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Preview for Versailles Symposium Fifth ESARDA

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The Fifth ESARDA Symposium on Safeguards and Nuclear Material Management will be held in Versailles, France on 19-21 April 1983. The "Call for Papers" is reproduced at the end of this issue.

The place will be the "Palais des Congrès" at Versailles which is situated at the west boundary of Paris. The oral sessions will be held at the first floor in the "Salle Richelieu" while the posters and the exhibition will be at ground floor in the "Salle Mazarin".

An exhibition of equipment related to fissile material control will be organized during the symposium. The centre of the circular hall will be used by the exhibitors while the posters will be accommodated around it. The area of each poster will be 2.5 m x 1.22 m with the larger side horizontal. The maximum height will be 2 m.

Coffee breaks will be organized in the same room as for posters and exhibition.

Normal slides (5 cm x 5 cm) and/or overhead projector can be used for oral presentation. A single operator will operate all the controls including room lights. The chairmen will have a preset time bell for limiting the oral presentations and an interphone with the operator to cut the sound amplification if necessary.

A lunch buffet will be organized the 19 April in the Congress Hall. A dinner will be organized in Paris on the evening of the 20 April.

Original papers on the subjects mentioned in the call for papers are expected to reach the secretariat not later than the 30 November 1982.

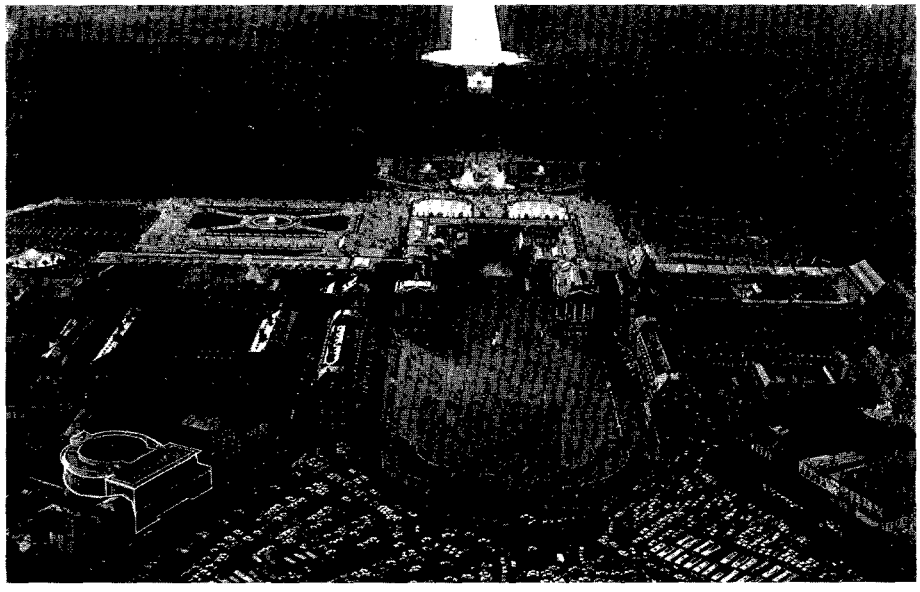
This deadline must be strictly observed to allow the selection committee to evaluate the papers and group the accepted ones in the appropriate sections. The symposium programme will be prepared during January 1983 and dispatched at the beginning of February.

The programme will contain all the information for the participants and include the registration card as well as the hotel reservation card. The organization will book rooms of different price levels according to the request of the participants in hotels situated in Paris on the left side (Rive Gauche) of the river Seine. A very frequent train connection is available from the stations of the "Rive Gauche" to Versailles.

Finally, a social programme will be organized for accompanying guests.

In addition a technical visit to EURODIF could be arranged on Friday 22 April if enough participants are interested. EURODIF is located near Valence about 100 km from Lyon in the South of France.

Panoramic view of Versailles



Characterization of the Results of the Post-Irradiation Experiment "Benchmark"

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Introduction

The Joint Research Centre (JRC) of the Commission of the European Communities (CEC) has carried out destructive post-irradiation analyses of samples from BWR and PWR fuel elements for many years. A total of about 120 pellets (see Table I) with a burnup range from 3.5 to 37.5 GWD/MTU were investigated in this activity called Benchmark (BM) experiment. The main goal was to create a bank of qualified data on burnup and isotope buildup of spent fuels, to be used in assessing some Safeguards techniques as well as reactor burnup codes and their cross section library.

All information on the reactors, fuel elements and investigated samples has been given in a recent summary report[1]. Details of the analytical and measurement procedures have also been given, together with all raw and elaborated data concerning burnup and heavy isotope (U, Pu, Am, Cm) buildups. Extensive references are given in the report to the papers periodically published by JRC on the Benchmark activity.

The experiments on Garigliano and Trino Vercellese fuel were performed within the framework of research contracts established between Euratom, the Ente Nazionale per l'Energia Elettrica (ENEL) and FIAT.

Those on Obrigheim and Gundremmingen were carried out within the framework of two cooperation agreements between Euratom, Kernkraftwerk RWE-Bayernwerk GmbH (KRB), Kernkraftwerk Obrigheim GmbH (KWO) and Kraftwerk Union Ag (KWU).

Analysis and characterization of the results

The following methods were used to qualify the BM data:

a) careful analysis of random and systematic uncertainty,
b) comparison with calculated data,
c) interpretation of data through the Isotopic Correlation Technique (ICT).

Table I Reactors and samples investigated in the framework of the Benchmark activity

Reactor	Identification	Fuel Assembly	^{235}U wt% Initial Enrichment	Number of Samples	GWD/MTU Sample Burnup Range
Garigliano (BWR)	Garigliano I	A-106	1.60	5	9.8-14.5
			2.10	13	8.9-12.7
	Garigliano II	SA-13	2.41	8	4.2- 8.6
Trino Vercellese (PWR)	Trino I	509-049	2.719	14	8.2-15.3
		509-032	3.13	8	7.2-17.7
		509-104	3.897	4	3.4-11.9
	Trino II	509-069	3.13	23	19.1-26.6
Obrigheim (PWR)		BE 124	3.00	19	15.6-36.3
		BE 210	2.83	8	24.2-37.5
Gundremmingen (BWR)		B 23	2.53	9	21.2-27.4
		C 16	2.53	7	14.4-20.3
TOTAL NUMBER OF SAMPLES				118	

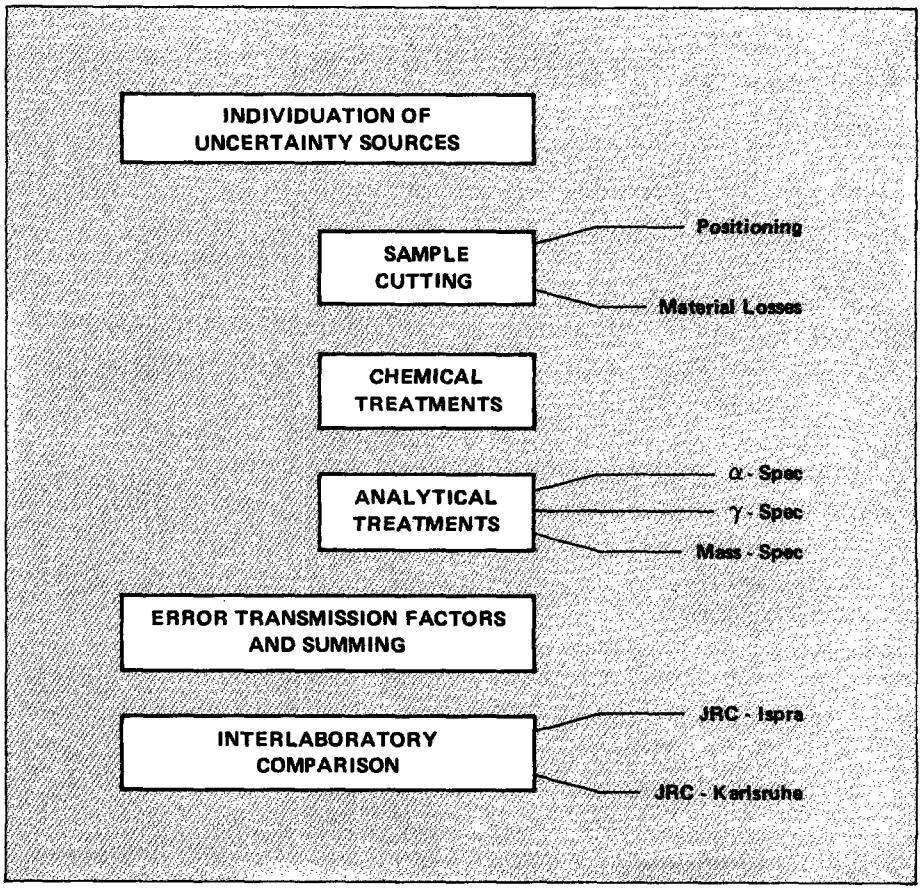

Figure 1 Model for Benchmark results uncertainty assessment

Experimental uncertainty analysis

The analysis of the uncertainty relevant to the determination of the BM results (burnup and heavy isotope buildups) required first of all the setting up of a model for the evaluation of possible uncertainty sources involved in the preliminary operations of the measuring procedure, such as sample cutting and chemical treatments (Fig. 1).

The next step concerns the evaluation of the raw data uncertainties and their transmission factors on the final burnup and buildup values.

To evaluate systematic uncertainties, pairs of samples of each batch were analysed in parallel in the two JRC-laboratories of Ispra and Karlsruhe.

The contribution to the total uncertainty originating in the sample cutting position was evaluated on the basis of the experimental axial burnup and buildup distribution and knowing the cutting position uncertainty (± 1 mm).

The eventual material loss in pellet cutting could significantly alter the Pu distribution in the pellet. Indeed, resonance capture in U-238 determines a complex spatial structure in the Pu production in the pin. The effect due to the eventual material loss is difficult to detect and it has been estimated with the aid of the multigroup transport cell calculation code LASER[2] simulating different situations of material loss.

The uncertainty in the α, γ and mass spectrometry measurements was assessed starting from the measured uncertainties in the raw data (Table II) and propagating them to the final results. The final uncertainties evaluated in this way are shown in Table III.

