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

Volume II: Preparation, Characterization and Transport of the Test Samples

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KERNFORSCHUNGSZENTRUM KARLSRUHE
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CENTRAL BUREAU FOR NUCLEAR MEASUREMENTS
Geel

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THE IDA-80 MEASUREMENT EVALUATION PROGRAMME ON MASS SPECTROMETRIC ISOTOPE DILUTION ANALYSIS OF URANIUM AND PLUTONIUM

VOLUME II:

PREPARATION, CHARACTERIZATION AND TRANSPORT OF THE TEST SAMPLES

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* Die Arbeiten wurden unter diesem Projekt durchgeführt, das Ende 1983 formell eingestellt wurde.

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ABSTRACT

A description is given of the test samples for the IDA-80 interlaboratory measurement evaluation programme.
The preparation is described from the delivery of authentic reprocessing plant input samples (WAK Karlsruhe), their treatment at the IRCh-KfK and their further preparation and bottling at CBNM-Geel.
All test samples have been characterized with best methods and instrumentation available at CBNM Geel and NBS Washington.
Joint certified values for U/Pu element and isotopes content of a real-life and of a synthetic input test material were established, to serve the evaluation of participant's results.
Full details of packaging, transport requirements and transport formalities in order to dispatch the samples world wide are also given.

Das IDA-80 Meßprogramm zur Bewertung der massenspektrometrischen Isotopen-Verdünnungsanalyse von Uran und Plutonium

Vol. II: Vorbereitung, Charakterisierung und Transport der Test-Proben

ZUSAMMENFASSUNG

Die Herstellung der Testproben für das analytische Interlabor-Meßprogramm IDA-80 wird eingehend dargestellt.
Das Probenmaterial wurde der Eingangslösung einer Wiederaufarbeitungsanlage (WAK, Karlsruhe) entnommen und zunächst im IRCh/KfK, danach im ZBKM/Geel aufgearbeitet und abgefüllt.
Alle Testmaterialien wurden vom ZBKM/Geel und dem NBS/Washington unter Einsatz leistungsfähigster Meßgeräte und -methoden analysiert. Die von diesen beiden Instituten gemeinsam zertifizierten Werte für U/Pu-Element- und Isotopengehalte der aus der Wiederaufarbeitungsanlage stammenden Testmaterialien sowie die einer Lösung ähnlicher Zusammensetzung dienen der Auswertung der von den Teilnehmerlaboratorien erzielten Meßergebnisse.
Darüber hinaus werden die Verpackung und die für den weltweiten Transport der Proben erforderlichen Formalitäten im einzelnen beschrieben.

Table of Content

Chapter II: Characterization of the Test Samples

W.H. Wolters, M. Gallet, W. Lycke, P. De Bièvre
Central Bureau for Nuclear Measurements, Geel

L.A. Machlan, J.W. Gramlich, J.D. Fassett, K.R.Eberhardt
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Chapter III: Compilation of technical and administrative requirements for the shipment of the test samples to the participating laboratories

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The conscientious work and devotion in the painstaking preparation, characterization and transport of the test samples by the following persons of CBNM is gratefully acknowledged:

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* on leave at CBNM from the Western Australian Institute for Technology, Perth

Two further volumes of the final report on the

"IDA-80 Measurement Evaluation Programme on Mass Spectrometric Isotope Dilution Analysis of Uranium and Plutonium"

are published:

Volume I, KfK 3760/EUR 7990e
"Design and Results"
(W. Beyrich, W. Golly, G. Spannagel, KfK
P. De Bièvre, W.H. Wolters, CBNM)

and

Volume III, KfK 3762/EUR 7992e
"Compilation of Evaluation Data"
(W. Beyrich, W. Golly, G. Spannagel, KfK)

PREFACE

Interlaboratory measurement evaluation programmes without carefully characterized test samples, have limited use. They only provide the size of the interlaboratory spread of the results. They do not allow a statement on how much a participant - or the grand mean value of all participants - deviates from a "value, closest to the truth", the latter being defined as the best value obtainable with "state of the art" measurement instrumentation and procedures where each step is under full control, is repeated many times and is rigidly assessed for all possible uncertainties, both random and systematic.

In order to be able to draw as many useful conclusions as possible, the preparation and characterization of the IDA-80 test samples, has been carried out with as large a scientific effort (several man-years) as has ever been invested in an interlaboratory test programme.

The results are "reference values" for the test materials with overall uncertainties which are considerably smaller than the interlaboratory spreads.

Chapter I: Preparation of the Test Samples

by

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1. Introduction

At the meeting of the ESARDA Working Group on Destructive Analysis in Harwell in October 1977, a first plan for a programme "IDA-78" was discussed and approved. In December 1978 the sample preparation was completed.

However, a few weeks before shipment, inhomogeneous plutonium distribution in samples of reprocessing input solution in form of solid particles had been observed. Consequently, both CBNM and IRCh-KfK examined closely the input samples intended for IDA-78 and arrived at the conclusion that participants could cast some doubt on the homogeneity and identity of the samples ready for distribution to the participants although such lack of homogeneity and integrity were in fact very unlikely.

Nevertheless on proposal from CBNM, the ESARDA Working Group decided unanimously at its meeting on November 1978 in Mol, Belgium to recommend preparation of new samples which would be proven unambiguously to be homogeneous. A new planning was then prepared on the basis of a LWR input material announced to be available at WAK Karlsruhe by the end of 1979. Also CBNM prepared U-Pu metallic spikes (Pu/U = 0.003) of 250 mg and metallic spikes (Pu/U = 0.005) of 30 mg suitable to spike LWR input samples.

In January 1980 we were informed that an FR 2 fuel material instead of LWR would be reprocessed and could be made available (see Table 1.1). It was decided to go ahead with the FR 2 input to avoid further delay of the programme (LWR fuel was expected towards the end of 1980 only).

The Feed solution was sampled on 1980.02.09 at WAK Karlsruhe and delivered to IRCh at KfK on 1980.02.11.

All samples were ready for shipment by the end of July 1980. In an additional experiment the influence of the particles was studied at IRCh on the unfiltered and undiluted input solution (see Section 4).

This chapter reports on the preparation of the samples according to the general IDA-80 lay-out (see Fig. 3.1). The test-sample preparation is given in Fig. 1.1. The sample preparation of all programme parts has been performed quantitatively starting from the diluted input solution B for part 1 and from a fission product free reference solution R for part 2. The comparison of the element concentration results of solution B, BS, B-dried and AS demonstrate a total spread of 0.06 % for Uranium and 0.1 % for Plutonium (see App. 1,2) except for the AS results on Plutonium which enlarged the spread to 0.4 %.

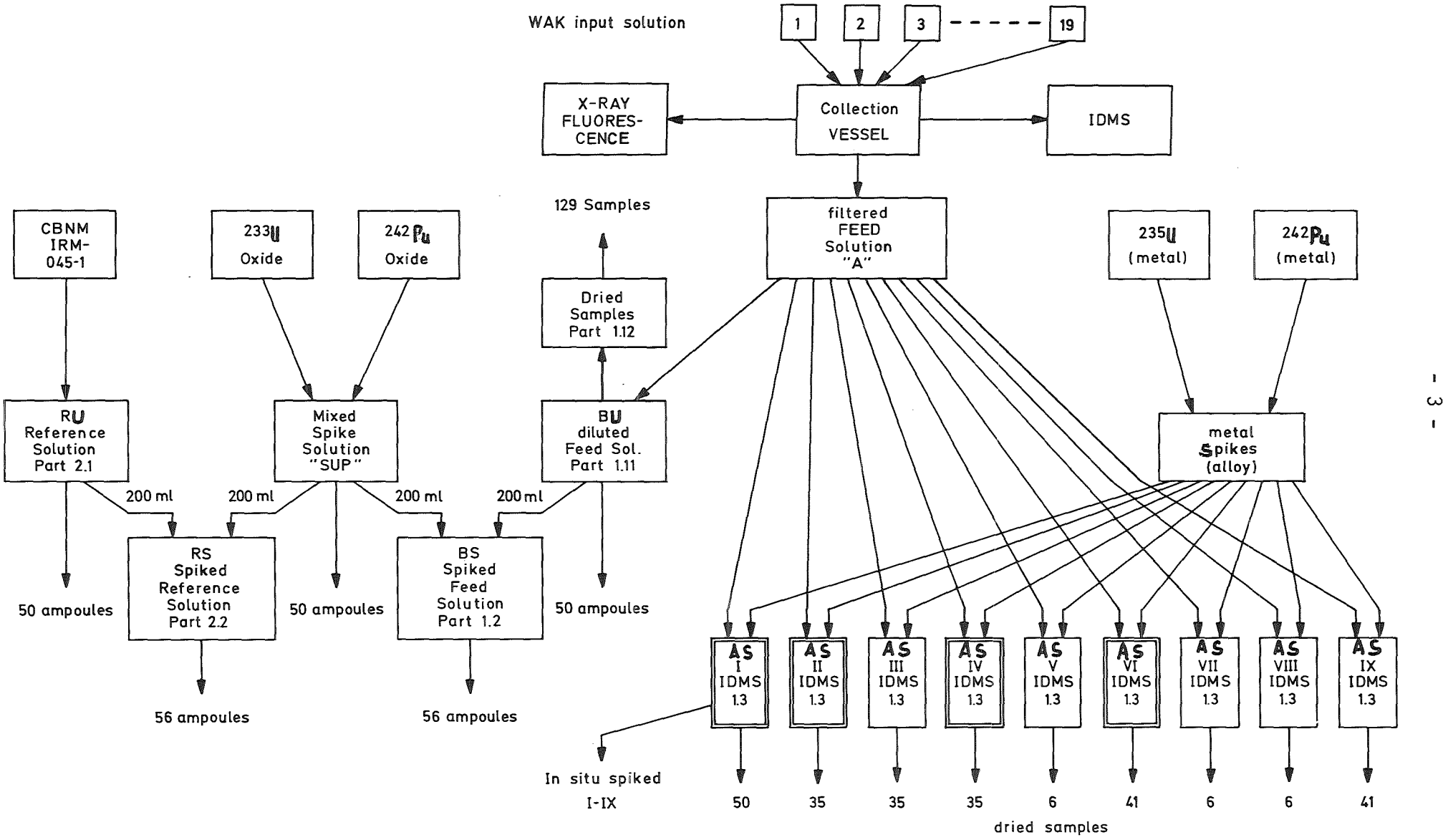


Fig. 1.1 IDA-80: Interrelation of Test Samples

Burn up			15 MWd/kg
U-concentration			250 g/l
Pu/U			$3.5 \cdot 10^{-3}$
Activity			7 Ci/l
Fission products			2.5 g/l
Isotopic Composition (mass %)			
^{234}U	0.080	^{238}Pu	0.195
^{235}U	0.579	^{239}Pu	69.924
^{236}U	0.177	^{240}Pu	25.130
^{238}U	99.237	^{241}Pu	3.189
		^{242}Pu	1.563
Pu =	0.585 g/kg		
U =	168.8 g/kg		

Table 1.1 Characteristics of the input solution (FR 2 fuel) established by WAK-Leopoldshafen

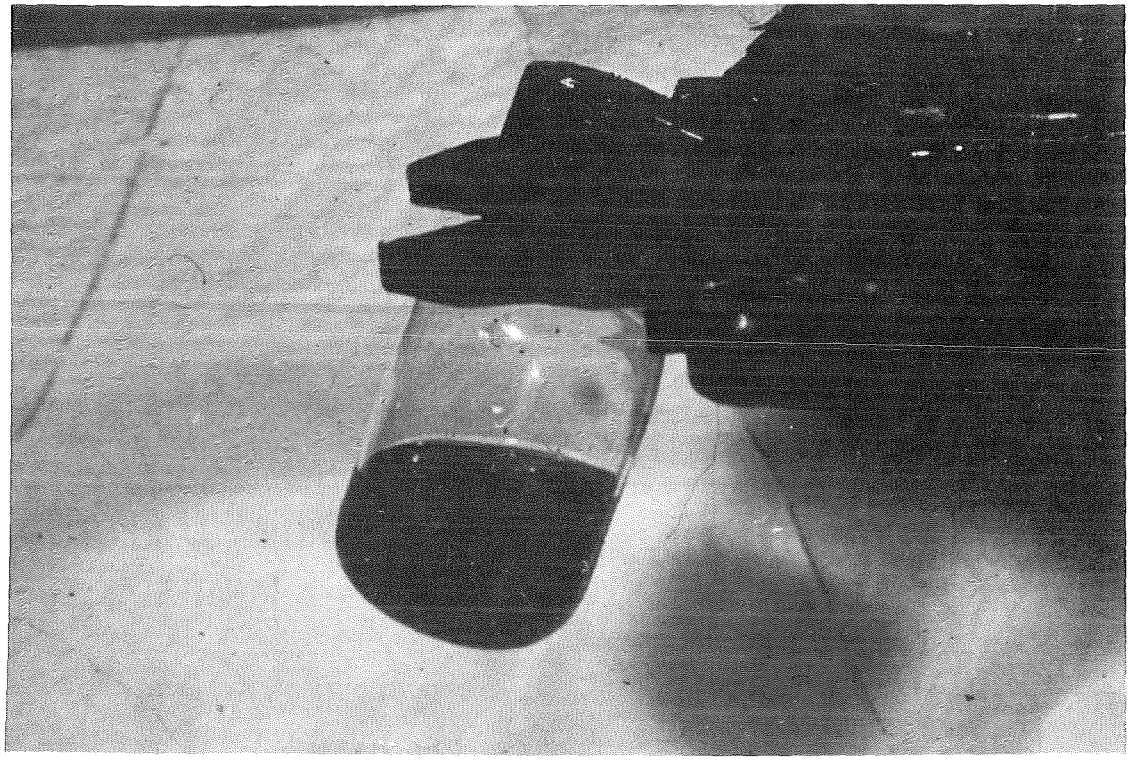
The comparison of the R and RS element concentration results demonstrates a total spread of 0.05 % for Uranium and 0.11 % for Plutonium (see App. 3,4).

2. Filtering of the input solution

The total input solution delivered had a dark brown colour and particles were observed. To ensure an absolutely homogeneous solution and representative samplings, 36 ml solution were filtered through a 0.4 μm Nucleopore filter (see Photograph 2.1). The residue was weighed and analyzed for Uranium and Plutonium by X-ray fluorescence (see Table 2.1 and Fig. 3.4.1).

