themselves and by the programme evaluators, both using the same raw data (the difference is due to calculation errors and use of wrong nuclear constants).

Figure 2 shows present capability to determine [235]U abundances.

Figure 3 demonstrates how good Pu concentrations can be measured.

Certified values for all test samples in the programme were established by CBNM and NBS.

2. The Group sponsored a determination of the "1982 State-of-the-practice in low enriched UF_6 isotopic measurements in Western Europe" (2) and Figure 4 displays one of the results of this interlaboratory measurement evaluation programme. With one exception, it demonstrates an astonishingly good agreement to within 0.1%.

3. The Group finished a compilation of "Current Methods for the Destructive Analysis of Nuclear Materials".

4. At each of its meetings, the Group focusses on a new development in measurement techniques e.g. for the period covered by this report : "Analytical Techniques for Recycled Uranium" or "The Use of Quadrupole Mass Spectrometers for Isotopic Analysis of UF_6" or "The Spectrophotometric Determination of PuO_2 + +"
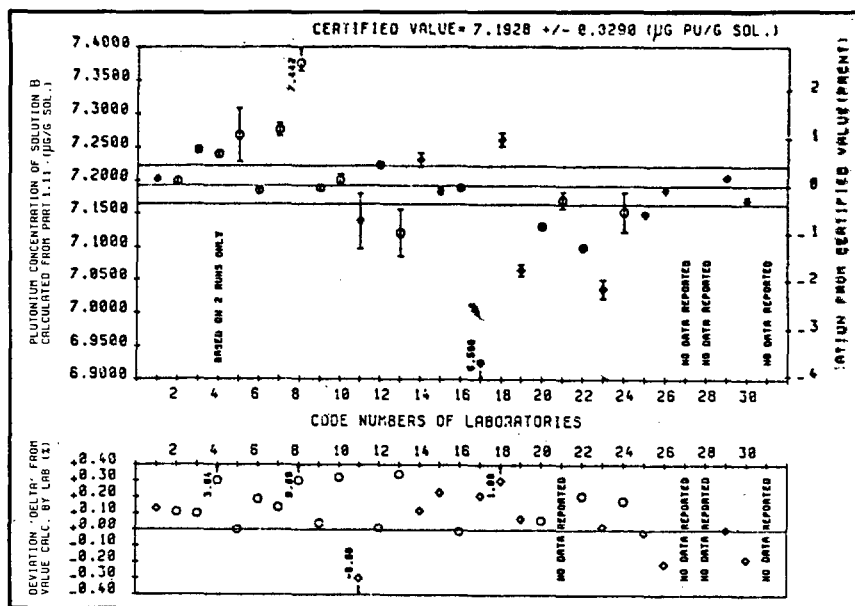
Fig. 1 - IDA-80: Determination of
Uranium Concentration. Solution B.

Legend:

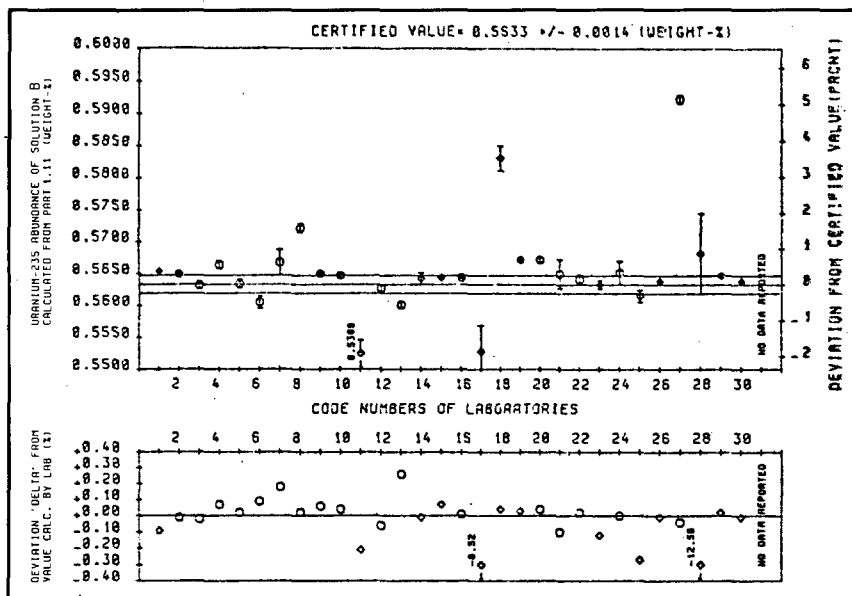O More experienced laboratories
◊ Less experienced laboratories