Then a simple model of burnup dependence of the uncertainty was developed assuming a linear dependence of raw data from burnup. The conclusion was, that in the limits of the model only U-235 and U-238 depletions or Pu-242 and Cm-244 buildups have a marked burnup dependence of their uncertainty. The errors on burnup and on other isotopes are not significantly dependent on burnup.

A table of the error sensitivity was then prepared showing which parameters have the greatest influence on the final uncertainties (see Table IV).

Finally, in order to draw some inferences on the laboratory bias, a comparison was carried out between the results obtained on the same samples by the two JRC-laboratories of Ispra and Karlsruhe. The results of the comparison were generally good, demonstrating that no major bias was present in the measurements. Only the results of Pu-235 and Cm-244 showed a greater discrepancy than expected. A discussion on the possible causes can be found in ref. 1.

A summary of the main results, isotope by isotope, is given there and is partially reported here in the following paragraphs.

Table II Evaluated percentage uncertainties for measured raw parameters

Parameter	Burnup (MWD/MTU)			
	7694	10932	19039	24637
$^{235}U/^{238}U$	056	0.59	0.66	0.71
$^{236}U/^{238}U$	1.64	1.57	1.38	1.24
$^{240}Pu/^{239}Pu$	0.41	0.42	0.45	0.48
$^{241}Pu/^{239}Pu$	0.65	0.64	0.60	0.58
$^{242}Pu/^{239}Pu$	1.70	1.52	1.01	0.67
$^{242}Cm/(^{239}Pu + ^{240}Pu)$	4.00	4.00	4.00	4.00
$^{244}Cm/(^{239}Pu + ^{240}Pu)$	8.51	7.43	4.47	2.42
$(^{238}Pu + ^{241}Am)/(^{239}Pu + ^{240}Pu)$	0.81	1.02	1.58	1.97
$^{238}Pu/(^{239}Pu + ^{240}Pu)$	1.00	1.00	1.00	1.00
Pu concentration	0.65	0.65	0.65	0.65
U concentration	0.58	0.58	0.58	0.58
Nd concentration	0.60	0.60	0.60	0.60
$^{148}Nd/^{238}U$	1.00	1.00	1.00	1.00
initial ^{235}U wt%	0.60	0.60	0.60	0.60
fission yield (^{148}Nd)	1.00	1.00	1.00	1.00

Table III Percentage analytical uncertainty of derived parameters at different burnup levels

DERIVED PARAMETERS	Burnup (MWD/MTU)			
	7694	10932	19039	24637
^{235}U Depletion	2.46	1.80	1.23	1.03
^{236}U Buildup	1.64	1.57	1.38	1.24
^{238}U Depletion	4.66	3.43	2.29	2.05
^{238}Pu Buildup	1.75	1.83	2.01	1.99
^{239}Pu Buildup	0.87	0.88	0.88	0.88
^{240}Pu Buildup	0.96	0.97	0.98	0.99
^{241}Pu Buildup	1.20	1.19	1.14	1.15
^{242}Pu Buildup	1.91	1.75	1.33	1.10
^{241}Am Buildup	18.7	–	–	20.00
^{242}Cm Buildup	4.17	4.18	4.19	4.21
^{244}Cm Buildup	8.59	7.53	4.64	2.75
Pu/U Mass ratio	0.73	0.69	0.64	0.61
^{148}Nd Burnup	1.40	1.41	1.42	1.43
^{137}Cs Burnup	–	–	–	1.8

Burnup from Nd-148

Two equal components of errors are summed up from Nd-148 determination and from Nd-148 fission yield. The burnup dependence of the error on burnup is weak. The interlaboratory comparison was good.

Depletion of U-235

The initial and the final U-235 percent content have not the same transmission factor. The initial content must be known with greater accuracy than the final one. The burnup dependence of the error is marked. The interlaboratory comparison was good.

Buildup of U-236

Only the error on U-236 determination is of any importance. The burnup dependence is appreciable. The interlaboratory comparison was good.

Buildup of Pu-238

The most important parameters with respect to buildup error are:

- the Pu-238 ratio measurement
- the uncertainty on the knowledge of the decay constant of Pu-238
- the Cm-242/(Pu-239 + Pu-240) ratio measurement,
- the U and Pu concentration determination.

The burnup dependence is weak.
The interlaboratory comparison gave questionable results for Obrigheim data, probably due to a difficult spectrum analysis mainly at high burnups.

Buildup of Pu-239

The Pu and U concentration errors only play a nearly balanced role in the final error. The burnup dependence is weak. The interlaboratory comparison was good.

Buildups of Pu-240, Pu-241, Pu-242

For all these isotopes the uncertainty in buildup is similar. The significant error components are three of approximately the same weight: the U and Pu concentration determination and the mass spectrometric determination of the isotope itself.
The burnup dependence is significant only for Pu-242. The interlaboratory comparison was good for all of them.

Buildup of Am-241

Only one error component is significant, i.e. the uncertainty of the measurement of (Pu-238 + Am-241) / (PU-239 + Pu-240). Not enough paired data were available to give a verdict on the Ispra-Karlsruhe intercomparison.

Buildup of Cm-244

The only significant error component is that on Cm-244/(Pu-239 + Pu-240) determination.
The burnup dependence was appreciable. The interlaboratory comparison showed that the evaluated error of Table III was probably underestimated.

Comparison with calculated data

The experimental data were compared with calculated values. Two codes, LASER[2] and RIBOT[3], were used for recalculation of the experimental data.

LASER is a multi-group, one-dimension transport burnup code, whose isotopic chain in the version available at Ispra is limited to Pu-242. The code was used mainly to check Pu isotope production and isotopic correlations, as shown in Fig. 2.

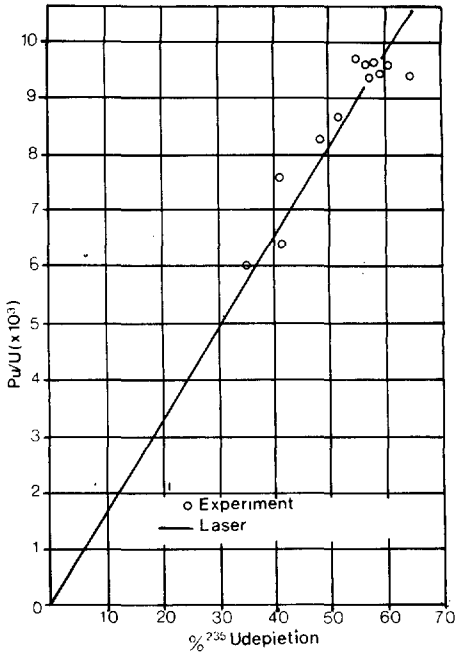

Figure 2 Trino Vercellese : Pu/U as a function of ^{235}U depletion

The results of the comparisons were always quite satisfactory. The exercise was performed on results from two PWRs, YANKEE ROWE and TRINO Vercellese.

$$(*)\ Z = \frac{100}{\sum_{i=1}^{n} r_i + \frac{^{148}Nd}{^{238}U} \cdot \frac{1}{y}}$$

N = negligible

Table IV Error sensitivity analysis of ^{148}Nd burnup, ^{235}U and ^{238}U depletion and heavy isotopes buildup at 24637 MWD/MTU

PERCENTAGE EXPERIMENTAL ERROR	0.60	0.71	1.24	0.01	0.12	0.48	0.58	0.67	4.00	2.42	1.97	1.00	0.58	0.65	0.97	1.00	0.04	0.90	0.40	1.10	2.70	1.30	0.90	0.06	0.06	
ROW DATA / DERIVED DATA	INITIAL ENRICH.T ^{235}U wt%	^{235}U at%	^{236}U at%	^{238}U at%	^{239}Pu at%	^{240}Pu at%	^{241}Pu at%	^{242}Pu at%	$\frac{^{242}Cm}{^{239}Pu+^{240}Pu}$	$\frac{^{244}Cm}{^{239}Pu+^{240}Pu}$	$\frac{^{238}Pu+^{241}Am}{^{239}Pu+^{240}Pu}$	$\frac{^{238}Pu}{^{239}Pu+^{240}Pu}$	U CONCENTRATION	Pu CONCENTRATION	$\frac{^{148}Nd}{^{238}U}$	^{148}Nd FISSION YIELD	(*) Z	$\lambda(^{238}Pu)$	$\lambda(^{239}Pu)$	$\lambda(^{240}Pu)$	$\lambda(^{241}Pu)$	$\lambda(^{242}Pu)$	$\lambda(^{241}Am)$	$\lambda(^{242}Cm)$	$\lambda(^{244}Cm)$	% TOTAL ERROR
BURNUP (Ft %) ^{148}Nd															1.00	1.03										1.43
^{235}U DEPLETION	0.94	0.40		N													N									1.03
^{236}U BUILDUP			1.24	N																						1.24
^{238}U DEPLETION		1.09															1.74									2.05
^{238}Pu BUILDUP				N	0.06	0.25			0.88			1.22	0.58	0.65			N	0.70	0.19	0.58				0.09		2.00
^{239}Pu BUILDUP				N	0.11								0.58	0.65												0.88
^{240}Pu BUILDUP				N		0.48							0.58	0.65			N									1.00
^{241}Pu BUILDUP				N			0.58						0.58	0.65			N				0.49					1.15
^{242}Pu BUILDUP				N				0.67					0.58	0.65			N					N				1.10
^{241}Am BUILDUP				N	0.54	1.28					18.76	5.87	0.58	0.65			N		1.72	1.64			2.43			20.00
^{242}Cm BUILDUP				N	0.18	0.25			4.00				0.58	0.65			N		0.61	0.58				0.41		4.21
^{244}Cm BUILDUP				N	0.18	0.25				2.42			0.58	0.65			N		0.61	0.58					0.41	2.75

RIBOT is a multi-group, zero dimension, diffusion burnup code developed by Comitato Nazionale per l'Energia Nucleare (CNEN). The code, in operation for many years, was just implemented with the Am and Cm chains. Its use for Benchmark data analysis was mainly intended for checking the RIBOT library for Am and Cm isotopes.