Filtered A-solution		36.25	ml		
Undissolved residue	0.3 mg/ml	10.88	mg		
U-content of residue		0.47	mg	or	4.3 %
Pu-content of residue		0.022	mg	or	0.2 %

Table 2.1 Residue of the filtered input solution



Photogr. 2.1 Filtered input solution

The residue only contained 0.005 % U and 0.07 % Pu as related to the amount of starting material (9 g U and 33 mg Pu).

Conclusions:

1. The solution could be accepted as homogeneous.
2. The observed solid particles did contain negligible amounts of U and Pu only.

3. Preparation of the IDA-80 test samples

In describing these preparations, we will follow closely the general lay-out of IDA-80 as given in Fig. 3.1. incl. the abbreviations.

3.1 Delivery of the undiluted input solution

Nineteen bottles with a total of 61 ml were sampled over 4 hours in the WAK Karlsruhe on 9 February 1980. The sample bottles were packed in plastic tubes containing each 2-3 bottles and transferred to the IRCh laboratory on 1980.02.11/12.

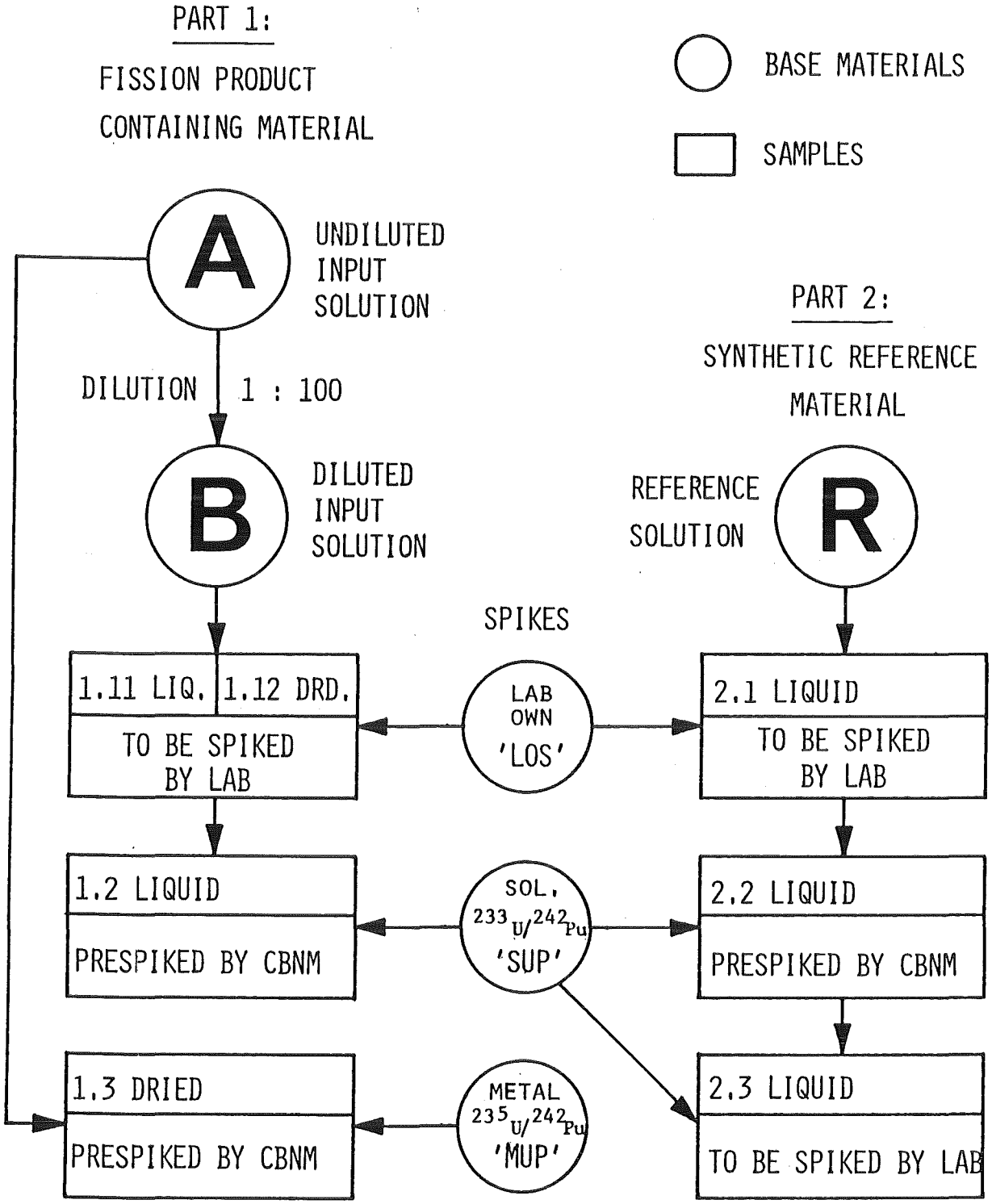


Fig. 3.1 General lay-out of IDA-80

The bottles were numbered as follow:

plastic tube 1	plastic tube 2	plastic tube 3	plastic tube 4
005045	005050	005043	005035
005046	005040	005034	005037
005038	005036	005044	

plastic tube 5	plastic tube 6	plastic tube 7
005033	005047	005053
005032	005039	005041
	005049	005051

The dose rates encountered are given in Table 3.1.

Distance	Background		Total		Sample	
	mrad	mGy*	mrad	mGy*	mrad	mGy
19 cm	480	4.8	1700	17	1220	12.2
50 cm	80	0.8	300	3	220	2.2
70 cm	10	0.1	120	1.2	110	1.1

* SI radiation unit 1 rad = 10^{-2} Gray

Table 3.1 Radiation dose rate of one sample bottle

3.2 Preparation and aliquoting of the diluted input solution BU.

9 ml of the undiluted input solution "A" was aliquoted and diluted to 900 ml at IRCh, KfK, to prepare the diluted input solution BU which was then transferred to CBNM for further treatment. Following sequence was applied:

1. 91 ml 6M HNO_3 were aliquoted and weighed.
2. 9 ml of the undiluted input solution were carefully weighed by difference. The solution was diluted with 91 ml HNO_3 and weighed again. The solution was carefully homogenized as solution B.
3. 800 ml 6M HNO_3 were weighed outside the hot-cell in a 1 l plastic-flask with screw-cap.

4. The solution of the first dilution step, see point 2, was added to the 800 ml HNO_3 to form the B-solution. The mass of the transferred solution was determined by difference weighing. The BU solution was homogenized and packed into a stainless steel lead lined container for shipment to CBNM (see Table 3.2.1).

	Mass in g
- 9 ml undiluted input solution, A	12.888 ± 0.001
- diluted with 91 ml 6M HNO_3 up to	121.448 ± 0.002
- transferred to weighed amount of 6M HNO_3 (810 ml)	121.342 ± 0.002
- total diluted input solution	1066.0 ± 0.2
- dilution factor	82.78 ± 0.02

Table 3.2.1 Preparation of the BU-solution

The work described under point 1 and 3 was performed outside, and point 2 and 4, inside the hot-cell.

3.2.1 Liquid BU samples, programme part 1.11

The aliquotation of the BU-solution was performed at CBNM according to the scheme given in Fig. 3.2.1.

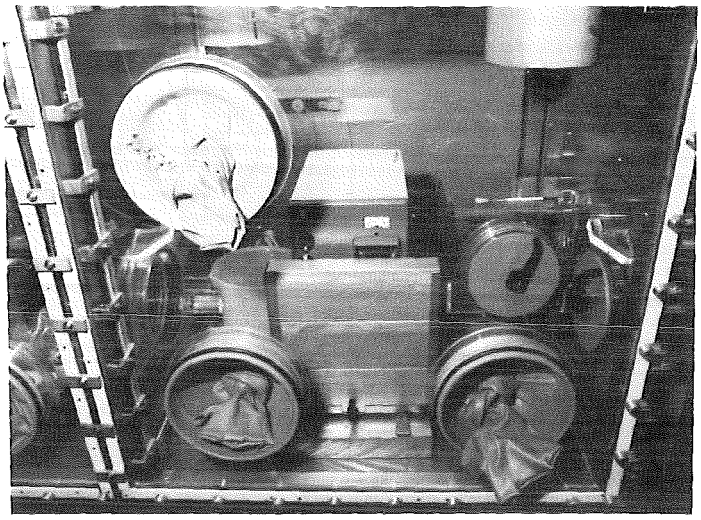
Fractions of 5 ml were taken from the BU-solution transferred into glass-ampoules and sealed immediately. The 50 ampoules were labelled:

BU 111 001 through BU 111 050

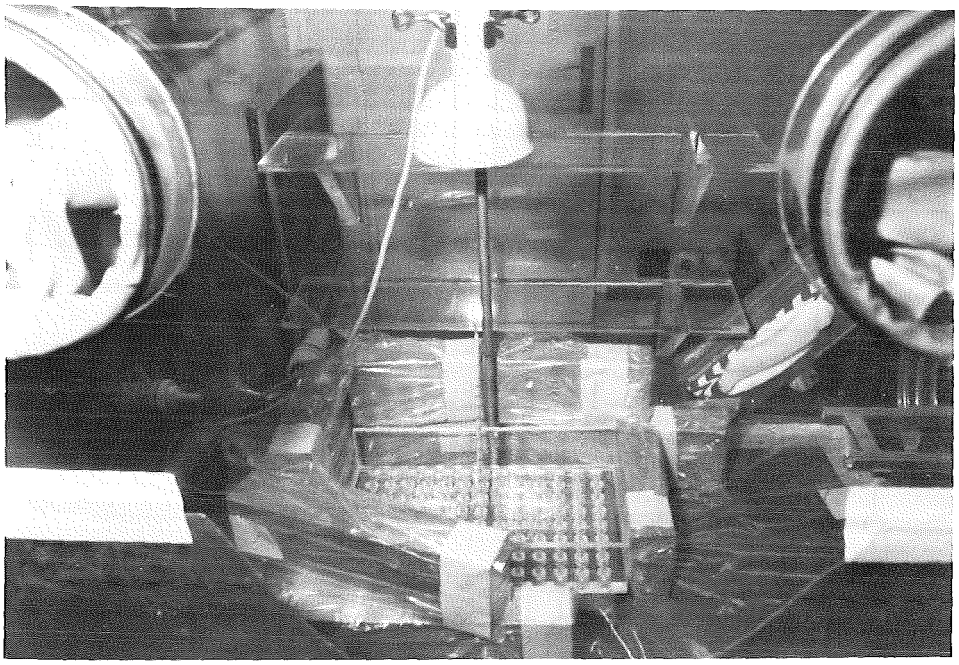
3.2.2 Dried BU samples, programme part 1.12 (see Fig. 3.2.1)

129 fractions of 1.2 ml were sampled by means of an automatic Metrohm burette in small vials (see Photogr. 3.2.2.1 - 3.2.2.2) and weighed on an analytical balance (see Table 9.1). All samples were slowly evaporated to dryness in a separate glove-box over 4 weeks at ~ 28°C. The vials were then closed with a plastic stopper and labelled:

BU I 112 001 through BU I 112 043
BU II 112 051 through BU II 112 093
BU III 112 101 through BU III 112 143 (see Table 3.2.2.1)