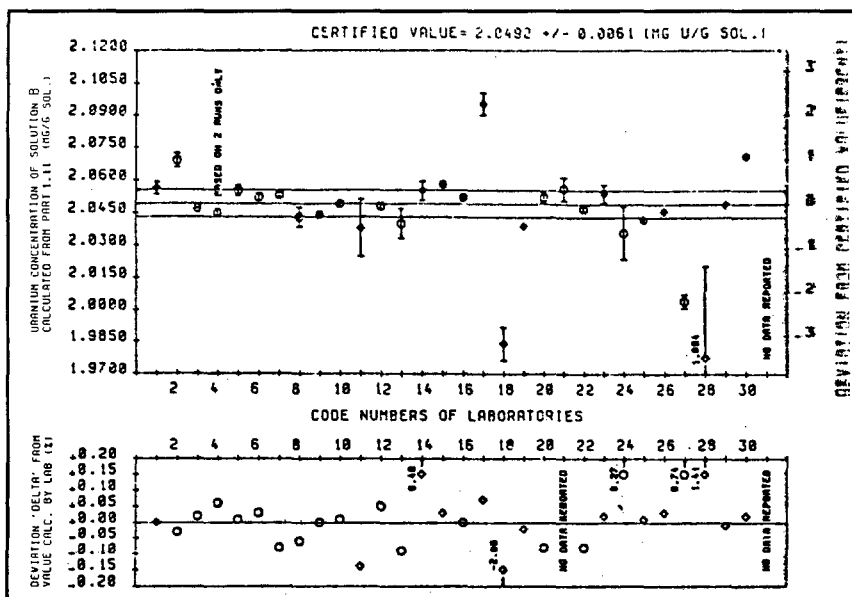Fig. 2 - IDA-80: Determination of
^{235}U Abundance

Fig. 3 - IDA-80: Determination of
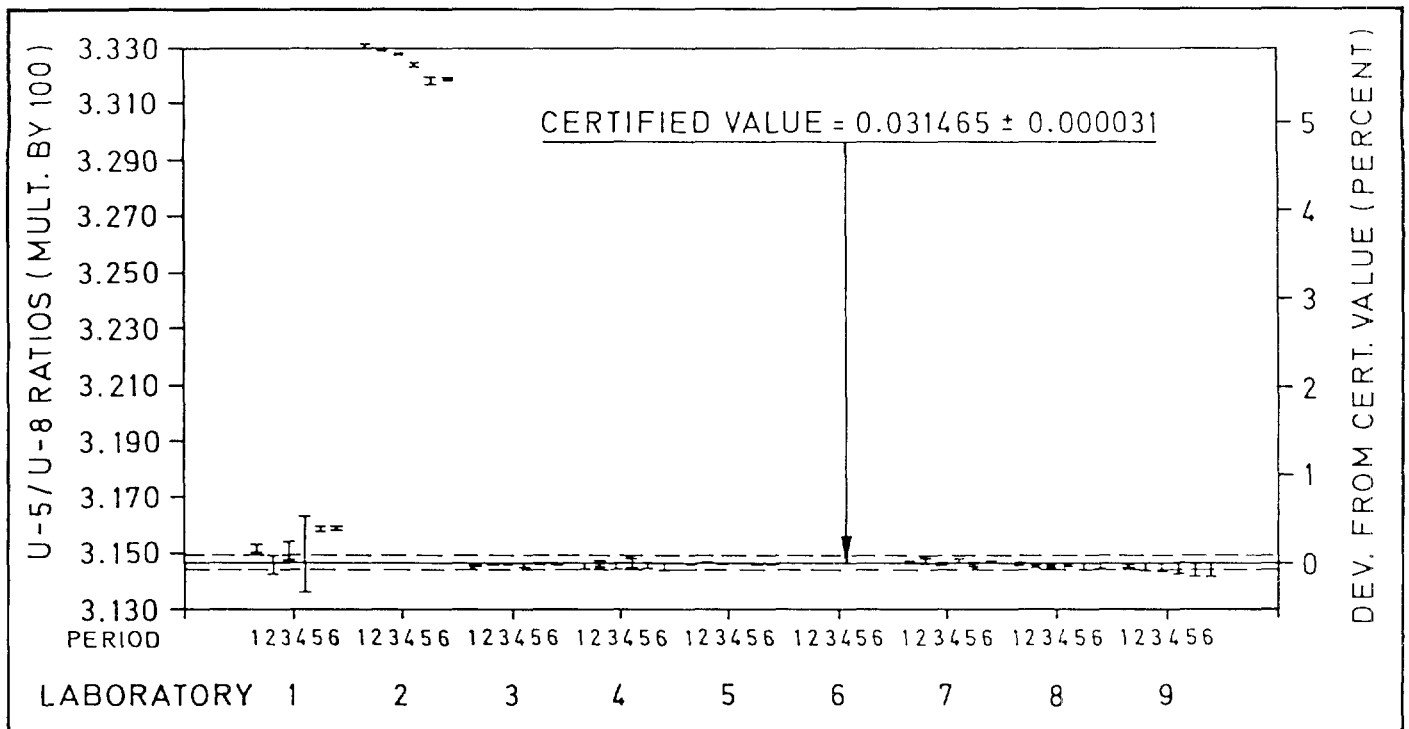Uranium Concentration. Solution B

Fig. 4 - Reported ^{235}U / ^{238}U ratios for each participating laboratory (in UF_6)

5. Perhaps one of the main activities was concentrated on the achievable analytical precisions and accuracies in verification methods for accountancy of nuclear materials. Again the aim was to establish the present "state-of-the-practice" as opposed to the "state-of-the-art".