The results obtained in the comparison with data from TRINO and Obrigheim PWRs were quite satisfactory for almost all isotopes. All Pu isotopes were in excellent agreement. The worst case (disagreement of a few percent) was Pu-242, presented in Fig. 3. All calculated transplutonium isotope values were acceptable. One of the worst cases was Cm-244 (Fig. 4). Only Pu-238 was underestimated by RIBOT by roughly a factor 2.

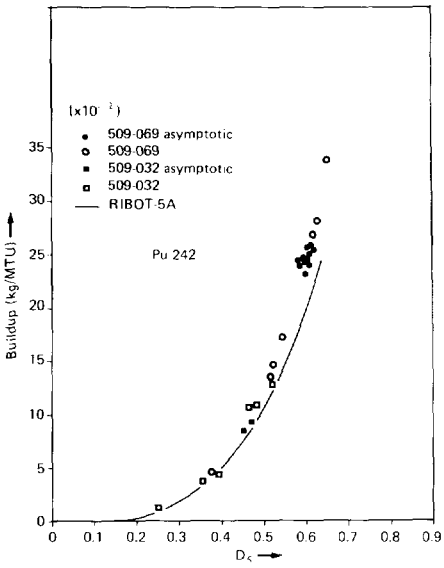

Figure 3 Comparison between calculated and experimental results for Trino Vercellese

Isotopic correlation technique (ICT)

One of the most powerful methods for the consistency check of experimental destructive post-irradiation data is the ICT. Intense use of this technique was therefore made throughout all the measurement campaigns and is summarized amply in the final report[1].

The destructive measurement of fission products and correlation amongst them was currently used in the BM activity as can be seen in Fig. 5, where the Cs-134/Cs-137 activity ratios are shown, correlated with the Nd-148 evaluated burnup for samples of PWRs.

There is a certain evidence of non-linearity in the whole burnup range and the explanation can be in the fact that the Cs-133 capture cross section has an important resonance capture part, making it dependent on the spectral conditions. A much better correlation is obtained with Cs-137, as shown in Fig. 6, where all destructive historical data produced in the JRC are plotted against burnup.

A certain number of correlations involving heavy isotopes were also extensively employed in the BM experiments, for internal data checking, reactor physics considerations and safeguards tool assessment. The technique now has worldwide application for the checking of relevant Safeguards parameters, such as burnups, buildups of Pu isotopes, Pu/U mass ratios (Fig. 7), decay times, irradiation histories, etc.

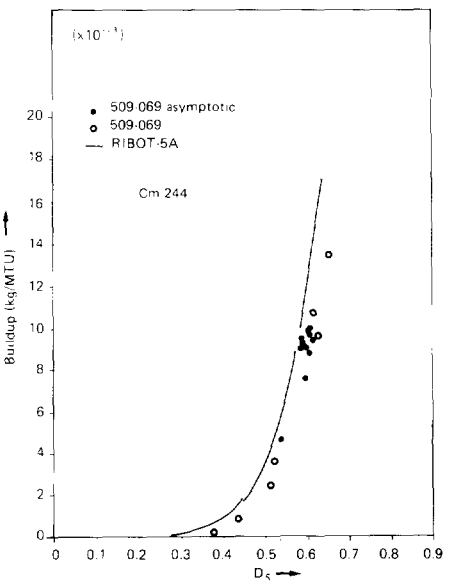

Figure 4 Comparison between calculated and experimental results for Trino Vercellese

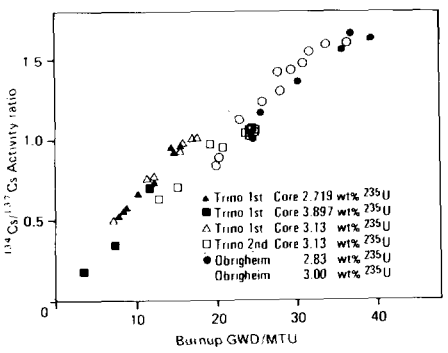

Figure 5 Correlation between the activity ratio $^{134}Cs/^{137}Cs$ and burnup for the PWR samples analysed at the JRC

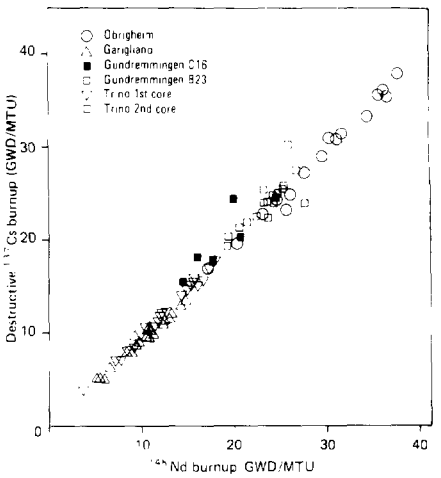

Figure 6 Correlation between ^{137}Cs and ^{148}Nd burnups for all JRC samples

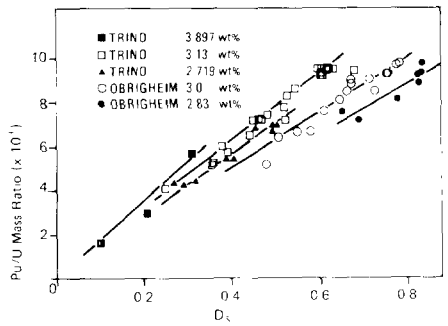

Figure 7 Correlation between Pu/U mass ratio and ^{235}U depletion for all PWR samples analysed at JRC

Conclusions

The BM experiment was a destructive post-irradiation analysis activity carried on for many years by the JRC of CEC.

A wide range of burnup and light water reactor types, the qualification and traceability of the data, the availability of pertinent reactor and irradiation data for pellet recalculation, make the results of the BM a useful tool for the assessment of Safeguards techniques.

The most relevant achievements of the activity, the characterization of data and the pertinent information needed for their recalculation are all collected in the above mentioned BM final report.

References

1 S. GUARDINI, G. GUZZI: «Benchmark : Reference destructive post-irradiation data on LWR fuels. Summary of 12 years of experience at JRC. Results and their characterization», EUR 7879.EN (1982)

2 C.G. PONCELET: «LASER: A depletion programme for lattice calculations based on MUFT and THERMOS», WCAP 6073 (1966)

3 P. LOIZZO: «RIBOT 5: A physical model for light water lattice calculations», BNWL 735 (1968)

KfK Film-Camera System for Safeguards

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Introduction

For safeguards in nuclear facilities, particularly in reactor plants, the instrumental surveillance by cameras is of an essential importance as a complementary measure to accountancy. Euratom and IAEA have been using for years film cameras for this purpose which have been adapted to this function by corresponding modifications. The development of the film-camera system presented here is oriented towards the same goal. It has been tried to improve its application range above all by three measures, i.e. by increasing the range of luminous intensity in which the camera operates, by increasing the film capacity and, last not least, by recording date and time on each frame.

Concept of the Camera System

Together with the camera system itself, the requirements for the system were developed in long range discussions with Euratom and IAEA. The manufacture of the camera system should be economically as favourable as possible. Therefore, the concept is based on the application of a super-8 movie camera; by accessories and modifications the system is constructed with the necessary properties. At last only the body, optics and mechanical components of the basic camera are used.

The camera system is characterized by the following properties:

- single-frame exposure controlled by an external timer with different selectable time intervals between 1 to 29 min, in steps of 1 min;
- auto-diaphragm and exposure time control from aperture value 22 to 1.2 at 1/30 sec and from 1/30 sec to 1 min at aperture value 1.2;
- possibility to accommodate 61 m film cartridges with 14,500 frames;
- recording of date and time on each frame, with a display controlled by the external timer for presenting this information and with an additional optical system for transmission to the film window;

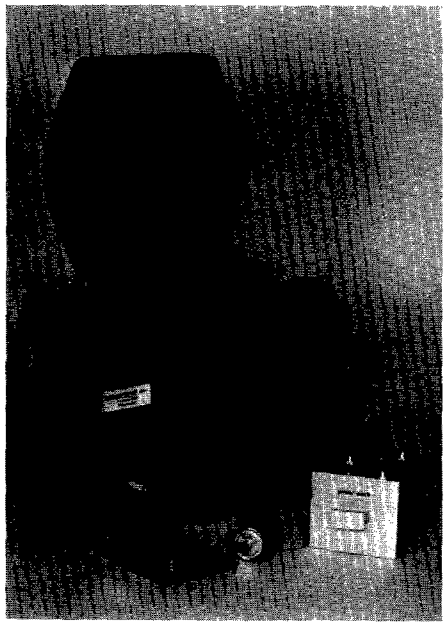

Figure 1 KfK film camera system with electronic box and battery case, 61 m film-cartridge

- power supply for the camera by batteries, modified power-economizing camera electronics so that 20,000 exposures can be made without battery change; independent battery for the timer for an operation time of at least two years;
- test system for testing the function of the entire system, of the battery charge and the film reserve.

Furthermore, the following accessories are to be provided for:

- a casing for a hermetic and safe placing of the cameras, with one camera in one casing;
- a reusable 61 m film cartridge which can be loaded with film by the safeguards organization itself.

Design

The camera system was constructed at first on the basis of the EUMIG-881 PMA camera. The latter was supplied a priori with an exposure time control mechanism (long-time exposure control) so that it was possible to install the combined diaphragm and exposure time control mechanism without interfering with the mechanics.

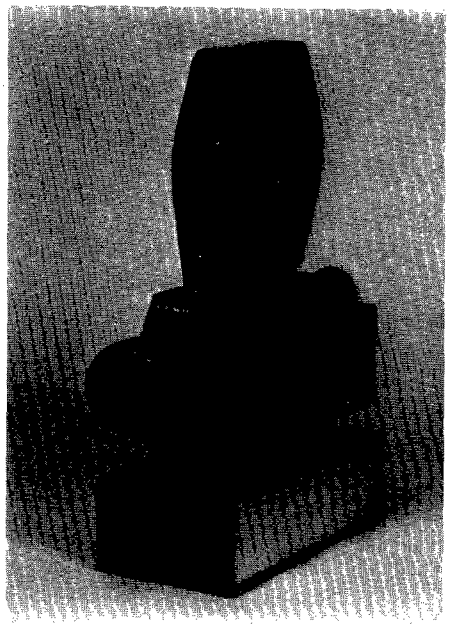

Figure 1 KfK filmcamera system, other side, with casing for date-time display and additional optics

After completion and successful testing of the prototypes the failure of the EUMIG company made it necessary to change the camera. Last not least as a result of very positive tests carried out in the Sandia National Laboratories, U.S.A., with this camera type, the ELMO 612 S-XL super-8 movie camera was selected.