Photogr. 3.2.2.1 Lead protected analytical balance



Photogr. 3.2.2.2 Evaporation glove-box

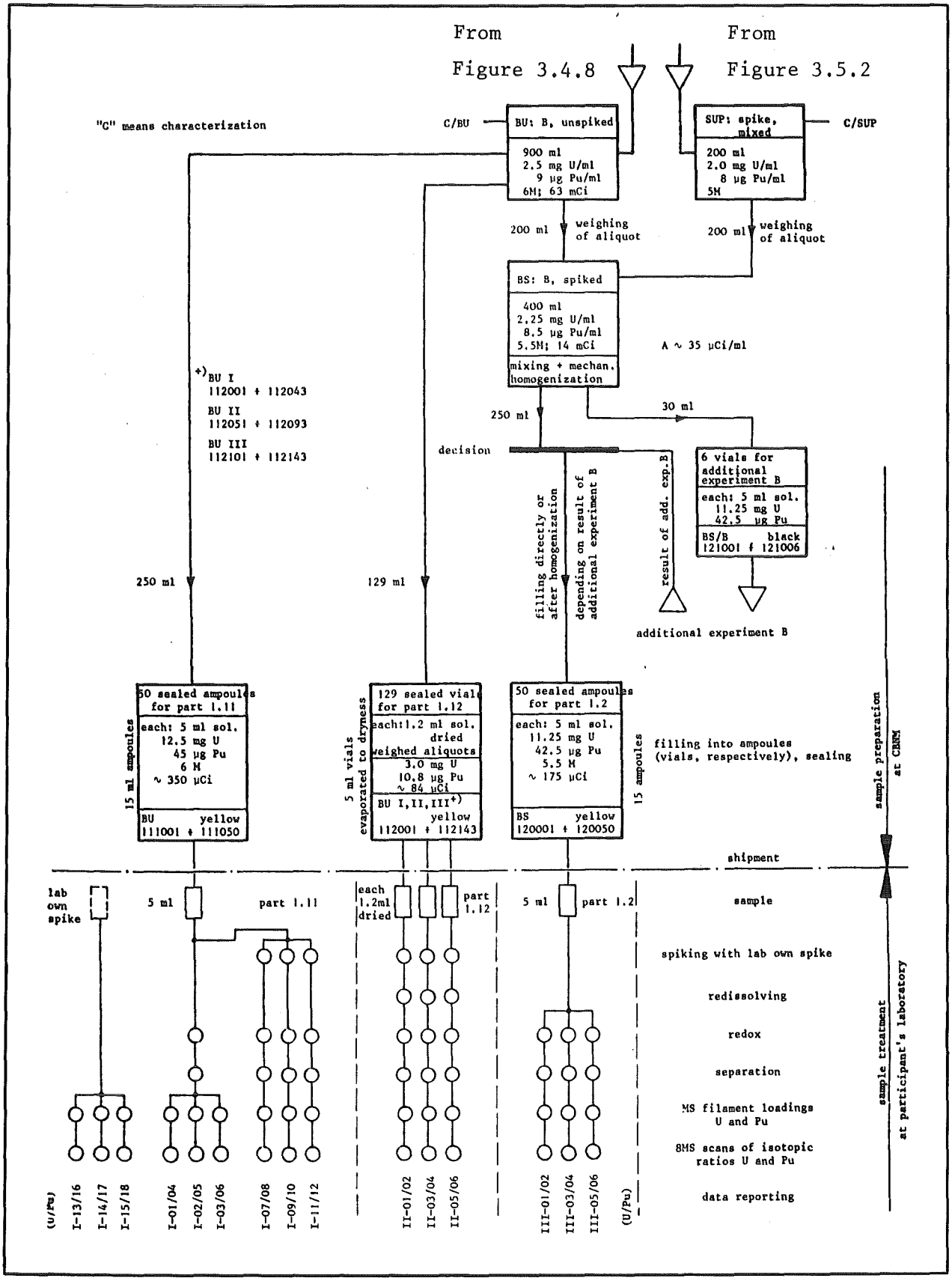


Fig. 3.2.1 Sample preparation for the BU-solution

Sample Number	Amount of solution(g)	Sample Number	Amount of solution(g)	Sample Number	Amount of solution(g)
112001	1.438 7 ± 5·10^{-4}	112051	1.436 2 ± 5·10^{-4}	112101	1.438 3 ± 5·10^{-4}
002	1.428 6 "	052	1.425 8 "	102	1.420 9 "
003	1.448 7 "	053	1.431 1 "	103	1.427 6 "
004	1.437 4 "	054	1.429 1 "	104	1.430 1 "
005	1.426 1 "	055	1.430 3 "	105	1.438 6 "
006	1.433 4 "	056	1.425 8 "	106	1.425 9 "
007	1.435 5 "	057	1.432 9 "	107	1.435 4 "
008	1.422 1 "	058	1.440 7 "	108	1.422 1 "
009	1.441 6 "	059	1.425 7 "	109	1.444 0 "
010	1.433 7 "	060	1.437 2 "	110	1.423 7 "
011	1.427 1 "	061	1.438 0 "	111	1.425 6 "
012	1.437 8 "	062	1.423 4 "	112	1.433 1 "
013	1.429 6 "	063	1.432 0 "	113	1.434 9 "
014	1.437 8 "	064	1.431 7 "	114	1.424 3 "
015	1.431 1 "	065	1.424 6 "	115	1.439 7 "
016	1.427 8 "	066	1.432 6 "	116	1.420 4 "
017	1.451 9 "	067	1.438 6 "	117	1.435 5 "
018	1.442 6 "	068	1.427 0 "	118	1.422 6 "
019	1.422 9 "	069	1.434 1 "	119	1.425 9 "
020	1.430 8 "	070	1.436 3 "	120	1.431 2 "
021	1.428 1 "	071	1.425 6 "	121	1.432 8 "
022	1.435 3 "	072	1.435 1 "	122	1.433 7 "
023	1.432 7 "	073	1.423 7 "	123	1.429 4 "
024	1.420 2 "	074	1.431 4 "	124	1.425 4 "
025	1.428 1 "	075	1.439 1 "	125	1.428 9 "
026	1.427 9 "	076	1.428 1 "	126	1.425 6 "
027	1.431 7 "	077	1.434 1 "	127	1.425 8 "
028	1.434 6 "	078	1.430 8 "	128	1.434 7 "
029	1.433 7 "	079	1.432 0 "	129	1.434 1 "
030	1.427 0 "	080	1.425 7 "	130	1.440 0 "
031	1.432 7 "	081	1.438 8 "	131	1.440 6 "
032	1.427 8 "	082	1.421 7 "	132	1.426 1 "
033	1.429 1 "	083	1.429 8 "	133	1.426 8 "
034	1.428 8 "	084	1.429 0 "	134	1.430 3 "
035	1.438 1 "	085	1.443 4 "	135	1.430 7 "
036	1.432 5 "	086	1.434 8 "	136	1.436 6 "
037	1.429 4 "	087	1.426 7 "	137	1.432 4 "
038	1.432 9 "	088	1.435 4 "	138	1.424 5 "
039	1.436 2 "	089	1.430 3 "	139	1.429 5 "
040	1.432 9 "	090	1.437 9 "	140	1.430 0 "
041	1.429 0 "	091	1.427 7 "	141	1.431 3 "
042	1.431 1 "	092	1.428 6 "	142	1.425 5 "
043	1.430 6 "	093	1.427 9 "	143	1.424 0 "

Table 3.2.2.1 Aliquoting of the BU-dried samples

Six B-dried samples have been characterized by CBNM only. Weighed fractions of CBNM spike were added to the dried sample. The dried sample was dissolved in 50 ml 6M HNO_3 in a 250 ml beaker glass. After homogenization and evaporation to 10 ml, the solution was transferred into a 20 ml glass vial. After evaporation to dryness the residue was dissolved in 600 μl 2M HNO_3 for purification. Because the solvent volume was three times greater than usual (200 μl is usual), the preconditioning procedures of the U and Pu was adapted. The element concentration results of the B-dried sample have not been used as a certified value. They have merely been considered as a value determined on the same type of test sample as those distributed and presenting the same difficulties in redissolution.

3.3 Preparation of the spiked diluted input solution BS, programme part 1.2 (see Fig. 3.2.1)

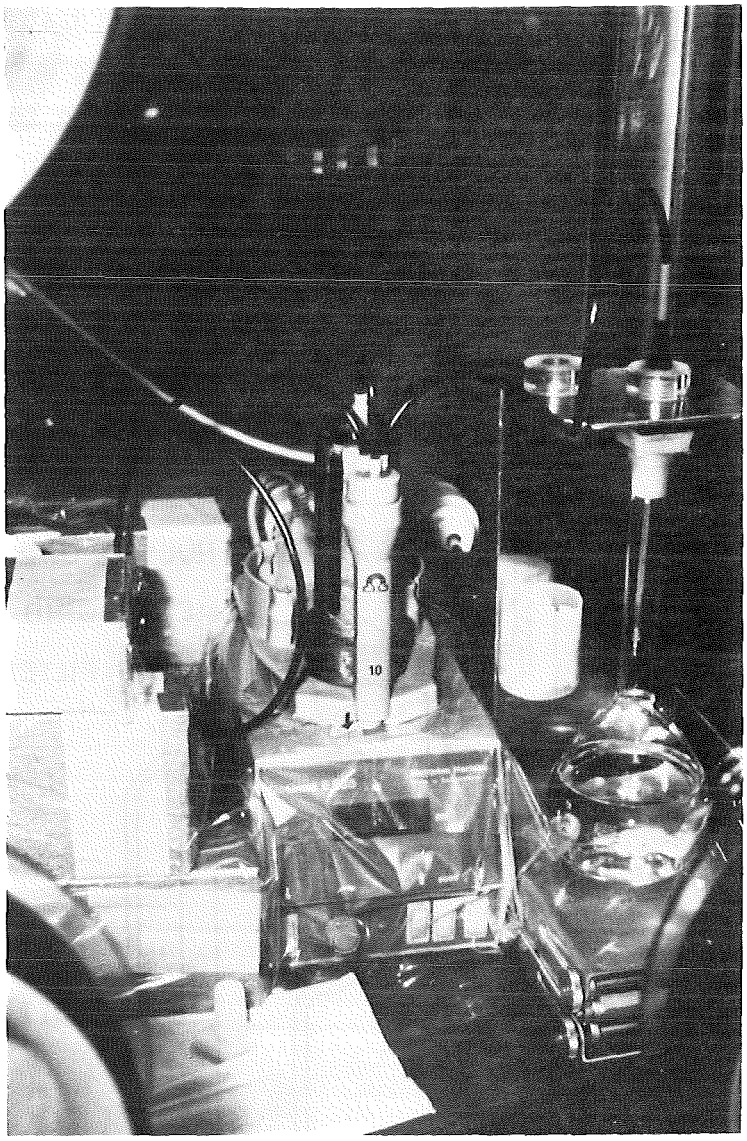
The preparation of the BS solution was performed by mixing weighed fractions of SUP-spike and BU-solution. The required end concentration was 8.5 μg Pu/ml and 2.25 mg U/ml at an acidity of 5.5M HNO_3. The isotope ratio targeted for $^{233}U/^{238}U$: ~ 0.8 and for $^{242}Pu/^{239}Pu$: ~ 1.16.

A weighed fraction of 200 ml SUP-solution was sampled in a 0.5 l Erlenmeyer flask. The BS solution was prepared by adding a fraction of 200 ml BU-solution by means of a remote controlled Metrohm burette (see Photograph 3.3.1). Weighed fraction of SUP and BU-solution see Table 3.3.1.

200 ml SUP-solution	231.37 g $\pm$ 0.01 g
200 ml BU-solution	238.89 g $\pm$ 0.01 g

Table 3.3.1 Fractions SUP and BU solutions

The solution was homogenized by means of a magnetic stirrer. The homogeneity was tested on six samples taken independently by measuring the isotope ratios $^{233}U/^{238}U$ and $^{242}Pu/^{239}Pu$. The homogeneity from this ratio measurements was found to be better than $\pm$ 0.05 %.



Photogr. 3.3.1 Remote controlled Metrohm burette for the aliquotation of the BU-solution

3.4 In situ spiking and aliquoting of the undiluted input solution A, programme part 1.3

This part was performed in the hot cell of KfK-IRCh at Karlsruhe by CBNM and KfK staff. All samples for the in-situ spiking were taken from the filtered clear solutions (see Fig. 3.4.1). Following sequence was applied:

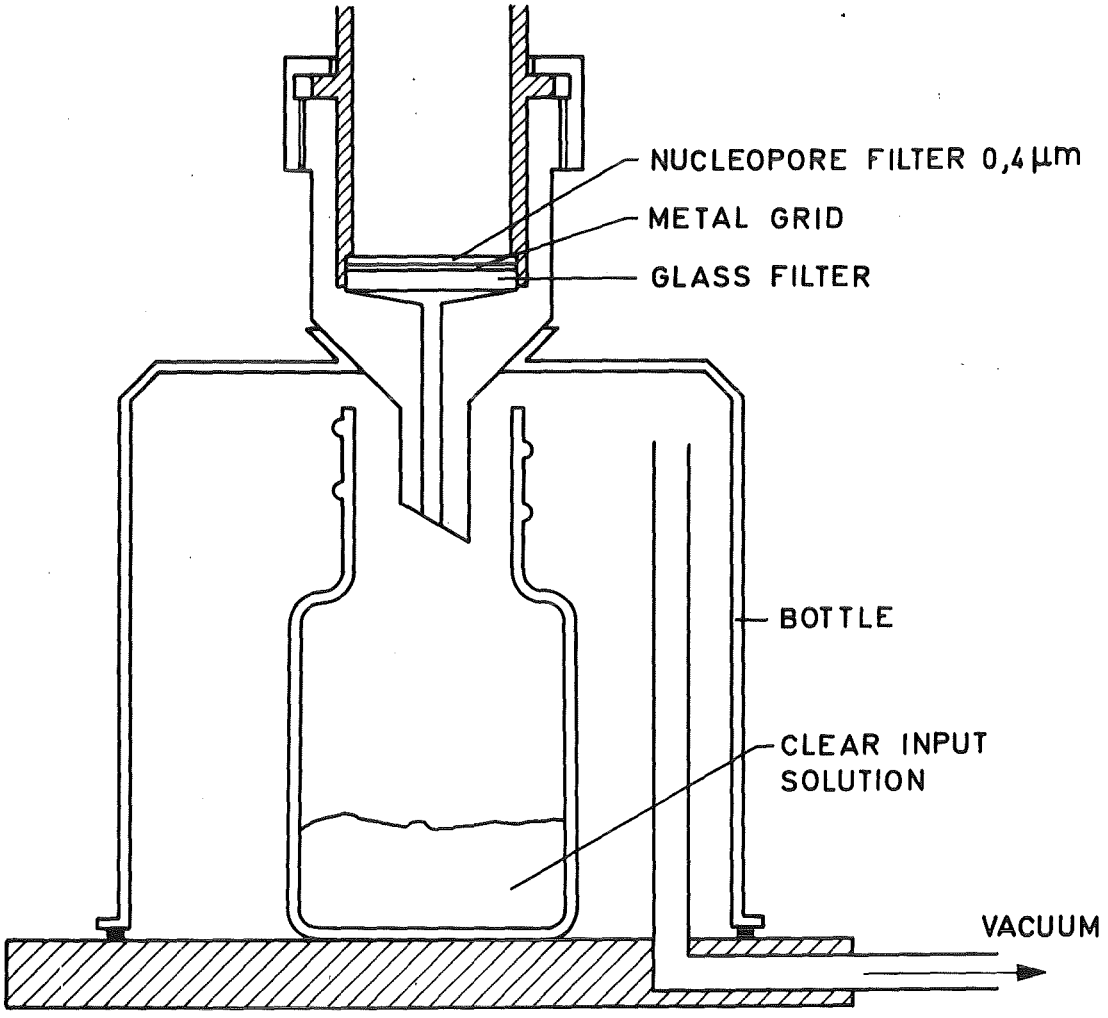
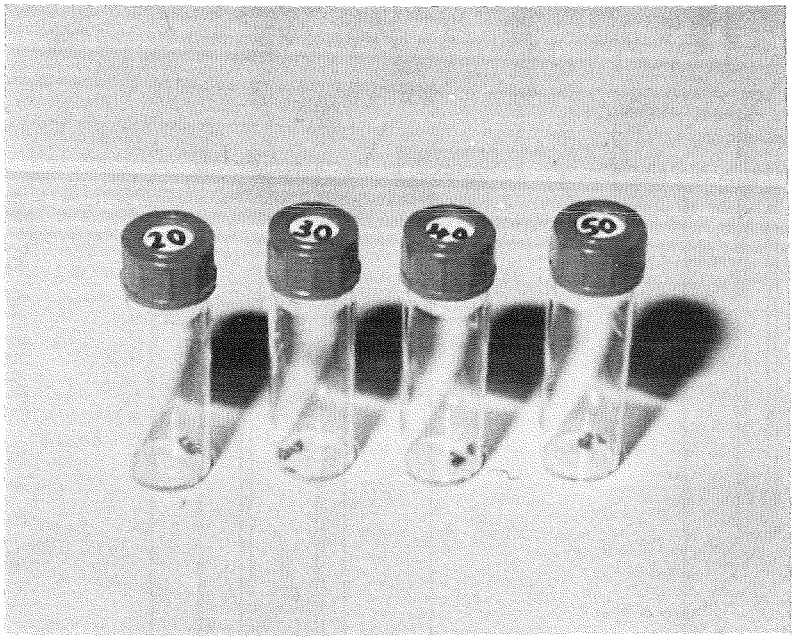


Fig. 3.4.1 Filtering device

a. The ampoule containing the metal U/Pu spike, was opened outside of the hot-cell. The mass of the spike was verified in the reaction vessel to be identical with the mass at the time of preparation. The spike mostly consisted of two small metal pieces (see Photograph 3.4.1).