After about four years of preparation, the Group established a list of achievable precisions and accuracies for different assay methods used on the commonly occurring nuclear materials. The values of these precisions and accuracies are values for which analytical laboratories should aim at (they are therefore called "Target Values") because they can readily be achieved. The list can be found elsewhere in this Bulletin (3) and will be presented at the May 1984 ESARDA Symposium in Venezia and the INMM Symposium in Columbus (U.S.A.).

The IAEA is already using the "Target Values" and has presented experience from this use to the Group.

The Group now has representatives of 37 nuclear analytical laboratories including observers from Euratom, IAEA, USA. Norway and Austria.

Literature

(1) W. Beyrich, W. Golly, G. Spannagel (KfK),
P. De Bièvre, W.H. Wolters, M. Gallet (CBNM),
L.A. Machlan, J.W. Gramlich, J.D. Fassett (NBS),
The IDA-80 Measurement Evaluation Programme on Mass Spectrometric Isotope Dilution Analysis of Uranium and Plutonium : some preliminary results
IAEA / SM-260 pp. 261 ff

(2) P. De Bièvre, W.H. Wolters, G. Müschenborn, W. De Bolle (CBNM), W. Beyrich, W. Golly (KfK)
The 1982 State-of-the-Practice in Low Enriched UF_6Isotopic Measurements in Western Europe
Proceedings 5th ESARDA Symposium 1983 pp. 177 ff

(3) P. De Bièvre (CBNM), S. Baumann (ALKEM), T. Görgenyi (NUKEM), E. Kuhn (IAEA), S. Deron (IAEA), P. De Regge (SCK),
1983 Target Values for Uncertainty Components in Fissile Element and Isotope Assay
ESARDA Bulletin, Proceedings 6th ESARDA Symposium 1984, ESARDA Bulletin, here

PERLA : Performance Test Laboratory for Safeguards

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For many years the major R&D effort in the area of non-destructive assay (NDA) and containment and surveillance (C/S) has been devoted to the development of instrumentation to be used for safeguards and for plant management. On many occasions, e.g. in ESARDA symposia or in IAEA advisory group meetings, opinions have been expressed calling for a more systematic and structured approach in the transformation of a laboratory prototype into a commercial product ready to be used in the industrial environment. This process requires various steps and in particular that of evaluating in near field conditions the performances of the instruments.

In this view and as part of its 1984-1987 multiannual programme, the JRC-Ispra is proposing to set up a performance evaluation laboratory (PERLA) for NDA and C/S. This laboratory should profit from the liberation of important resources in terms of facilities, staff and budget due to the interruption of the SuperSARA project. More precisely, the new staff will need reconversion and special training and a number of existing laboratories at the ESSOR complex will have to be modified and adapted to their new functions.

It is well known that the JRC, which has been developing instruments and methods for safeguards since 1969, has sometimes encountered difficulty in testing them in field conditions because of the lack of necessary facilities. As a consequence, the JRC has found it necessary to establish collaboration with the nuclear industry and some R&D laboratories of the member states to perform field tests. Therefore, even though the relationships with industry and laboratories have always been excellent, it was felt necessary to have available a laboratory on the spot where the near-field conditions can allow the testing of instrument performance, thus avoiding the boundary conditions and constraints imposed by the facilities.

It is the intention of the JRC to continue and possibly to increase this type of collaboration but with the set up of PERLA, the JRC will be able to concentrate on one site the possibilities to:

- establish the performances of instruments and methods on a large variety of nuclear materials,
- recalibrate the instruments,
- train inspectors and operators
- perform new developments.

It is envisaged to cover most of the techniques used in NDA namely : gamma spectrometry, passive and active neutron techniques, and calorimetry. Concerning C/S, various types of seals and optical surveillance devices will be taken into consideration. One must also not forget that NDA can be applied to spent fuels and with this in mind other classes of instruments will be tested such as Cerenkov viewers and special gamma scanning devices.

As PERLA is intended to operate in near-field conditions, it has to be provided with a large variety of nuclear materials. These have been defined through a careful analysis of the various fuel cycles namely for LEU, HEU and Pu. The nuclear materials which will be procured, will be as representative, this means as close as possible in forms, composition and characteristics, as possible of the numerous products of the cycles considered.

To conclude, we think that as far as the objectives are concerned, a good and sound basis has been established on our own experience and wishes or needs expressed by potential users. The practical implementation has started at different levels such as the transfer of know-how for the new staff, contacts with operators for realizing basic studies and developing proposals for the construction of the laboratories and contacts with nuclear materials dealers and operators for the procurement of various samples.

A paper on the subject will be presented at the 6th ESARDA Symposium, Venice, 14-18 May 1984 (paper 5.2).

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