The system named KfK Mark IIb-film camera consists, as can be seen from outside, of three parts: camera body with picture optics, electronic box with battery case below the camera and the casing on the side of the body for display and additional optics for recording data and time (see Figs. 1 and 2).

Besides the body, from the original camera the picture optics, view finder, the photometric optics as well as the diaphragm and shutter mechanism have been taken, the latter modified for long-time exposure.

The lens is a zoom for manual operation, F 1.2, 8.5 - 51 mm.

A rotary disk serves as a shutter. The camera is designed for single frame exposure. The normal exposure time is 1/30 sec, whereby the rotary disk shutter performs each time a full circle. By an accessory unit in the rotation mechanics, in the case of long-time exposure, the

rotary shutter is opened, stopped and released for finishing the pass only after the end of the exposure time (which can be between 1/15 and 30 sec). The electric motor for the shutter operation has been maintained.

The light is measured by the photodiode originally installed at the film window, also for the time control, in the case of long-time exposure. The view finder with split-field telemeter has been maintained in the original state, too. The camera is originally designed for 61 m cartridges; a cover plate over the cartridge case guarantees a light-proof closing, when normal film cartridges are used. The film counter registers up to the film length in the large-capacity cartridge.

In the electronic box below the camera, directly connected with it, the timer and clock, the control electronics for the camera and the batteries are installed, with operating element at the external front.

The timer with oscillator quartz generates the trigger pulses for the camera and controls as a clock the date-time indication to be recorded onto the frames. For the exposure triggering constant time intervals from 1 min to 29 min, in steps of 1 min, may be preselected. For the date-time recording the clock feeds an LED display at the camera; the reading may be fetched on a second display at the front plate and the clock may be adjusted. Power supply for timer and clock comes from two mercury batteries, separately from the power supply of the camera electronics. Their capacity is sufficient for several years at a power consumption of a few μA. Thus, it is guaranteed that the timer and clock go on unchanged, when the camera batteries are replaced.

The control electronics of the camera is equally designed corresponding to the demand for a possibly low power consumption. In the intervals between the exposures it is switched off. The pulse from the timer triggers in a preliminary interval at first the diaphragm setting according to the light conditions and the change from normal-time exposure to long-time exposure, respectively. A second sequence pulse, issued by the timer as well, then triggers the exposure itself, in the case of normal-time exposure with full course of the exposure, in the case of long-time exposure for opening the disk shutter, the shutting of which is then induced by a third pulse within a time depending on the measured luminous intensity.

For the power supply of the camera control electronics and of the electro-mechanical elements in the camera, especially of the diaphragm motor and the driving motor for the shutter as well as of the LED displays, six 1.5 V alkaline batteries with a capacity of about 4 A are provided. This capacity is sufficient with safety reserve for the exposure of one 61 m film. The batteries are installed in a case in the electronic box; this case can be replaced as a whole.

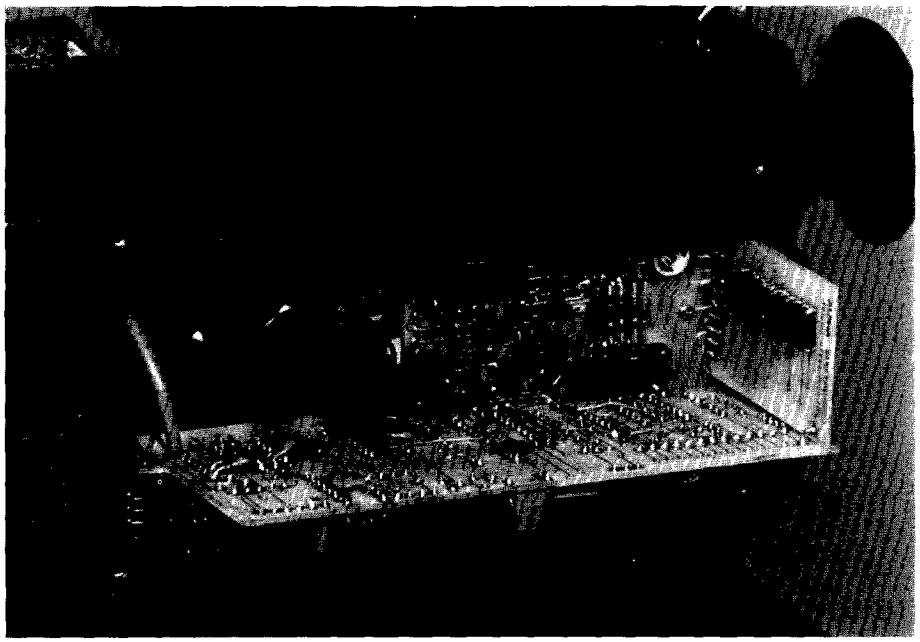

Figure 3 Display (right side) and parts of the optic system for date and time recording (middle)

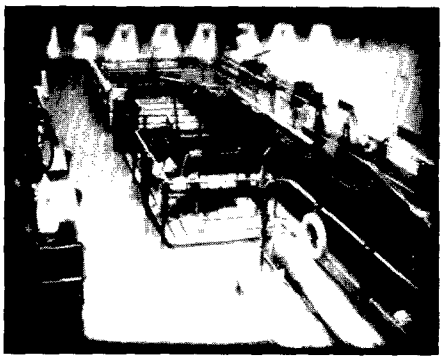

Figure 4 : Frame with date and time

A small testing system makes it possible to make sure by pressing a button on the front plate of the electronic box, whether a film is inserted and not fully passed through and furthermore, whether fresh batteries have been introduced for enabling the exposure of a 61 m film. Moreover, the regular function of the entire system can be checked integrally from the pulse triggering of the electronic timer to the exposure.

The display and the optical system for the date-time recording are installed on the camera below the lateral covering cap. The display consists in an eight-figure LED indicator. Over two mirrors and a supplementary lens the information is transmitted directly onto the film. The LEDs are approched during 30 msec for every exposure. Thus, the recording of date and time on each frame is not influenced either by the camera aperture or by the exposure time.

Closing Remark

As far as surveillance by cameras is concerned, for Euratom as well as for the IAEA the emphasis is still clearly on using film-camera systems for the time being. With the film-camera system proposed here tests have been carried out to find out what possibilities film cameras have, above all with respect to three properties:

- comparatively cheap manufacture
- comparatively small space requirement
- possibility of autarchic operation.

By recording date and time on the frames, using large-capacity cartridges and the automatic diaphragm and exposure time mechanism the applicability is considerably extended; further accessories, such as e.g. the exposure triggering by stochastic trigger or motion detectors, would be possible. But it is to be expected that the end of this development will imply the completion of the development of surveillance film cameras in general. In future video-systems and their adaptors for surveillance tasks will characterize the main activities in research and development work for instrumental optical surveillance.

In-Field Use of a Quadrupole Mass Spectrometer for Isotope Analysis of UF_6

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JRC-Ispra

Introduction

The exact determination, by plant operators and safeguards inspectors of the content of fissile materials in the various parts of the fuel cycle is generally performed by weighing and sampling the bulk material to perform destructive assay for chemical and isotopic composition determinations.

The ability to perform frequent determination of the isotopic composition of such material is thus essential.

For verification purposes, safeguards inspectors usually draw samples of the material to be controlled, and ship them outside to analytical laboratories designated by them where, much later, the samples are analysed. This approach is encountering two major problems. One is the increasing and serious administrative difficulties of transporting samples from one facility to the analytical laboratory. The second problem is linked to the long time delay before receiving the measurement results from the analytical laboratory.

To overcome or even decrease the extent of these difficulties, the JRC-Ispra has developed a transportable instrument for isotope analysis with the aim of performing destructive analyses at a facility.

Although it is a rather complex technique, mass spectrometry was considered as an appropriate tool for in-field measurements. Because of its compactness and its leightweight, a quadrupole mass spectrometer (QMS) was chosen.

Instrument

The instrument developed in our laboratory is composed of:

- a quadrupole mass filter coupled with a cross-beam ion source with tungsten cathode filaments;
- a Faraday collector connected to an electrometer amplifier;
- a radio-frequency unit;
- a set of electronic function modules Balzers QMG 511 interfaced to a Digital MINC 11 computer;
- an inlet system provided with four sections for batch sample introduction, especially designed to allow analyses of low and high enriched UF_6 samples through two independent lines.

It must be pointed out that this equipment is a prototype partly constructed in the JRC-Ispra workshop and partly constructed using commercial material assembled in our laboratory together with Balzers and Digital instrumentation. In Fig. 1 an overall view of the instrument is presented.

The main specifications of our prototype instrument can be summarized as follows:

- abundance sensitivity at "unit" resolution: < 10 ppm related to the intensity of the peak 333 ($^{238}UF^{+}_{5}$);
- detection limit at "unit" resolution with Faraday detector: 3 ppm of base peak intensity $^{238}UF^{+}_{5}$;
- memory factor: 1.002 when changing over from a 2.5% to a 0.7% sample even after an extended throughput of the higher material through the source;
- sample consumption of the ion source: 1.5 mg/h with a UF_6 pressure in the expansion tank of 0.3 Torr;
- operation time without cleaning the ion source: 1000 h;
- precision (relative internal standard deviation) of double ratio measurements: 1×10^{-3};
- accuracy (relative external standard deviation) with sample to standard calibration obtained by comparing measured $^{235}U/^{238}U$ ratios with quoted $^{235}U/^{238}U$ ratios of samples having enrichments from 0.3% to 5.8% : < 0.2% (average value).