Photogr. 3.4.1 Reaction vessel with metal spike

b. The reaction vessel with the spike was weighed in the hot-cell a second time. This provided unadditionally a second verification of the analytical balance.

c. One ml of the input solution was transferred into the reaction vessel by means of an Eppendorf-pipette and weighed immediately. A teflon funnel was used for the transfer of the solution. The precise weighings required that the neck of the vessel did not get wet in order to avoid evaporation errors. The input solution, acidity 3M HNO_3, did not react immediately. Reaction began after more than 1 hour (see Fig. 3.4.2).

d. The metal spike was dissolved in $\pm$ 7M HNO_3/0.1M HF for about 4 hours on an aluminium block maintained at 100°C (see Fig. 3.4.3) and monitored with a digital thermometer.

A total of nine samples were spiked in this way (see Table 3.4.1). In all spiked samples a whitish gelatinous precipitation was observed which did not dissolve in 6-10 molar nitric acid. All 9 solutions were diluted to 6 ml 6M HNO_3 and shaked for homogenization.

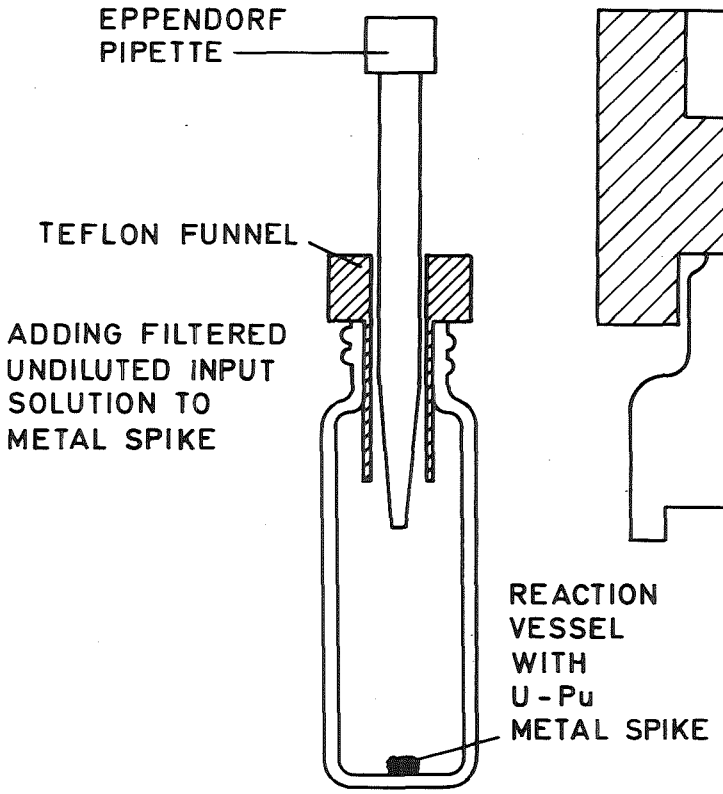


Fig. 3.4.2. Adding filtered undiluted input solution to metal spike in reaction vessel

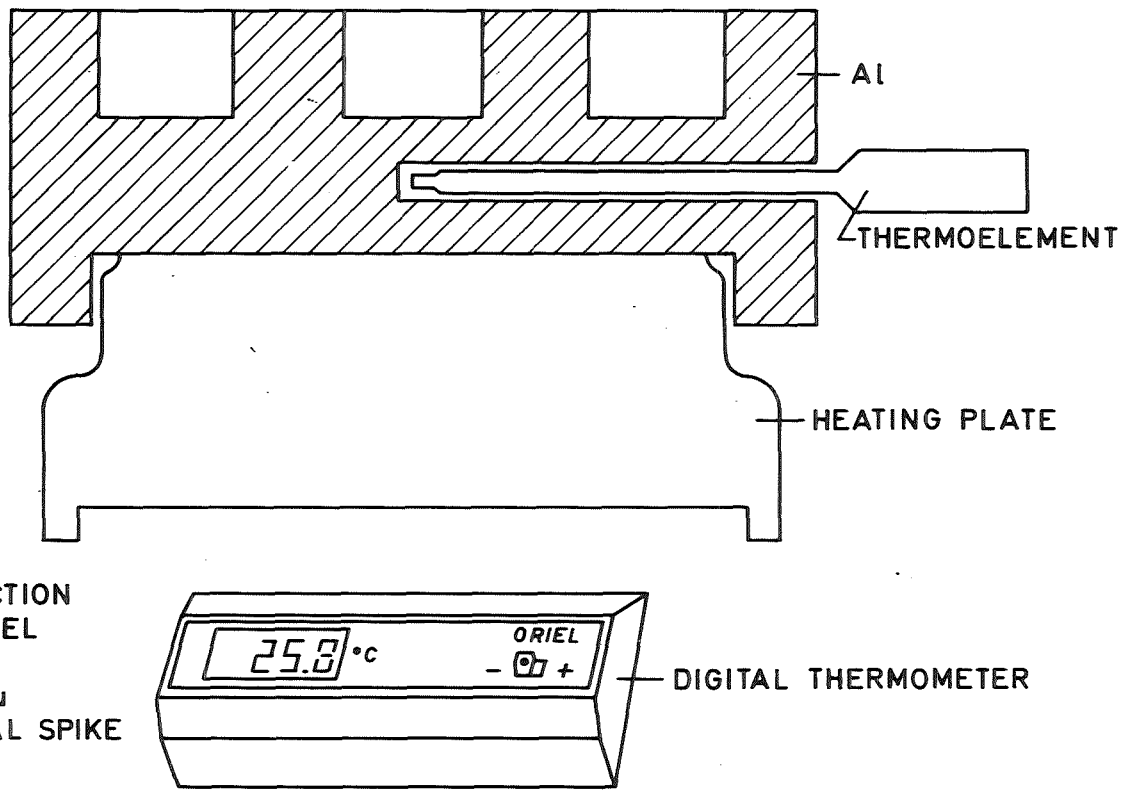


Fig. 3.4.3 Heating plate

Code of spiked sample	Flask number	Spike number	Amount of spike (mg)	Amount of sample (g)
AS I	55	117	$254.60 \pm 2 \cdot 10^{-2}$	$1.436\ 8 \pm 5 \cdot 10^{-4}$
AS II	25	102	245.30 "	1.428 3 "
AS III	40	107	245.77 "	1.441 4 "
AS IV	50	104	249.89 "	1.438 4 "
AS V	30	105	243.40 "	1.427 0 "
AS VI	15	101	250.41 "	1.430 9 "
AS VII	10	118	244.37 "	1.418 3 "
AS VIII	20	108	243.57 "	1.415 5 "
AS IX	35	110	244.59 "	1.438 7 "

Table 3.4.1 Masses of metallic spiked samples and input solution

After sedimentation the solution was filtered, with the exception of the last 0.5 ml, for use in the IDA sample aliquotation (see Figs. 3.4.4 and 3.4.5).

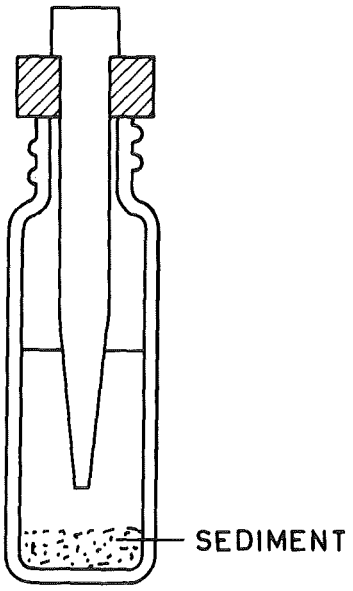


Fig. 3.4.4 Spiked solution after sedimentation

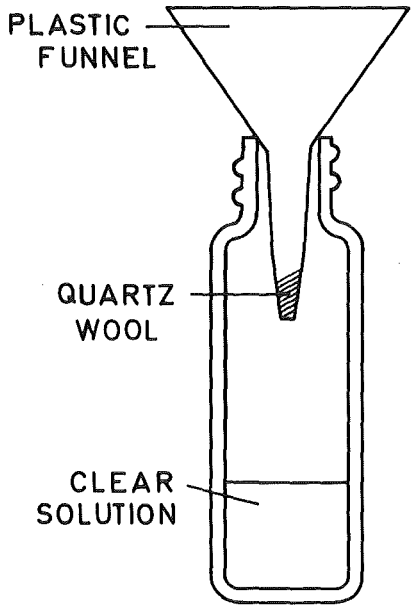


Fig. 3.4.5 Filtering of the spiked solution

Spiked samples AS I through AS IX were aliquoted in 100 μl samples, evaporated and closed with a plastic-stopper (see Figs. 3.4.6 and 3.4.7). The sample vials were labelled as follow:

AS I	130 101	through	130 150
AS II	130 201	through	130 235
AS III	130 301	through	130 335
AS IV	130 401	through	130 435
AS V	130 501	through	130 506
AS VI	130 601	through	130 606
and	130 611	through	130 645
AS VII	130 701	through	130 706
AS VIII	130 801	through	130 806
AS IX	130 901	through	130 906
and	130 911	through	130 945

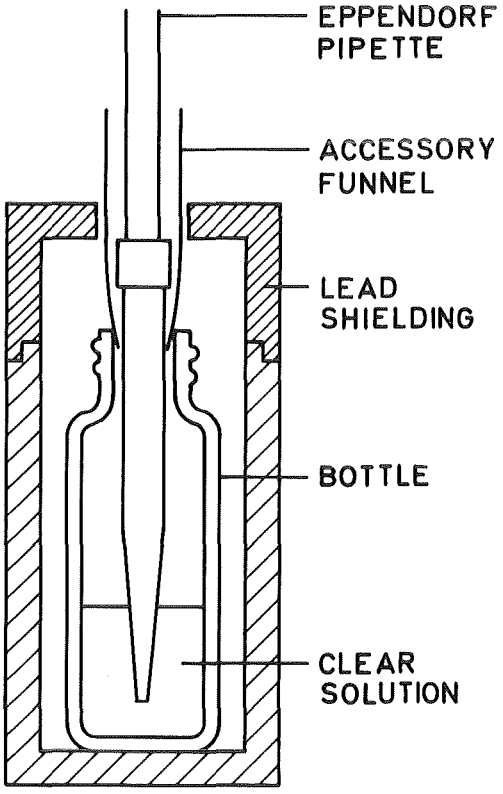


Fig. 3.4.6 Sampling of the clear solution

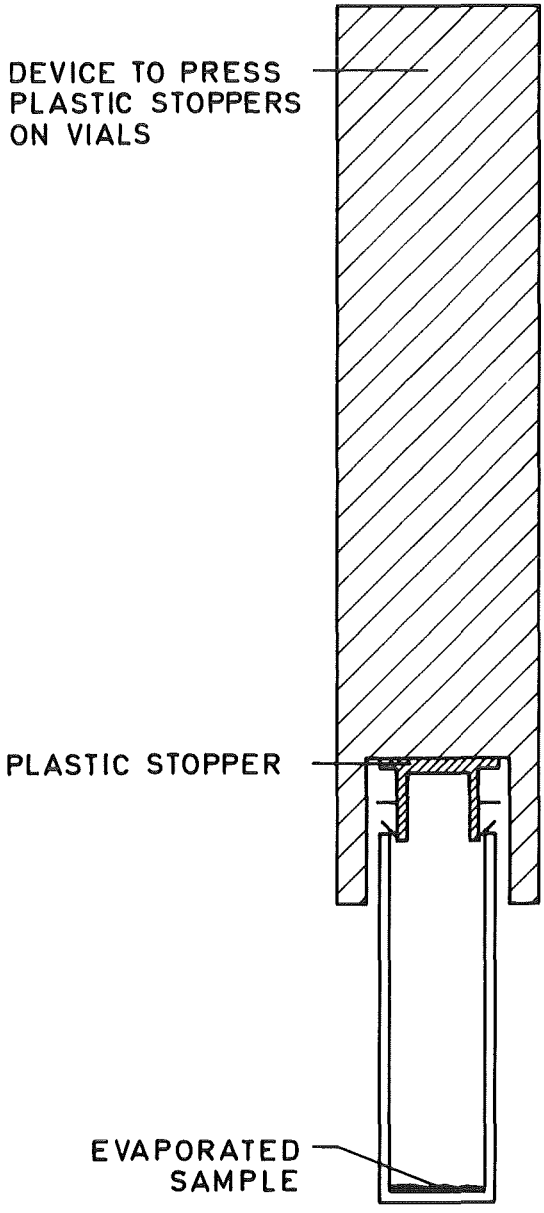


Fig. 3.4.7 Closing device

3.4.1 Chemical treatment of the residue found in the spiked input solution

0.5 ml of the spiked solutions, containing the main part of the particles, was treated with 1 ml concentrated HNO_3 / 0.1M HF solution and heated to 100°C for about two hours. One ml of this solution was filtered and 5 samples were aliquoted from each retreated sample. The samples were labelled as follows:

AS I	F55 R1	through	F55 R5
AS II	F25 R1	through	F25 R5
AS III	F40 R1	through	F40 R5
AS IV	F50 R1	through	F50 R5
AS V	F30 R1	through	F30 R5
AS VI	F15 R1	through	F15 R5
AS VII	F10 R1	through	F10 R5
AS VIII	F20 R1	through	F20 R5
AS IX	F35 R1	through	F35 R5

Measurements of the ratios $^{235}U/^{238}U$ and $^{242}Pu/^{239}Pu$ of both the filtered solution and the solution of the residue, as well as determination of Uranium and Plutonium by X-ray fluorescence showed that the metallic spike had dissolved completely (see Table 3.4.1.1).

- Mass of the residue of 4 spiked samples	< 0.2 mg
- U-content	$\pm\ 32\times10^{-3}$ mg
- related to the total U-content of the spiked sample	$\pm\ 2\times10^{-3}$ %
- Pu-content	below detection limit (3×10^{-4} mg Pu-0.006 %)

Table 3.4.1.1 U-Pu content of the residue

Fig. 3.4.8 summarizes the full sample preparation for programme part 1.3. Only aliquots of AS I, AS II, AS IV and AS VI were distributed to the participants (in dried form). The others were kept as a reserve.