The measurement procedure is the following: the unknown sample and a reference standard varying in enrichment by no more than 10% are introduced into the source alternatively. The $^{235}U/^{238}U$ isotope ratios of both the sample and the standard are determined by the use of the MINC 11 computer following the measurement procedures. One complete measuring cycle (9 single isotope measurements for both the sample and the standard) requires about 20 minutes including manual operations for changing the samples and flushing the source to minimize memory effects. The $^{235}U/^{238}U$ ratios of the sample are recorded by the program as R_1, the $^{235}U/^{238}U$ ratios of the

Figure 1 View of the transportable quadrupole mass spectrometer

standard are recorded as R_2. Each analysis requires some double ratio (R_1/R_2) measurements the number of which is chosen by the operator.

In order to have internal standard deviations of better than 10^{-3}, 10 to 15 double ratio measurements are usually carried out. Besides the determination of the $^{235}U/^{238}U$ ratio, and because precise measurements of minor isotopes ^{234}U and ^{236}U are increasingly important, the computer program has also been supplied with a routine capable of determining the $^{234}U/^{235}U$ and the $^{236}U/^{235}U$ ratios.

Results

Three in-field experiments were carried out in February 1979, late in 1981 and in February 1982, with the aim of testing the prototype instrument after long journeys and repeated loading and unloading operations with the instrument partially disassembled and the most delicate parts packed into wooden cases.

To facilitate the movements the entire instrument is mounted on racks with retractable wheels.

The first experiment was conducted at Balzers A.G. Works. The main aim was the test of the inlet system developed at JRC-Ispra. The test was performed with the instrument installed in a van and connected to a PDP 11 computer of the Balzers' Laboratories.

In general, fifteen measuring cycles per sample were carried out in order to have sufficient reproducibility of the measurements. The relative precision ranged in fact from about 0.2% to 0.02% according to the enrichment of the samples.

Precision and accuracy of this experiment were mainly affected by the inefficient temperature control in the van and by the incomplete elimination of vibrations caused by the rotary pumps system.

For practical reasons it was preferred to carry out future measurements by installing the equipment in the plant's laboratories. During the second in-field experiment at the URENCO plant, three plant UF_6 samples from depleted to enriched at 3.3 % ^{235}U were analysed using 10 measuring cycles. The relative precision of the measurements was satisfactory (less than 0.1%) and the relative accuracy was not worse than 0.5% for the depleted and natural samples. However, the sample enriched at 3.4% gave a relative accuracy of 0.07%.

In order to reduce the duration of the analyses it was decided for the future to perform only 5 cycles of measurements per sample. In fact, during the third in-field experiment at the BNFL Plant (Capenhurst), each of the six analyses of production UF_6 samples took about 2 hours only. The enrichment of the samples ranged from 2.12 % ^{235}U to 5.56 % ^{235}U. Only in a few cases was the quality of the measurements slightly affected by the reduced number of double ratio measurements. The relative precision was, in general, better than 0.05%. Considering the limited number of measurements per sample, the relative accuracy was also very satisfactory.

A summary of the results obtained is presented in Table I.

The measurement of the minor isotope ratios were only carried out in order to have an indication of the possibilities of the computer program.

Conclusions

The results clearly show that in-field measurements of UF_6 isotopic ratios can be performed by a quadrupole mass spectrometer with satisfactory precision and accuracy for samples enriched up to about 6%. It must be pointed out that under field conditions the repeated venting and the insufficient cleaning of the analyser system could affect the quality of the measurements.

The planned improvements of the pumping units and of the inlet system (automatic computer controlled operation) will lead to a steadier high vacuum and better reproducibility of the measurement conditions. They will also improve the performance and yield to a smaller size of the whole equipment making it easier to transport.

Modifications of the computer programs are in preparation to allow analyses of UF_6 samples enriched up to about 90%. In the meantime suitable improvements of the electronics are under study.

Owing to the promising results obtained, the Euratom Safeguards Directorate, Luxembourg, is considering to test the prototype instrument under inspection conditions.

Note: A detailed description of the instrument and a thorough discussion of the results obtained will be presented at the Internation Symposium on "Recent Advances in Nuclear Material Safeguards" to be held in Vienna from 8 to 12 November 1982. The paper has been assigned with the number IAEA-SM-260/51.

Table I Results obtained during three in-field experiments for $^{235}U/^{238}U$ measurements

SAMPLE	QUOTED $^{235}U/^{238}U$ (percent)	MEASURED $^{235}U/^{238}U$ (percent)	ΔC (percent)	
1st IN-FIELD TEST:				
BCMN 29M/2	0.4402	0.44658 ± 0.0008	+ 1.45	(a)
BCMN 32M/1	0.7254	0.73201 ± 0.0006	+ 0.91	(a)
BCMN 31M/2	3.3859	3.4129 ± 0.0019	+ 0.80	(a)
BCMN 43M/1	5.3245	5.3768 ± 0.0008	+ 0.98	(a)
2nd IN-FIELD TEST:				
U1	0.34449	0.34570 ± 0.0005	+ 0.35	(b)
U2	0.72639	0.72314 ± 0.0004	− 0.45	(b)
U3	3.42453	3.4221 ± 0.003	− 0.071	(b)
3rd IN-FIELD TEST:				
B1	2.1680	2.1726 ± 0.0013	+ 0.18	(b)
B2	2.7902	2.7810 ± 0.0014	− 0.32	(b)
B3	3.4340	3.4350 ± 0.0013	+ 0.029	(b)
B4	3.5294	3.5306 ± 0.0007	+ 0.034	(b)
B5	4.2113	4.2199 ± 0.0018	+ 0.20	(b)
B6	5.8906	5.8947 ± 0.0022	+ 0.07	(b)

$$\Delta C \quad \frac{\text{MEASURED QUOTED}}{\text{QUOTED}}$$

(a) without sample-to-standard calibration

(b) with sample-to-standard calibration

Safeguards Reporting by Use of Word-Processing Equipment

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Introduction

It all started recently following a number of breakdowns of my secretary's typewriter. She said: "I cannot use this machine any longer, its quality is simply too poor".

I realized very quickly that word-processing equipment could facilitate her tasks (letters, mailing lists, minutes of meetings, symposium papers, etc.).

What about safeguards reports ? She types ICRs, PILs, MBRs, etc. A copy of the PIL for each MBA is continuously being updated. Copies (about 125 pages of PIL and other inventory listings) are given to inspectors. They are also used at PITs. The final version at PIT is sent to Euratom together with MBR.

We report for all Danish installations: two research reactors, a HEU and LEU fuel-fabrication MBA, a storage MBA, a hot cell, an ore-processing installation, various laboratories, a number of private firms possessing depleted U, and a mine.

My accounting system is manual. During the 17 years I have been in charge of the safeguards office, I have reported according to seven different agreements, sometimes two in parallel. Many changes have been made even during "stable" periods. Therefore, it has not been practical to develop a universal computer accounting and reporting system covering also operational data.

Word-processing equipment

My preliminary investigation indicated that the large computer at Risö was inconvenient for ordinary secretarial tasks. Considering the effort needed to introduce the system, the speed of work, the dependence on computer staff union, security, etc., I found that we should purchase our own system. Then I studied systems using storage of data on discs. Even a small disc would be sufficient for, e.g. a particular PIL for a particular MBA, or, for that matter, any other task of my secretary.

Three types of equipment could facilitate our work:

a) equipment without calculating capability and without file,
b) equipment with calculating capability but without file, and
c) equipment with calculating capability and with file.
Data transmission by phone is possible by adding a unit.

Type a) equipment

PIL updating would be facilitated very much. Furthermore, for the many local transfers of materials a master copy with common ICR information supplemented by individual information would facilitate the work of my secretary as well as that involved in my drafting and auditing. Errors found at my audit could easily be corrected.

For safety reasons a unit with two discs should be used so we can copy important information and store the copy separately. Both discs would be subject to security measures.

Type b) equipment

This equipment could also facilitate updating of the special inventory lists including information on all such bottles containing UO_2 powder or UO_2 pellets, which together constitute a batch. After a change the total quantity of UO_2 is calculated and multiplied by the U-factor and the enrichment to give the new batch data that should be introduced in the PIL.

Type c) equipment

If PIL data are introduced directly into the file, then the equipment may :

- type PIL according to batch names,
- type PIL according to location,
- produce a disc with data in accordance with Euratom electronic standards, so the Euratom computer can read the information without punching,
- alternatively, transmit PIL data by phone to Euratom (in which case security measures may be taken),
- transfer PIL data to computers used by inspectors (they could calculate the total quantity subject to inspection as well as a sampling plan),
- calculate lines of information for MBR.

Similarly, transfer of ICR data to the file could lead not only to typing on ordinary forms to be used as transfer documents, but also to electronic transmission on a monthly basis to Euratom. Furthermore, the equipment could calculate lines of information for MBRs.

For my secretary the transfer of data to the file could be quite different from normal typing on forms, but perhaps the transfer could be done automatically without any delay. The time factor will be very important in selecting the equipment.

The essential problem for me is: How can I perform the independent audit of the input to the file? At present I am auditing the updated PIL pages, while my secretary is typing other reports. If she uses the equipment all the time, how can I audit? A rapid printout of changes will be necessary.

For the Euratom the direct transfer of data to the computer will speed up data processing and reduce punching and auditing costs. If a financial arrangement could be made, e.g. on the basis of the number of data, then I would be more interested in getting type c) equipment.

An inadequate Euratom audit occurred some years ago, when Euratom forgot to punch a couple of pages of my PIL before transmission to the IAEA. This led to a MUF of several kilos of HEU in the IAEA computer, and a comment from the IAEA inspector.

For the inspector an automatic evaluation of the actual inventory is better than a sampling plan made in Luxembourg on the basis of old data.

In conclusion, I feel that type c) equipment should be compatible with the Euratom computer in Luxembourg. Preliminary discussions have taken place, and I have not yet made any final decision with respect to the equipment I should try to get.

What now ?

At a meeting on 31 August this year, the ESARDA co-ordinators found that investigation of data transmission should have a high priority and that the problem was of general interest. Hopefully, co-operation between interested parties could be established and lead rapidly to useful results.

Verification of Filling Levels in Tanks by Means of the D_2O Tracer Technique, and Use of a SPECTRAN 677 IR Process Photometer

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Keywords
Reprocessing
Tank recalibration
Verification of tank levels
D_2O tracer technique
Analysis of D_2O in H_2O
Infrared photometry

Abstract

For the verification of tank filling levels in calibration of the dip tube used for measurements in reprocessing, application of tracers (Li, Mg, D_2O) is one of the eligible means. The routine application of the tracer measuring technique, however, has been impeded so far by the relatively high costs and by the very expensive analysis of the tracer samples. As an alternative to the mass spectrometric tracer analysis exclusively used until now, application of an infrared process photometer for measurement of the heavy water (D_2O) concentration in H_2O was tested at the Karlsruhe Nuclear Research Centre (KfK); tracer tests with D_2O in an 800 l ring tank had already been performed earlier by the Karlsruhe Reprocessing Plant (WAK) which means that reference values were available. The results of assay show that the photometric method is clearly superior to the mass spectrometric assay both in terms of acuracy and costs and hence the first method is suited for application on a routine basis.

Introduction

In a reprocessing plant the process tanks are remotely handled and emptied. The filling levels of the tanks are determined by the familiar dip tube method. This system of measurement has to be recalibrated at given intervals with water.

Within the framework of international nuclear Safeguards it is of interest to have an independent method of verifying a tank calibration and tank filling, respectively. Such a verification may be done, e.g. by analysis of the degree of dilution of an added amount of a tracer substance.

In 1975 WAK used for the first time on a trial basis heavy water (D_2O) as the tracer. However, the routine application of this technique of verification was impeded by the fact that the analytical system available at the time (mass spectrometric determination of D_2O) required the addition of at least 1% of D_2O, related to the final filling level of the tank, in order to provide adequate information and that it is consequently too expensive for Safeguards.

Investigations[1] performed at the Institute of Radiochemistry of the Karlsruhe Nuclear Research Centre using a SPECTRAN 677 IR process photometer supplied by Bodenseewerk Geosystem GmbH, Überlingen, have shown that the photometric assay of the D_2O concentration in H_2O is likewise suitable for verifying tank calibrations. Compared with the mass spectrometric analysis the method indicated offers the advantage of reducing the D_2O demand by the factor 30 whilst the accuracy of measurement is clearly improved.

Experimental

For calibration work an 800 l ring tank was available having the same dimensions as the tanks used in the WAK process part for the head-end measurements.

The calibration was performed with deionized water whose density was in conformity with that in the table according to Küster-Thiel; successively, portions of about 8 kg were added which were weighed on a calibrated METTLER precision balance. The water consumption was verified by means of a cylindrical piston counter.

Figure 1 shows the experimental set-up used for the tracer measurements.

The heavy water was added at a filling level of 90 liters and calibration subsequently continued. After approximately 100 liters each had been added the tank content was mixed by air sparging. Then the samples were taken. The amounts of water and D_2O lost in sampling were corrected in the subsequent computations.

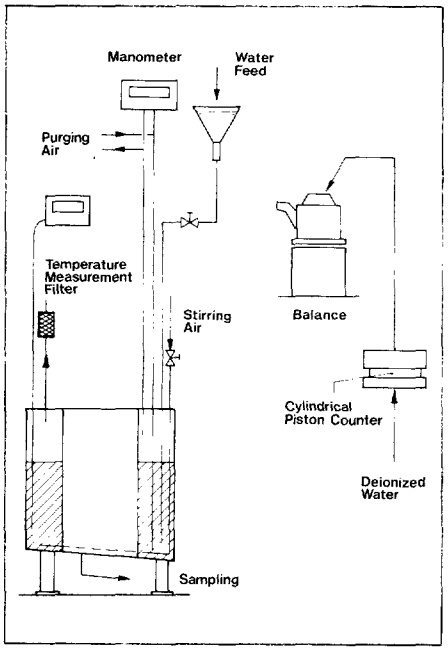


Figure 1 Set-up for calibration of a head-end tank, type WAK

The quality of calibration was assured by comparison with the preceding runs without tracer (linear regression of the pairs of values of pressure/volume, CUSUM error diagram).

Analytical System

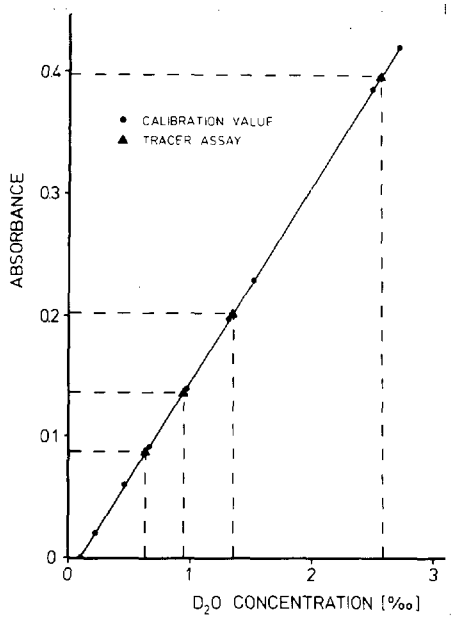
For the analysis of the H_2O/D_2O isotope mixture an infrared process photometer supplied by Bodenseewerk Geosystem GmbH, Überlingen, type SPECTRAN 677 IR[2] with a thermostat-controlled liquid process cell[3] was used. The method of

1. Analytical application work in industrial practice, based on the SPECTRAN process photometer, is being performed under the Technology Transfer programme of the Federal Ministry for Research and Technology (BMFT) and funded by the Technology Transfer Coordination Office of the Karlsruhe Nuclear Research Centre.
2. Under license of the Karlsruhe Nuclear Research Centre
3. Developed by the Karlsruhe Nuclear Research Centre

measurement underlying the process photometer is based on the selective absorption of infrared radiation in liquid and gas mixtures, respectively, by the component to be determined.

The process photometer works as a single-beam system using the bi-frequency technique and thus ensures a high long-term stability of the measurement signal and of the zero point. Non-specific changes in transmission characteristics of the liquid cell are automatically compensated for by this method.

Figure 2 *Calibration plot for the evaluation of tracer samples with the process photometer. (To improve transparency, not all values have been entered.)*



Evaluation of the Tracer Samples

To determine the D_2O content with the process photometer a calibration plot was recorded in the range from 143 to 2719 vpm D_2O in H_2O, including the concentration range of the tracer samples. The calibration mixtures were prepared from the same heavy water batch (D_2O content 99.75 mol.%) from which the D_2O was supplied into the ring tank. The D_2O content of the fully deionized water used for ring-tank calibration and the D_2O content of the fully deionized water for the calibration mixtures were assumed to be 1/7000. The qualitative analysis of the D_2O content in the fully deionized waters provided agreement within the accuracy of measurement.

Figure 2 shows a calibration plot; fitting to the measured values was done by linear regression. The D_2O concentration in the tracer sample was determined in each case from the measured absorbance value and the regression line.

With the samples taken three independent series of measurement were performed in accordance with Fig. 2. In Fig. 3 the relative deviation from the nominal value of the analytical value so determined has been plotted as a function of the tank content. The error bars entered show the relative standard deviation of the respective mean value of the tracer assays. It is evident that up to a tank filling of approximately 400 liters the measured negative deviations of the analytical values from the nominal value lie outside the uncertainty of measurement of concentration evaluation. Beyond approximately 600 liters of tank content the relative error of meausurement is about 1-2 per mil.

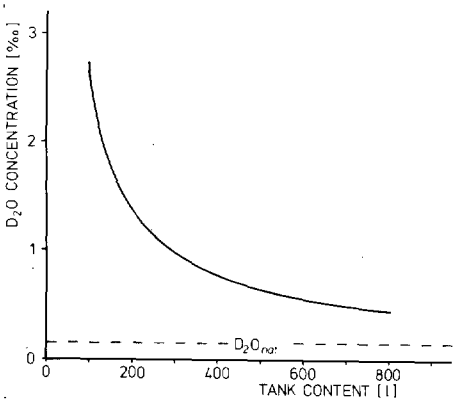
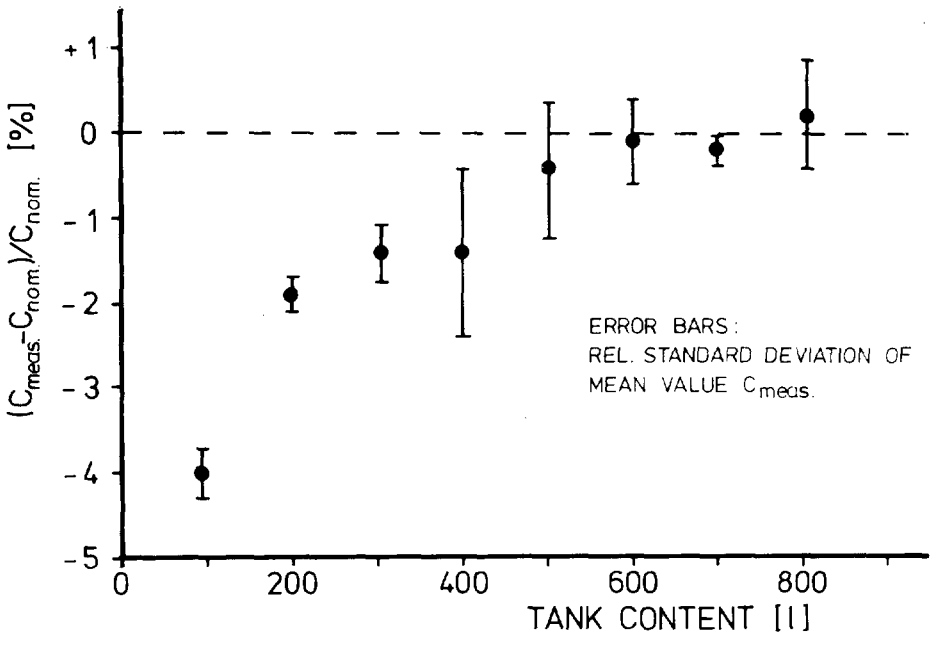


Figure 4 Dependence of the D_2O concentration on the tank content after addition of 0.25 litre D_2O with a content of 99.75%

However, the accuracy of measurement attainable in the assay of tracer samples is not identical with the accuracy with which the tank content can be determined because only the difference between the D_2O concentration of the tracer sample and the natural D_2O fraction in the deionized water can be taken into account. In Fig. 4 the D_2O concentration of the liquid in the tank has been entered after 0.25 l of D_2O (99.75%) had been added. The dashed line indicates the natural D_2O content of the water. It can be recognized that for higher filling levels the natural D_2O content makes up a relatively high fraction of the D_2O concentration; for 800 l this fraction amounts to approximately 30%. In this case, the uncertainty in the determination of the tank content increases accordingly by about 50% as compared with the uncertainty in the assay of the D_2O concentration; it attains approx. 0.3% = approx. 2.4 l (for 800 l of filling).