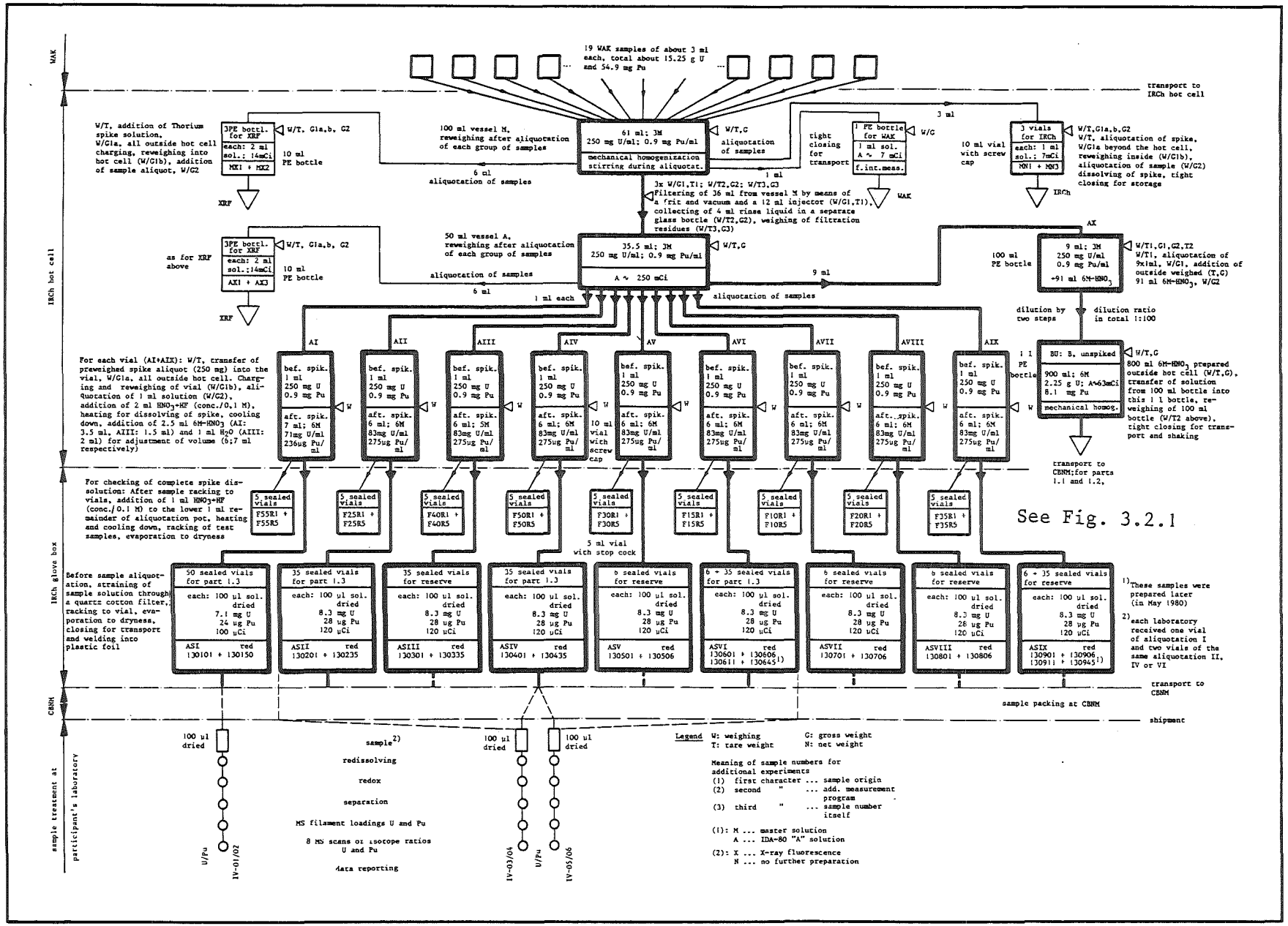


Fig. 3.4.8 Sample preparation for part 1.3

3.5 Preparation of the fission product free reference solution RU

The preparation of fission product free solutions was achieved weeks before the in situ-spiking and the preparation of the BU and BS solution.

The reference solution was prepared from an existing parent solution (synthetic Input solution CBNM-IRM 045-1). See approximate characteristics in Table 3.5.1.

Isotopes	atom %	Isotopes	atom %
^{235}U	1.2	^{238}Pu	0.1
^{238}U	98.8	^{239}Pu	76.5
		^{240}Pu	19.8
		^{241}Pu	3.1
		^{242}Pu	0.5
Concentration:		250 mg U/g solution 2 mg Pu/g solution	

Table 3.5.1 Characteristics of the reference solution

The solution had been made up from UO_2 powder (CBNM Lot Nr. BC 00728) and from PuO_2 (CBNM Lot Nr. 519). The required concentration for the RU-solution was 2 mg U/ml and 10 μg Pu/ml. After dissolution of semi-quantitative amounts of UO_2 and PuO_2, the solution was diluted to 1000 ml with 6M HNO_3 and homogenized by means of a magnetic stirrer.

3.5.1 Aliquoting of the RU-solution, programme parts 2.1/2.3

After six days 50 ampoules were filled successively with 10 ml "RU" solution for programme parts 2.1/2.3. The ampoules were sealed immediately and labelled as RU 213001 through RU 213050 (see Fig. 3.5.2). Additionally 200 ml of the RU solution were taken for the preparation of the RS solution, progr. part 2.2 (see Photogr. 3.5.1.1).

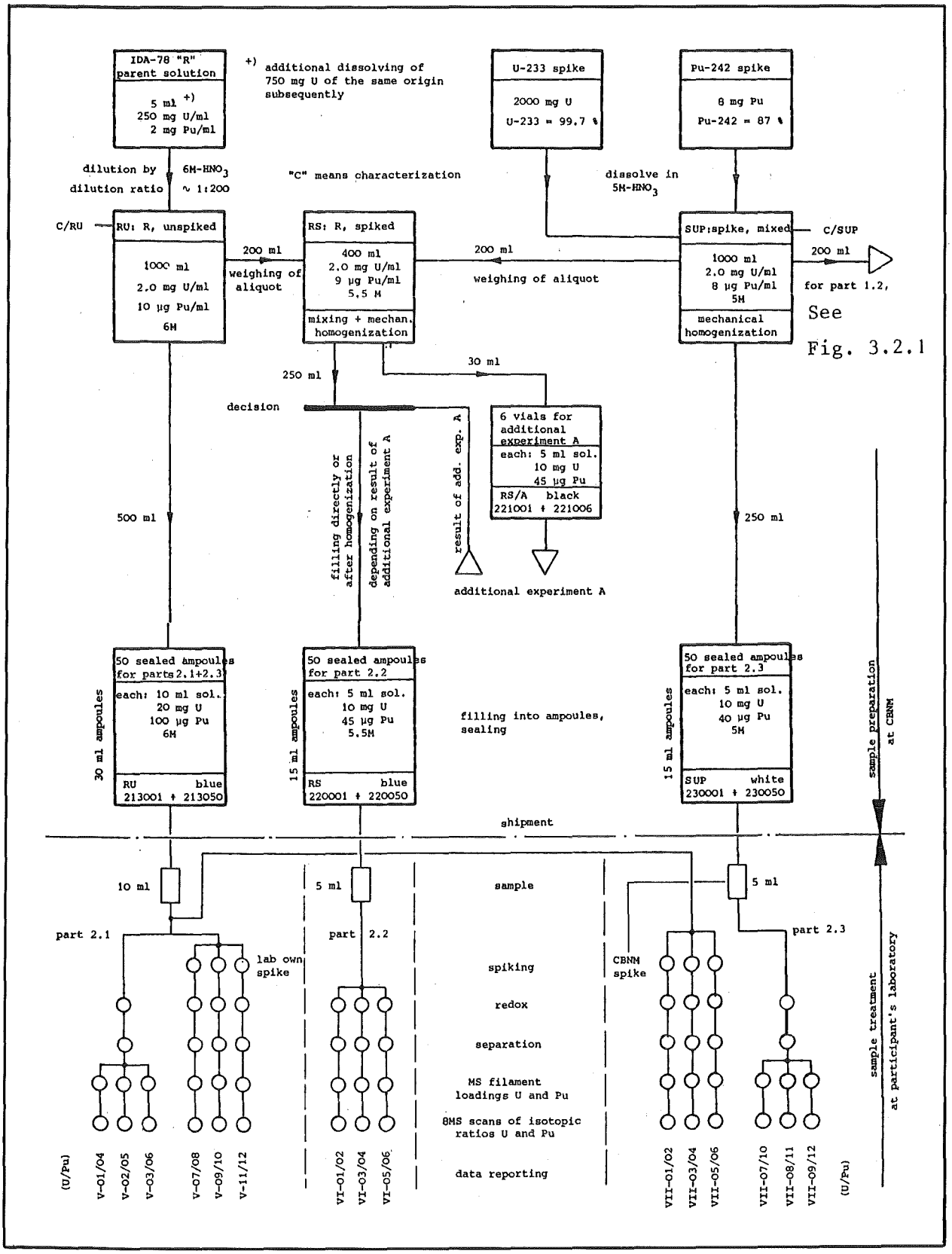
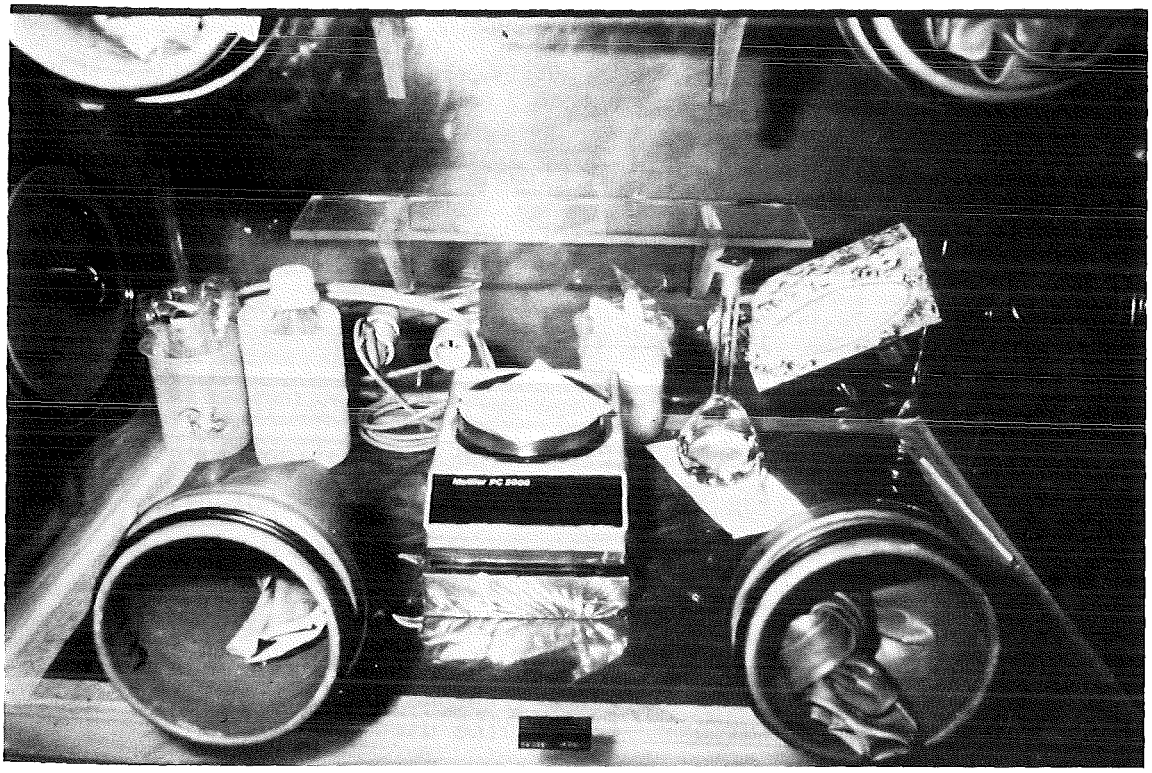


Fig. 3.5.2 Sample preparation for part 2



Photogr. 3.5.1.1 Top loader balance for the preparation of the RS and BS solution

3.5.2 Spiked fission product free reference solution RS, programme part 2.2

The RS solution was prepared by mixing weighed fractions of chemically and isotopically well-defined plutonium and uranium solutions. The required concentrations were 2 mg U/ml and 9 μg Pu/ml in 5.5M HNO_3 solution. The achieved isotope ratios for $^{233}U/^{238}U$ and $^{242}Pu/^{239}Pu$ were about 1.

After aliquoting the required samples for part 2.3, the weighed fraction of 200 ml SUP-solution, (see Section 5.2) was weighed again. Four times 50 ml of the RU-solution were transferred to the SUP solution. The exact mass was calculated by difference weighings (see Table 3.5.2.1).

200 ml SUP-solution	232.91 $\pm$ 0.01 g
200 ml RU-solution	232.83 $\pm$ 0.01 g

Table 3.5.2.1 Fraction SUP and RU solutions

The solution was homogenized by magnetic stirring. The homogeneity was tested on six independent samples by measuring the isotope ratios $^{233}U/^{238}U$ and $^{242}Pu/^{239}Pu$ respectively. The homogeneity was found to be better than $\pm$ 0.1 %.

50 ampoules with 5 ml solution were sampled and sealed immediately. The ampoules were labelled as RS 220001 through RS 220050 (see Fig. 3.5.2).

4. Sampling for the additional experiment

The 19 small bottles of input solution were collected in one vessel and homogenized (see Fig. 4.1). Because of the high γ-radiation all operations on the undiluted input solution had to be carried out in a hot-cell (KfK/IRCh see Section 6.1). Three aliquots of 1 ml were then taken, weighed and spiked with CBNM metal spike (see Table 4.1).

Sample Identification	Mass of 1 ml Input Solution	Identification of Spike used	Mass of Spike
MN 1	$1.4468 \pm 5 \cdot 10^{-4}$ g	114	$261.84 \pm 2 \cdot 10^{-2}$ mg
MN 2	$1.4188 \pm 5 \cdot 10^{-4}$ g	111	$242.82 \pm 2 \cdot 10^{-2}$ mg
MN 3	$1.4239 \pm 5 \cdot 10^{-4}$ g	113	$246.57 \pm 2 \cdot 10^{-2}$ mg

Table 4.1 In situ spiking of unfiltered input solution

The metallic spikes were dissolved in the undiluted input samples and spiked samples were measured at the IRCh Mass Spectrometry, see Table 4.2.