It was verified in separate experiments to which degree residues of HNO_3 and uranyl nitrate which might still be present in process tanks flushed for recalibration are susceptible of influencing the results of measurements. For aqueous HNO_3 solution a rise in absorbance by $\Delta E = 4x10^{-5}$ per mg/l HNO_3 was found. For a solution containing 10 mg/l uranium a rise in absorbance by $\Delta E = 1x10^{-4}$ was found. If one assumes that approximately 10 ml/l HNO_3 and about 10 mg/l uranium each are still present in a process tank in the course of recalibration, the resulting systematic error in the determination of the D_2O content is less than 1 ppm D_2O

Figure 3 : Relative deviation of the measured D_2O concentration in the tracer sample C_{meas} from the nominal concentration C_{nom}.



and hence within the limits of errors of measurement.

Discussion

The results of measurements performed to determine the D_2O content in tracer samples using a SPECTRAN 677 IR process photometer show that this analytical method is clearly superior to the mass spectrometric assay previously tested by WAK, both under economic aspects and with a view to the accuracies attainable.

To achieve a satisfactory accuracy of measurement only a D_2O feed of 0.4 per mil of the tank volume is required for process photometric analysis, as compared with at least 1% in mass spectrometric analysis. Moreover, the purchasing and maintainance costs of a process photometer are clearly lower than those of a mass spectrometer. The accuracy attainable in the determination of the tank content is approximately 3 per mil which means that a volume of 800 l can be determined with an accuracy of about 2.4 l.

The lower D_2O contents of the tracer samples as compared to the nominal values, found in the range up to 400 l, are very probably attributable to inadequate mixing of the tracer with the deionized water added. This interpretation is supported particularly by the fact that with increasing tank content, i.e., after a longer stay time and better mixing of the tracer, the absolute difference between the nominal and measured values decreases. On the other hand, the relative accuracy of measurement of the method is higher than under conditions of great dilution of the tracer, which is due to the linear relationship existing between absorbance and concentration in the process photometer with increasing tracer concentration, i.e., lower tank content.

Therefore, adequate mixing of the tracer must be ensured when tracer experiments are to be performed. This may be achieved, e.g., by adding the tracer in portions which have already been diluted with deionized water. Moreover, it is reasonable to homogenize the tank content by pumping. The D_2O analytical technique presented here shows spectral cross sensitivities with respect to nitrate, which, in the presence of rather high amounts of, e.g., uranyl nitrate, may result in considerable errors of measurement. Therefore, volume verifications in tanks containing uranium or plutonium products are not possible, in principle, by a photometric method using a D_2O tracer measurements.

But tank recalibrations can be made with the desired accuracy of measurement provided that after flushing the residual contents of HNO_3 and uranium, respectively, do not exceed 10 mg/l each.

Activities of the ESARDA Working Groups

Low-Enriched Uranium

A.G. Hamlin, NMACT, Harwell
Convenor of the ESARDA Working Group on Safeguarding Low-Enriched Uranium Conversion / Fabrication Plants

Historical Review

This Working Group was first conceived in 1977 as a model of a new departure for ESARDA - a form of Working Group in which the principal input would be from the operators within the nuclear industry who bore directly the burden of the application of Safeguards. After a period of initial orientation, the first meeting of the Group was held in March 1978, and attracted the support of all of the LEU Fuel Fabricators in the EEC.

As had been intended, the Group immediately addressed itself to the practical problems of Safeguards application, starting with the assessment of operators' mutual problems, and limitations, either as a basis for their resolution among the members themselves, or for their definition as items requiring research effort for solution. By the end of the second meeting, a work plan had been established aimed, as far as was possible, at establishing common procedures for Safeguards requirements among the different operators. This work plan envisaged that members would:

a. exchange information on the reliability, accuracy, precision and range of application of the various measurement practices in use in the plants of the participating LEU Fuel Fabricators. The best practices could then be identified and steps taken to encourage the common adoption of these methods where possible in existing plants and the information could be used as input to the design of new plant.

b. define a programme of the topics which would be actively considered by all plant operators in parallel with the measurement practices comparison above could be undertaken. These topics should all be of such a nature that an exchange of data and methods was likely to lead to the adoption of common standards at least cost (e.g. LEMUF analysis, verification procedures, PIT procedures, etc.).

c. define the areas where further development is necessary and attempt to minimise the duplication of prototype effort in these areas (e.g. NRTA systems, enrichment storage facility design, NDA techniques, sampling methods, sealing mechanisms, etc.).

d. exchange information on Nuclear Material control system uranium accounting procedures and documentation with a view to standardising source data and easing data processing problems.

e. agree methods for Physical Inventory, sealing of common items, etc., to reduce lost time when full PIL is necessary.

The Group believed that this information exchange had little commercial significance and would be to the advantage of all concerned.

Co-operative development along these lines extended over the five subsequent meetings of the Group which tended to become more widely spaced as the amount of individual effort required by members increased. The development is best described by topic than by chronological sequence.

Measurement Practices

The Group found that methods and standards of measurement of nuclear material were very similar among members, but that there were weaknesses in the evaluation of systematic error and in the use of common reference materials.

The Group suggested that ESARDA should establish a group to examine the supply of sintered uranium oxide pellets (up to 10,000) as a common reference material[1] and that the operation should encompass the necessary inter-laboratory consensus to establish the accepted composition of the standard. A group should also be established to examine the errors introduced by the sampling process and to develop methods for their control.

Subsequently contact with JRC-Geel established that a limited supply of standard UO_2 pellets was being prepared, but not on the scale envisaged as necessary by the Group.

In order to allow the Group to organise an intercomparison experiment between the members to establish the importance of systematic error between laboratories, JRC-Geel was requested to prepare a larger quantity of sintered pellets and to relate this to the previously mentioned reference material. In the meantime this standard reference material has been certified and is available as EC RM No. 110.

Preliminary consideration of the sampling problem[2] showed that the errors could not be defined and practical experiments were conducted by JRC-Ispra in co-operation with a member of the LEU Working Group. These indicated that the error of sampling bulk uranium oxide was negligible, but the Group decided to conduct further experiments under more rigorous conditions. These confirmed that under suitable conditions sampling errors could be controlled at a negligible level.

The Working Group considered it worthwhile to extend co-operative investigation to the establishment of the errors involved in sampling UF_6 but considered that the errors involved in sampling more heterogeneous materials were best investigated by individual members for report to the Group.

The Group also examined the weighing process which plays a large part in the control of nuclear material in fuel fabrication and organised a substantial collaborative programme involving the circulation of a set of seven standard weights among members under a statistical programme to be operated by JRC-Ispra.

Verification Schemes

The verification of inventory and flows in a LEU fuel fabrication facility is a complex undertaking which can be very expensive in process interruptions if carried out by conventional methods.

Initially, members of the Group made individual studies to assess the possibility of detecting empty or partially empty fuel pins in complete fuel assemblies representing the outcome of the process.

Considerable collaborative experience was exchanged relating to the use of rod scanners, which are largely used for quality control, as an accountancy tool. The results were encouraging and the assistance of the ESARDA Working Group on Non-destructive Analysis was sought for a fuller evaluation. This Group concluded that the present variety in fuel pins and rod scanner types precluded the measurements obtained being of adequate accountancy standard for individual rods but that forbatches of rods a very satisfactory average figure was obtainable.

This approach implied that, after verification of the rods, a system of containment and surveillance would be needed to maintain continuity of knowledge until the rods were finally incorporated in fuel assemblies, and that the assemblies themselves should be sealed for life. After some discussion of this problem, and consideration of JRC developments and members' practical experience, the Group decided that the proposed JRC developments were too complex and offered little advantage over more conventional approaches. They decided to seek further advice from the ESARDA Working Group on Containment and Surveillance.

The Group assessed the problem of verification of input to the plant and were of the opinon that input is best verified at the output of the relevant conversion plant where the containers are available individually for verification of content and can be sealed. The question of specification of suitable seals was referred to the ESARDA Working Group on Containment and Surveillance.

The Group also devoted attention to possible means of reducing the cost and intrusiveness of inventory taking in other parts of the production cycle. These divided broadly into administrative techniques and the preservation of knowledge by sealing and other containment and surveillance techniques.

Administrative techniques involved collective discussion of criteria for preparation of the list of inventory item(s), exchange of experience on the taking of the list and procedures suitable for preparing sampling plans for verification.

One member of the Group subsequently reported on an inventory carried out much in line with the criteria discussed. The inventory was effective, but a number of areas for further development became evident and the whole procedure was intrusive. As a result, other members developed alternative approaches resulting in less intrusion into operations. One of these which was well received by the Group was a proposal for partial inventory taking, which provided a reasonable degree of verification without interrupting production.

Sealing and containment techniques for inventory items were discussed in the ight of practical conditions of LEU Fuel Fabrication, but little of practical use in this context was found to be available. The feasibility of developing suitable containers was referred to the ESARDA Working Group on Conainment and Surveillance, who produced an assessment for consideration by the Group. This called for considerable finance and awaits a decision on implementation. It appeared to be more appropriate to the intensity of Safeguards applied to MOX fuel.

Instruments capable of verifying quantities of LEU non destructively were limited, and only the PHONID instrument developed for HEU by JRC-Ispra appeared to have any potential. The Working Group requested experiments to be conducted at the facility of one member to assess its performance with LEU.