Sample Identification	Pu-Element concentration		U-Element concentration	
	mg/g	MEAN VALUE	mg/g	MEAN VALUE
MN 1	0.5935	0.5891	167.88	167.89
MN 2	0.5842		167.92	
MN 3	0.5897		167.89	

Table 4.2 Element concentration of the unfiltered undiluted input solution (Mass Spectrometry)

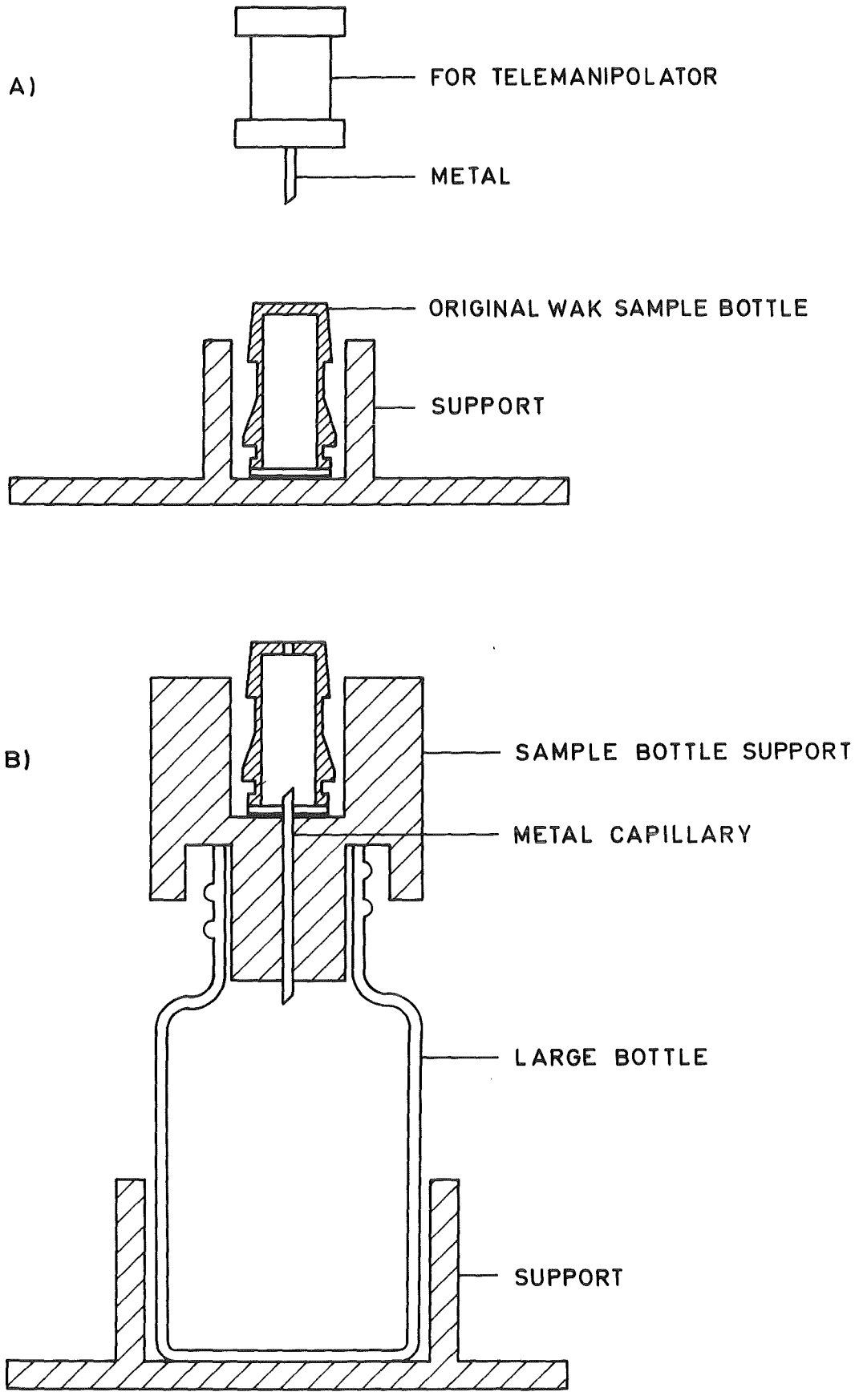


Fig. 4.1 Device for transferring the input solution into the collection vessel

Next, 3 samples of 2 ml of the unfiltered input solution were sampled and weighed for X-ray fluorescence. The solution was mixed with a well known amount of Thorium reference solution (see Table 4.3). Measurements of Uranium and Plutonium were carried out by IRCh-KfK Karlsruhe (see Table 4.4).

Sample Identification	Mass of 0.5 ml Th-solution	Mass of 2 ml input solution
MX 1	0.6071 g	$2.8717 \pm 5 \cdot 10^{-4}$ g
MX 2	0.6066 g	$2.8703 \pm 5 \cdot 10^{-4}$ g
MX 3	0.6060 g	$2.8705 \pm 5 \cdot 10^{-4}$ g

Table 4.3 X-ray fluorescence

Sample Identification	Pu-Element concentration		U-Element concentration	
	mg/g	MEAN VALUE	mg/g	MEAN VALUE
MX 1	0.583	0.582	168.3	167.6
MX 2	0.581	accuracy:	167.4	accuracy
MX 3	0.582	$\pm$ 1.3 %	167.1	$\pm$ 0.5 %

Table 4.4 Element concentration of the unfiltered undiluted input solution (X-ray fluorescence)

After following the procedure described in Section 2, 3 samples were taken, measured and compared with the unfiltered input solution (see Table 4.5).

Sample Identification	Mass of Thorium solution	Mass of 2 ml Input solution
AX 1	0.6063	$2.8740 \pm 5 \cdot 10^{-4}$ g
AX 2	0.6070	$2.8747 \pm 5 \cdot 10^{-4}$ g
AX 3	0.6080	$2.8649 \pm 5 \cdot 10^{-4}$ g

Table 4.5 Sampling amounts of the filtered undiluted input solution (X-ray fluorescence)

Results see Table 4.6

Sample Identification	Pu-Element concentration		U-Element concentration	
	mg/g	Mean Value	mg/g	Mean Value
AX 1	0.588	0.587	170.1	170.3
AX 2	0.594	accuracy	170.2	accuracy
AX 3	0.580	± 1.3 %	170.4	± 0.5 %

Table 4.6 Element concentration of the filtered undiluted input solution (X-ray fluorescence)

5. Spike materials used in the programme

5.1 Metal spike

For part 1.3 which required an "in situ spiking" of the undiluted input solution "A",a metal alloy spike was prepared with enriched ^{235}U and ^{242}Pu (see section 8).

The Uranium metal ($^{235}U/U$ = 0.92) belonged to CBNM stock (CBNM Lot 125AG and 125PE). The Plutonium metal ($^{242}Pu/Pu$ = 0.87) was purchased from the CEN, Fontenay-aux-Roses (CBNM Lot 573).

The U/Pu alloy (CBNM Lot 576) was characterized both by quantitative preparation and IDMS at CBNM and delivered in sealed glass-ampoules containing 250 mg each and having the approximate composition of U/0.3 % Pu (see chapter II, Sections 5.3 and 5.4).

5.2 U-Pu mixed spike solution SUP

For parts 1.2 and 2.2, a mixed spike solution of ^{233}U and ^{242}Pu was prepared from ^{233}U ($^{233}U/U$ = 0.997 - CBNM Lot 559) and ^{242}Pu ($^{242}Pu/Pu$ = 0.875 - CBNM Lot No. 577). The required concentration was 2 mg U/ml and 8 μg Pu/ml. The solution was prepared semi-quantitatively by weighing the required amounts of Plutonium and Uranium. After dissolving the materials separately the mixed spike solution was achieved by blending. Details are as follows:
9.3 mg ± 0.5 mg PuO_2 were weighed and dissolved in a 100 ml Erlenmeyer flask with 5 ml concentrated nitric acid. 2.412 ± 0.001 g U_3O_8 were weighed and dissolved in the same way. The Uranium and Plutonium solution were transferred into a 1 l glass-flask with screw cap and diluted to 100 ml 5M HNO_3.

After homogenization with a magnetic stirrer, 50 ampoules were filled with 5 ml solution and sealed immediately (see Fig. 3.5.2). The ampoules were labelled: SUP 230001 through SUP 230050 (part 2.3).

A further 200 ml of SUP-solution was aliquoted and weighed in a glass bottle with screw-cap for the spiking of the RU-solution (see Section 3.5.2) and BU-solution (see Section 3.3).

The aliquotation of the SUP-solution for these three programme parts was carried out simultaneously to guarantee comparable samples.

6. Laboratory Facilities

6.1 Hot-Cell (KfK IRCh)

A hot-cell with a 100 mm lead protection wall was used for the handling of the undiluted input solution, for the preparation of the diluted input solution BU and for the sampling of the X-ray fluorescence and mass spectrometric measurement samples. The transparent area consisted of a 20 cm thick lead glass shield. This hot-cell was built in 1970 by Wälischmiller, Markdorf and equipped with 4 gas tight manipulators A 202 of the same manufacture.

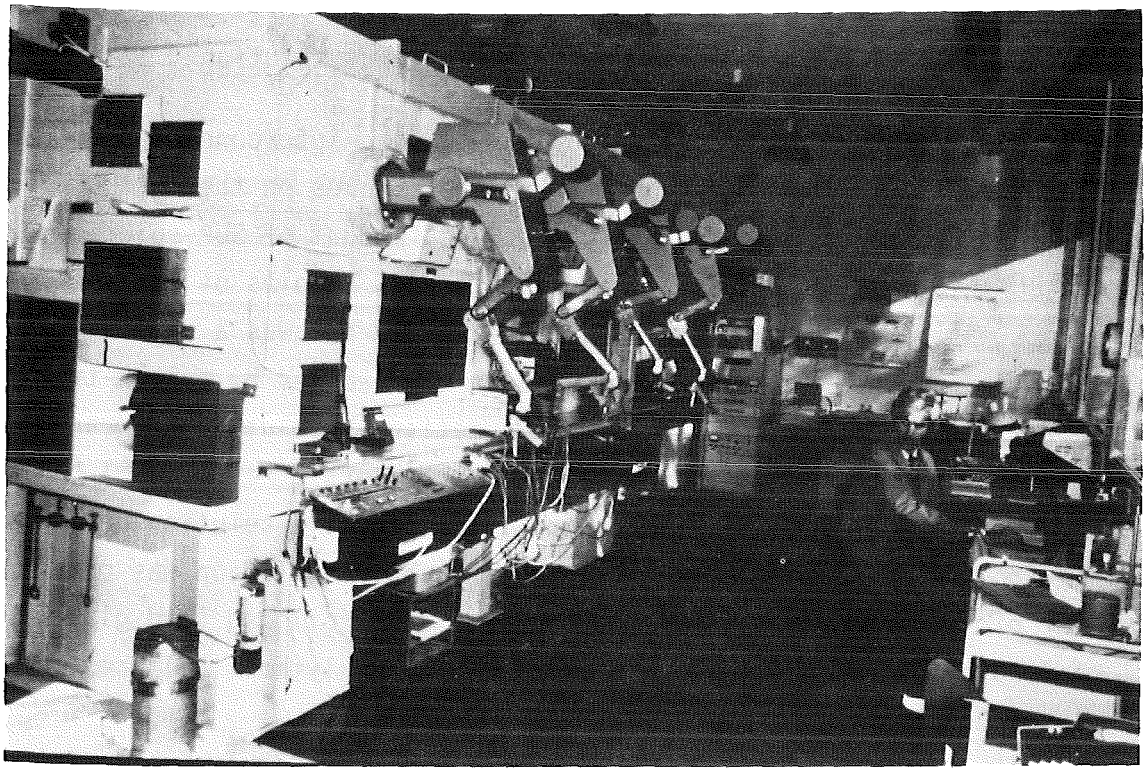
The equipment needed for the IDA-80 work consisted of an electronic balance Mettler H20E, a filtration device and an adjustable hot plate. Before work, the hot-cell was decontaminated, the work surface was covered with plastic foil and the top of the manipulators were changed (see Photograph 6.1.1).

Metallic α-work glove boxes

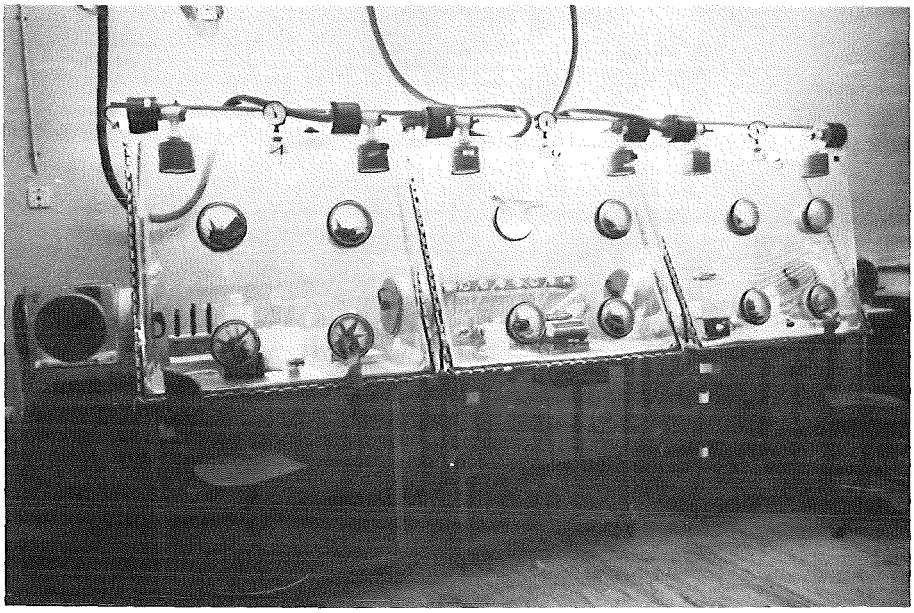
For the aliquotation of the AS samples two stainless steel glove boxes built by Draht and Schrader, Buchhausen, Type B3 were used. In addition to a wall of 10 mm stainless steel, a 5 mm lead foil protected the operators against the radiation of the IDA samples. The transparent area consisted of a 10-12 mm thick lead glass shield.

6.2 Glove boxes for the treatment of the reference solution

Three new PVC glove boxes (Type VT, Jahan Paris), were interconnected for the handling of the solutions of part 2 (see Photograph 6.2.1). One side of the glove box train was arranged for the preparation of the RU- and RS-solution. The other side was installed for the preparation of the mixed spike solution "SUP". These glove boxes were installed with an entry lock and equipped with a magnetic stirrer, ampoule holder and transfer pipettes (see Photograph 7.1). The middle glove box was accessible from two sides and equipped with a Mettler Electronic Top Loader Balance.