Computer-based Nuclear Materials Accountancy

The Group has exchanged information on individual accountancy systems and has provided a forum for the discussion of programs of Safeguards interest developed by JRC-Ispra for the calculation of MUF and LEMUF. Some operators have installed the later versions of these programs (NUMSAS) with a view to exchanging experience in their operation. These installations revealed a number of modifications that would be desirable,

1. DOC/142/59/79 Need for the preparation of sintered pellet UO_2 reference material for uranium determinations. H, Muntau and M. Bresesti
2. HM/mh/1722/117/79 - Ispra 8.6.79 'Inquiry on Sampling Techniques"
3. Criteria for Preparation of the List of Inventory Items (Luxembourg 16.5.79 - XVII E-4/md.

some deficiencies in the available supply of information, particularly of a statistical nature, and some basic doubts on the availability of the data necessary for statistical accounting.

A number of members of the Group were initially operating near-real-time accountancy systems. The Group appreciated the possible benefits of this system both as an operational control tool, and as a means of providing a means of Safeguards control that would be more effective than total inventory taking. Exchanges on this topic eventually resulted in a contractual agreement between JRC and one member for the development of a NRTA system under practical conditions with a view to making the resultant experience available to all members. This system was to be established in the light of guidance from the Group discussions.

The last full meeting of the Working Group was held in June 1980. At this point the Group felt that a fairly long suspension of meetings was desirable.

The Working Group itself was a pioneering venture, and represented a first serious attempt:

a. to get nuclear operators in a particular sector of the industry together to discuss the application of Safeguards as a common problem, the solution of which would benefit from a common approach.

b. to interface operators directly with Safeguards inspectors and both jointly with research workers as a means of defining problems and starting to solve them.

During its series of meetings, the Group had defined a number of problems to which its various members were devoting attention. These investigations would take some time to mature and would, the Group felt, be best conducted by small rather informal sub-groups having closely related interests. In fact this method of working, if successful, could influence the future structure and balance of the Group which some members felt needed some adjustment if it was to continue effectively. The Group therefore adjourned and was reconvened in September 1982.

At this meeting, the Group reviewed progress and it became clear that, while this had been substantial in some areas of direct interest to operators, such as computerised systems of material control and in quality control of the weighing operation, in other areas of more marginal interest to operators, progress had been less. The lack of progress had been due to lack of motivation and consequent difficulty in devoting scarce operational resources to the problems.

In the light of this experience, and after some discussions as to its purpose, the Group concluded that it had a valid function in promoting co-operation and avoidance of duplication in the continuing development of practical systems of material control in LEU fabrication. Evidence before the Group suggested that such systems would tend to become more and more similar, and would not become a major differentiation between operators as regarded competitiveness, therefore mutual benefit should result from the continuance of the group. As the general standard of material control became more uniform, the research and development representatives would be able more clearly to identify worthwhile areas of activity and the Euratom representatives would have information enabling them to formulate more efficient and effective Safeguards approaches, thus perhaps also leading to better definition of R & D projects.

The Group agreed to a further round of meetings based on the following Action Plan.

1. The Working Group would be composed of representatives from plant operators, Safeguards Authorities and research establishments.
2. A round of discussions would be held at a series of meetings over the next two years at various establishments, to exchange and demonstrate experience gained in Nuclear Materials Management by members of the Working Group.
3. The Working Group would endeavour throughout its works, to ensure that Safeguards procedures can draw largely, if not wholly, upon comprehensive Nuclear Materials Management Systems.
4. The orking Group would restrict actions to the pragmatic development of current Nuclear Materials Management technology taking due account of the requirements of both plant operations and Safeguards. The work commitment of the plant operators would be limited to the transfer of knowledge and assisting each other to implement improvements at least average cost to all members. This may involve some mutual training of specialists.

The role of the Research and Inspection Organisations would be to advise upon the developments in Nuclear Materials Management Systems and identify Safeguards aspects where R & D is required.

Some published articles on the Working Group are given below.

1. The ESARDA Approach to Facility Oriented Safeguards Problems. R. Stewart et al., 20th Annual Meeting, Institute of Nuclear Materials Management, Albuquerque 1979
2. The ESARDA Working Group on Safeguarding Low-enriched Uranium Conversion/Fabrication Plants, S. Saiger, 21st Annual Meeting, Institute of Nuclear Materials Management, Palm Beach, 1980
3. Activity of ESARDA Working Group on Safeguarding Low-enriched uranium Conversion/Fabrication Plants, S. Saiger, ESARDA Second Symposium on Safeguards and Nuclear Material Management, Edinburgh 1980.

Mathematical and Statistical Problems

M. Franklin and A. Musto
Convenor and Secretary respectively of the Working Group

The Mathematical and Statistical Working Group held its first formal meeting at Karlsruhe on 21 and 22 June 1982. The meeting finalised as far as the participants were concerned the terms of reference of the working group. The final form was as follows.

Draft Terms of ESARDA Working Group on Mathematical and Statistical Problems

The aims of the ESARDA Mathematical and Statistical Working Group is to act as a forum for technical discussions between plant operators, Safeguards authorities and research people in order :

1. to study problems of practical urgency identified by other ESARDA working groups. Involvement to be only at the express request of such other groups. It is understood that the Working Group will involve critically reviewing existing techniques and identifying gaps in statistical or mathematical modelling knowledge. The Working Group can also study general mathematical modelling and statistical problems associated with materials accountancy but not associated with any particular type of plant,
2. to make recommendations on mathematical and statistical techniques for Safeguards application,
3. to stimulate R & D in order to produce new mathematical or statistical techniques for Safeguards problems,

4. to stimulate the preparation of mathematical and statistical software for Safeguards problems.

It is understood that the interest of the Working Group will cover the Safeguards fields of measurement, MUF analysis, verification, containment and surveillance and the optimisation of Safeguards effectiveness. The Working Group at any point in time will work on a list of specific tasks which reflect Safeguards needs at that point in time.

The above terms of reference were distributed to the convenors of other working groups and the ESARDA coordinators for information and comment.

The rest of the meeting was taken up by presentations or dealing with:

- optimisation verification in MOX plants
- simulation of NRTA measurement system
- statistical methods for reprocessing input verification
- statistical methods for seal verification.

Optimisation of Verification in MOX Plants

Mr Van der Stricht (Safeguards Directorate CEC) presented a paper entitled : "Problem Statement: Study of the Relation of the Results of the Fortnightly Verification and Physical Inventory Taking for Mixed Oxide Fuel Fabrication Plants". This paper gave a detailed description of the current Safeguards approach for achieving assurance against both protracted and abrupt diversion in a reference mixed oxide fuel fabrication plant. For the stated reference case, the problem is to determine what safeguard evaluation relevant to protracted diversion can be drawn after a period of six months based on :

- the verification of the physical inventory at the beginning of the material balance period
- the verification of all the inventory changes at the boundary of the MBA
- the Safeguards activities carried out to meet short term requirements i.e. "attribute" measurements to detect gross defects.

The quality of conclusion should be compared with those achievable after a 6-month material balance, closed with a full physical inventory. If the two statements are not equivalent one should try to establish which other activities could be added to those already performed for the detection of abrupt diversion so as to obtain equivalence. Mr Van der Stricht felt that any proposed statement about protracted diversion, derived from the activities designed to detect abrupt diversion, should be based on sequential analysis of MUF-D. It is hoped to have a presentation for the next meeting of the approaches which have been considered to compare the effectiveness of the two verification strategies. This will serve to identify the modelling and/or computational difficulties if any.

Simulation of Near Real Time Accountancy Measurement System

Mr Nägele (KfK) presented a paper on the simulation of the measurement system of a NRTA system of a large reference reprocessing plant. The purpose of the study is to test the performance of sequential testing procedures for MUF analysis. One of the interesting features of the KfK approach is the fact that individual measurements are not simulated. Instead the MUF time series is directly simulated. This is achieved by using a set of hypothetical material movement accouts and measurement history information to compute synthetically the variance-covariance matrix of the MUF time series. This variance covariance matrix plus any hypothesised loss scenario become the parameters for a direct simulation of the MUF time series. Individual realisations of the MUF time series are subjected to sequential tests. The generation of realisations and their sequential testing are repeated to evaluate the performance of the tests. This technique of creating the variance-covariance matrix as a basis for simulating the MUF time series results in a great saving of computational effort.

Statistical Methods for Reprocessing Input Verification

Mr Foggi, on behalf of the former ICT Working Group, presented a paper entitled "Problems Related to the Evaluation of the Data Measured at the Reprocessing Plant Input". During evaluation of data from the ICE experiments the various laboratories involved used differing statistical tests and procedures to identify outliers. Mr Foggi requested advice from the Working Group on selection of the statistical procedure which would yield the "best" results for the Safeguards Authorities.

As a result of the discussion on this topic the Group set up a task group to read the ICE report and give an opinion on the different methods used. Mr Van der Stricht emphasised that he wished to see emerging from this opinion a clear indication of use to a Safeguards Authority.

The ICE report has subsequently become available and it is hoped for the next meeting of the Working Group to give a presentation by the authors on the findings presented in the report. This will allow the Working Group to get a better appreciation of the issues involved.

Statistical Methods for Seal Verification

Mr B. Causse d'Agraives presented a paper entitled "Problems and Requirements Related to the C/S Techniques Used for the Verification and Identification of Seals or Structures". Mr Causse d'Agraives described seals under development at Ispra which are identified by their ultrasonic signature. The problem is to compare two or more such signatures and decide whether or not they are from the same seal. The problem is the same for various other means of seal identification such as surface topography.

Mathematically there are two main problems which the Working Group was asked to consider. These are, how to decide when the two signatures are in phase and how to quantify the similarity between the two signatures. Statistical modelling which treats the signal as a constant function with a measurement noise imposed is likely to be inadequate since it is unlikely to recognise changes of amplitude or shift which are intuitively recognisable. Mr Butterfield suggested that a variation on the sign test may be suitable for choosing a starting point and agreed to write a note on this which he will present at the next meeting.

Date of the Next meeting

The next meting will be held on 15-16 November 1982 at the delegation of the Commission in Vienna.

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