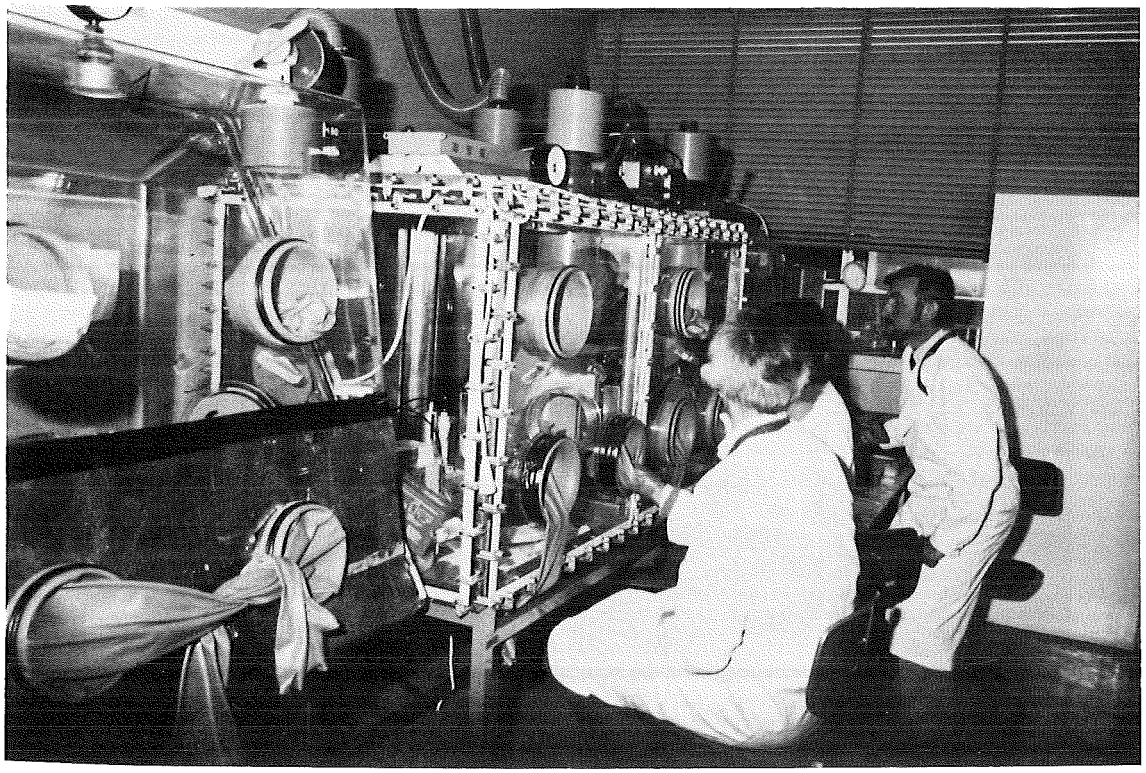
Photogr. 6.1.1 Hot-cell laboratory at KfK-IRCh



Photogr. 6.2.1 Glove box for part 2

6.3 Glove boxes for the treatment of the diluted input solution

Three new glove boxes (Type VT, Jahan Paris) were interconnected for the handling of the diluted feed solution B. The glove boxes on the left and right side were PVC boxes for handling the weighed amount of SUP-solution and the spiked BU-solution (BS), and to evaporate the 129 samples of part 1.12. An entry lock was installed on each side. The middle box was a standard type III metal frame box divided into 2 smaller glove-boxes by a leak-tight partition: one part for the handling of the γ-active BU-solution by means of a piston burette from Methrom, the other part for a Mettler analytical balance for the aliquotation of part 1.12. To handle the γ-active solution one glove box was equipped with a lead castle in which the active solution was placed. Part of the other glove boxes were shielded with lead foil and lead glass. A wall of lead bricks was installed in front of the analytical balance (see Photogr. 6.3.1).

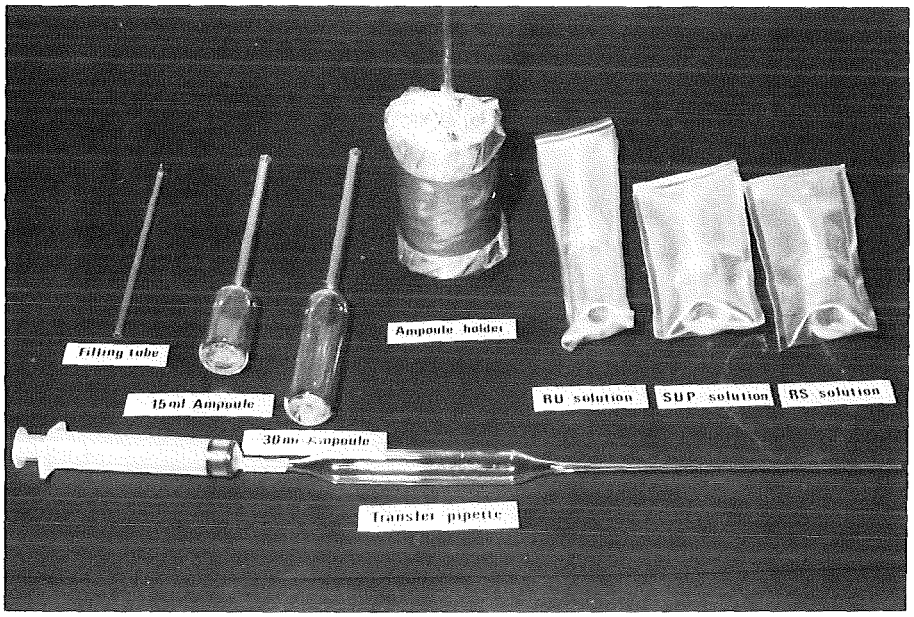


Photogr. 6.3.1 Glove boxes for the treatment of the diluted input solution

7. Sampling procedure

The glass ampoules used for the samples of parts 1.11, 1.2, 2.1, 2.2 and 2.3 have been manufactured at the CBNM glass workshop. To facilitate opening of the vials, these were filled to 1/3 only (e.g. 5 ml samples in 15 ml ampoules and 10 ml samples in 30 ml ampoules). This also allowed safe filling.

Every effort was made during sampling to keep the ampoules clean at the outside and at the inside of the ampoule necks. To prevent inside contaminations a clean glass-tube with collar was used for each ampoule filling (see Photograph 7.1).



Photogr. 7.1 Sampling tools

For parts 1.12 and 1.3 small glass vials with a plastic stopper were used. Dimension: diameter 15 mm, height: 50 mm. The inactive solutions for parts 2.1, 2.2, 2.3 and the active solution for part 1.2 were transferred with a syringe equipped with a tapered drawn-out glass tube. Solution of part 1.3 was taken by means of an Eppendorf pipette. For parts 1.11 and 1.12 the solution was taken with a Multidosimat E535 from Methrom equipped with a remote-controlled motor driven piston burette.

Each ampoule and vial, except those for part 1.3, was checked for contamination by the CBNM Health Physics service. Those for part 1.3 underwent checking at KfK. In no case was a contamination detected.

Upon termination of the sample preparation the glove boxes were checked on contamination by Health Physics staff. Not a single contamination could be detected.

8. Preparation of the U-Pu metal spike by high-frequency levitation melting

High-frequency levitation melting can yield accurately defined alloy samples. Its essential feature is the homogeneous mixing of compounds of known mass into a single sample essentially without loss and without contamination [1].

During the preparation the sample floats in a magnetic field without any direct contact with its surroundings. From the masses and purities of the components the concentration of each element in the sample can be calculated. Possible small losses are considered as the uncertainty of the method.

This technique was used for the preparation of 10 g amounts of a U-5 wt % Pu mother alloy and the successive dilution to a 10 g amount of U-0.3 wt % Pu.

Under the assumption that the losses during these preparations are only due to Pu, the composition of the spike alloy can be calculated as follows

$$U\text{-}(0.294 \pm 0.003)\ \text{wt \% Pu}.$$

This alloy was rolled, cut into pieces of about 250 mg mass and packed in glass ampoules under intert gas.

---

(1) J. Van Audenhove, J. Joyeux, Sample preparation by metallurgical methods, Nucl. Instr. Meth. 102 (1971) 409-415.

9. Weighings

All weighings including the revision of the used balances and the verification of the accuracy by test weighings were performed by the CBNM Metrology.

The weighings were done on sensitive balances using a set of calibrated weights. They were all of the type "substitution-weighing".

Corrections for buoyancy were applied using the following equation:

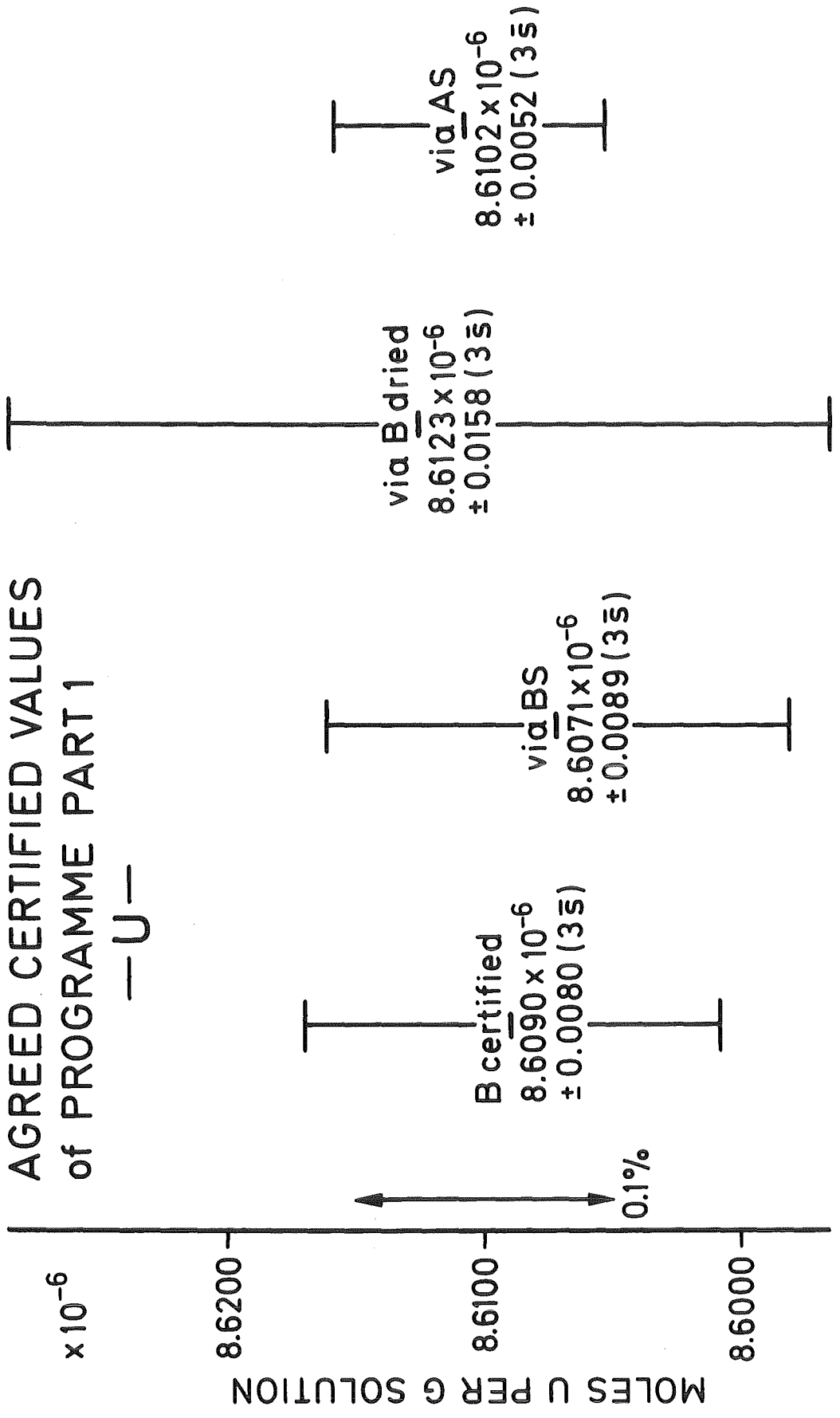
$$M_2 - M_1 = [W_2(1 - \frac{P_2}{d_w}) - W_1(1 - \frac{P_1}{d_w})] + V_1(P_2 - P_1) + v_2P_2$$

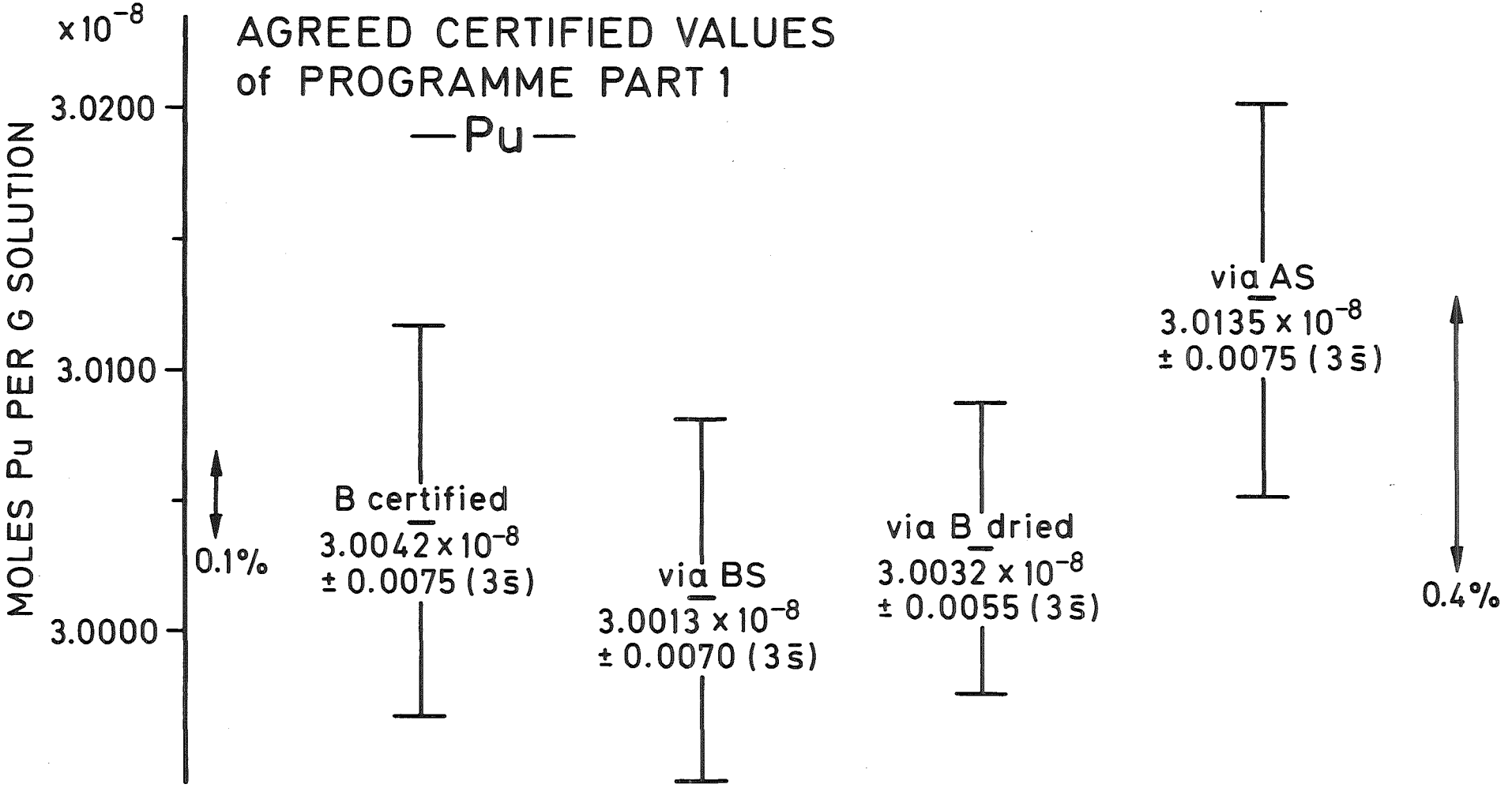
M_1 = mass of the flask
M_2 = mass of the solution + flask
W_1 = weight of the flask
W_2 = weight of the flask containing the solution
P_1 = density of the air during weighing of the flask
P_2 = density of the air during weighing of the flask + solution
V_1 = volume of the flask
v_2 = volume of the solution
d_w = density of the calibrated weights.

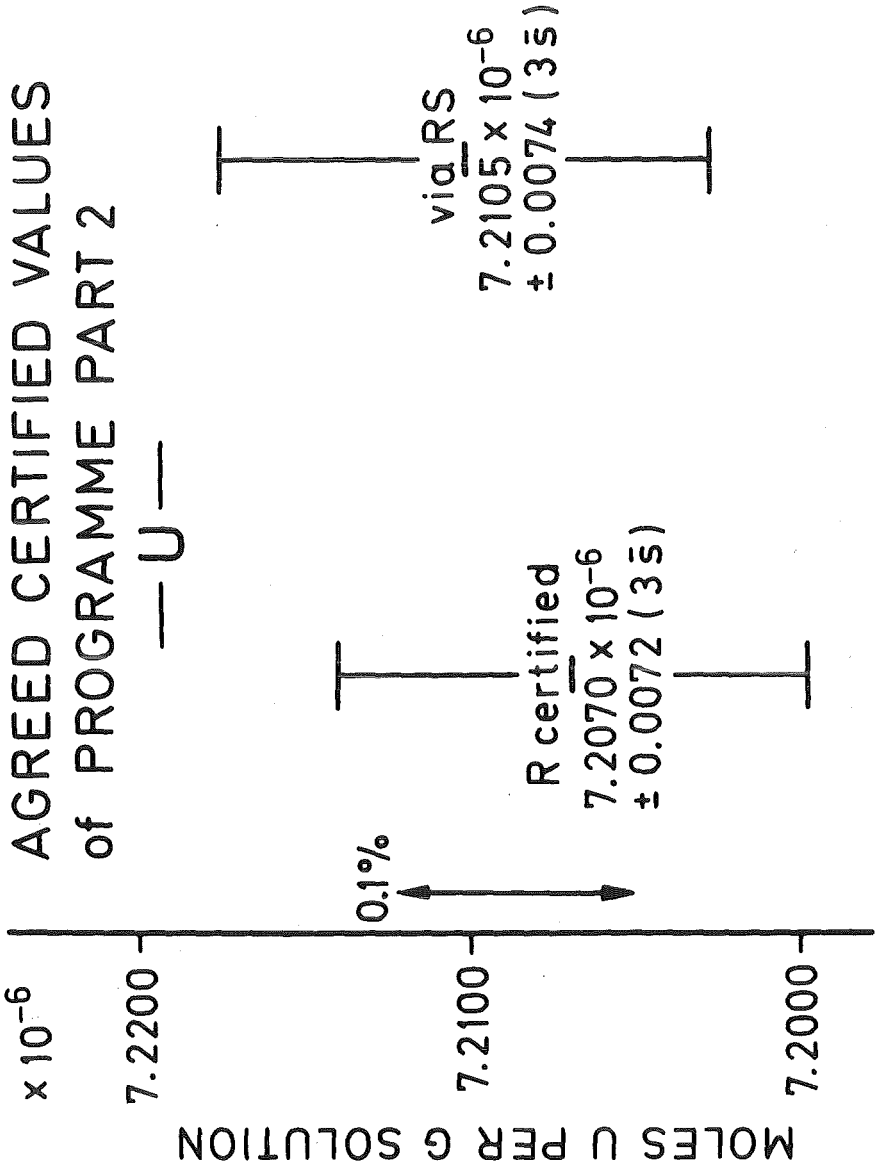
For the preparation of the solutions and samples different balances were used (see Table 9.1).

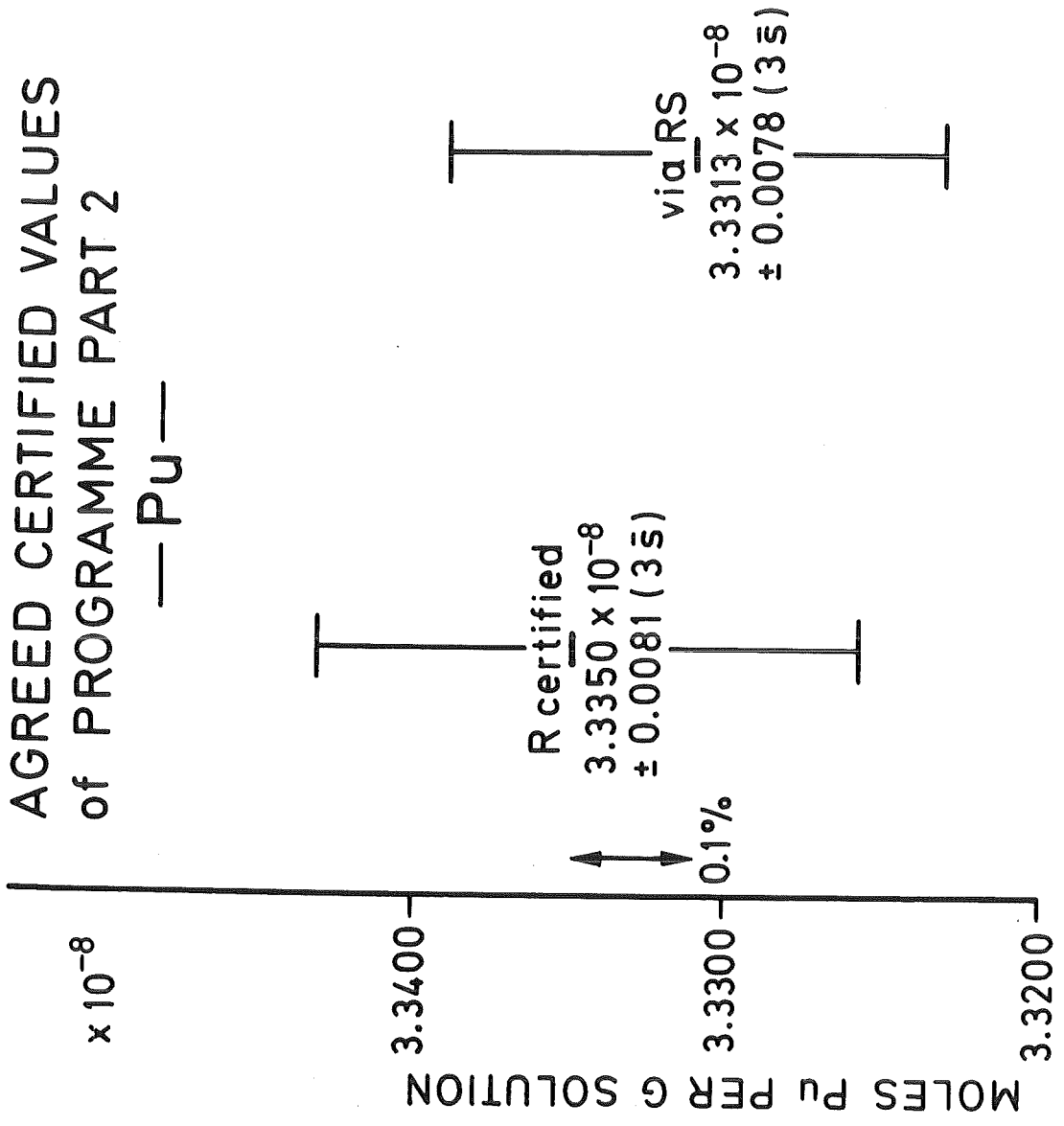
Type of Balance	weighing range	weighed solution	programme part
Top loader Mettler balance, Type PC 2000	2000 g	RU-solution RS-solution BS-solution	2.1 2.2 1.2
Mettler balance Type HL 52	mechanical 159.9 g electric 0.2 g	BU-dried	1.12
Top loader Mettler balance, Type PN 5	5000 g	BU-solution	1.1
Sartorius electronic balance, Type 2002MP1	160 g	Metallic spike verification	
Mettler electronic balance, Type H20E	160 g	AS-solution	1.3
Mettler M 5	20 g	Metallic spikes	

Table 9.1 Balances used in IDA-80 test sample preparation


AGREED CERTIFIED VALUES
of PROGRAMME PART 1
— U —
MOLES U PER G SOLUTION
x 10⁻⁶
8.6200
8.6100
8.6000
0.1%
B certified
8.6090 x 10⁻⁶
± 0.0080 (3s̄)
via B S
8.6071 x 10⁻⁶
± 0.0089 (3s̄)
via B dried
8.6123 x 10⁻⁶
± 0.0158 (3s̄)
via A S
8.6102 x 10⁻⁶
± 0.0052 (3s̄)


AGREED CERTIFIED VALUES
of PROGRAMME PART 1
—Pu—
x10⁻⁸
MOLES Pu PER G SOLUTION
3.0200
3.0100
3.0000
0.1%
B certified
3.0042 x 10⁻⁸
± 0.0075 (3s̄)
via BS
3.0013 x 10⁻⁸
± 0.0070 (3s̄)
via B dried
3.0032 x 10⁻⁸
± 0.0055 (3s̄)
via AS
3.0135 x 10⁻⁸
± 0.0075 (3s̄)
0.4%


AGREED CERTIFIED VALUES
of PROGRAMME PART 2
— U —
via RS
7.2105 x 10⁻⁶
± 0.0074 (3s̄)
R certified
7.2070 x 10⁻⁶
± 0.0072 (3s̄)
0.1%
x 10⁻⁶
7.2200
7.2100
7.2000
MOLES U PER G SOLUTION


AGREED CERTIFIED VALUES
of PROGRAMME PART 2
— Pu —
MOLES Pu PER G SOLUTION
x 10⁻⁸
3.3400
3.3300
3.3200
0.1%
R certified
3.3350 x 10⁻⁸
± 0.0081 (3s̄)
via RS
3.3313 x 10⁻⁸
± 0.0078 (3s̄)

Page

List of Figures

List of Tables

Page

List of Photographs

Chapter II: Characterization of the Test Samples

by

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1. Introduction

Putting accurate values with certified uncertainties on the test samples of an interlaboratory measurement evaluation programme which can serve as references (or "baseline") for the participating laboratories is a highly demanding task: it is expected that "true" values are contained somewhere in the quoted uncertainty range.

Furthermore, in order to be a real "reference" the uncertainty range thus defined should be smaller by a factor 3 - but preferably 5 to 10 - than the interlaboratory spread of the results.

Numerous measurements, reliable and experienced analysts and careful work are obvious components of such process. More important is the painstaking search for all possible systematic errors in the instrumentation and the measurement procedure. Once systematic errors have been identified, they can be corrected to provide a measurement that is essentially free of systematic errors (apparent at the given level of uncertainty). Thus only the uncertainty in the applied corrections affects the overall uncertainty range of the "baseline".

It has been attempted to achieve that in IDA-80 for all of the test samples,in particular for the two main solutions distributed as B and R test samples.

Because of

a) the scope of the programme: establish how well the nuclear community can determine U and Pu isotopes and elements in general and input samples in particular
b) the world-wide participation

CBNM has invited NBS Washington to join in the establishment of the "baseline" and its uncertainty, so that independent attempts of two Reference Material laboratories would yield a most reliable value to serve the participants in the programme.

2. Review of the measurement programme of the characterization laboratories CBNM Geel and NBS Washington

From all test materials used in the programme (see lay-out in Table 2.1), samples have been taken according to schemes which ensure proper representativeness and using sampling plans which - in a number of cases - would have revealed inhomogeneities if present.

These samples have been identified in Table 2.2 for convenience of the participants in the programme.

On these samples CBNM and NBS have been working fully independently. Only when measurements had been achieved were values compared. Within the measurement uncertainties no differences were observed relevant for the programme.

On the basis of

- careful evaluation of all corrections for systematic errors,
- traceability to reference materials (type, quality and length of the traceability route),
- study of 2nd order effects,

in a case-to-case discussion (August 1982), was one value agreed for each material. This value has been called "agreed certified value". This value is supported by both institutes to be "as close to the truth" as possible on the basis of all evidence and experience as available at both places. At that time it was given a preliminary conservative uncertainty range which was used in the IAEA-SM-260/33 paper presented in Wien in November 1982 and in the evaluation reports prepared for discussion at the participants' meeting in March 1984.

These values took 1.5 years of measurements to be established.

In a second round of discussions (1983), this time on the establishment of the uncertainties on the values, these were established as follows:

a) at each institute:
   3 times the "standard error" or "standard deviation of the mean" ($\bar{s}$)

b) on the "agreed certified values":

$$\sqrt{\frac{(3\bar{s})^2\ \text{CBNM} + (3\bar{s})^2\ \text{NBS}}{2}}$$

These uncertainties are found in the tables hereafter and will be used in the final report drafted after the participants' meeting.

It may be useful to indicate that both the certified values and their uncertainties were established (at CBNM and NBS) totally independent from the evaluation work (at KfK Karlsruhe) and the participants' results.

A final remark is that the key values of the programme are those of the diluted B and R solutions. Consequently, all measurements in the different programme parts have been evaluated against these. They have been arrived at by the best, most accurate and shortest characterization routes possible e.g. via measurements on the B (part 1.11 BU) and on the R solution (part 2.1 RU) and not via measurements on the dried samples or/and via prespiked samples.

The consistency of element concentration determinations of programme part 1.11, 1.12, 1.3 and programme part 2.1 and 2.2 is given in appendices 1, 2, 3, 4 of Chapter I.

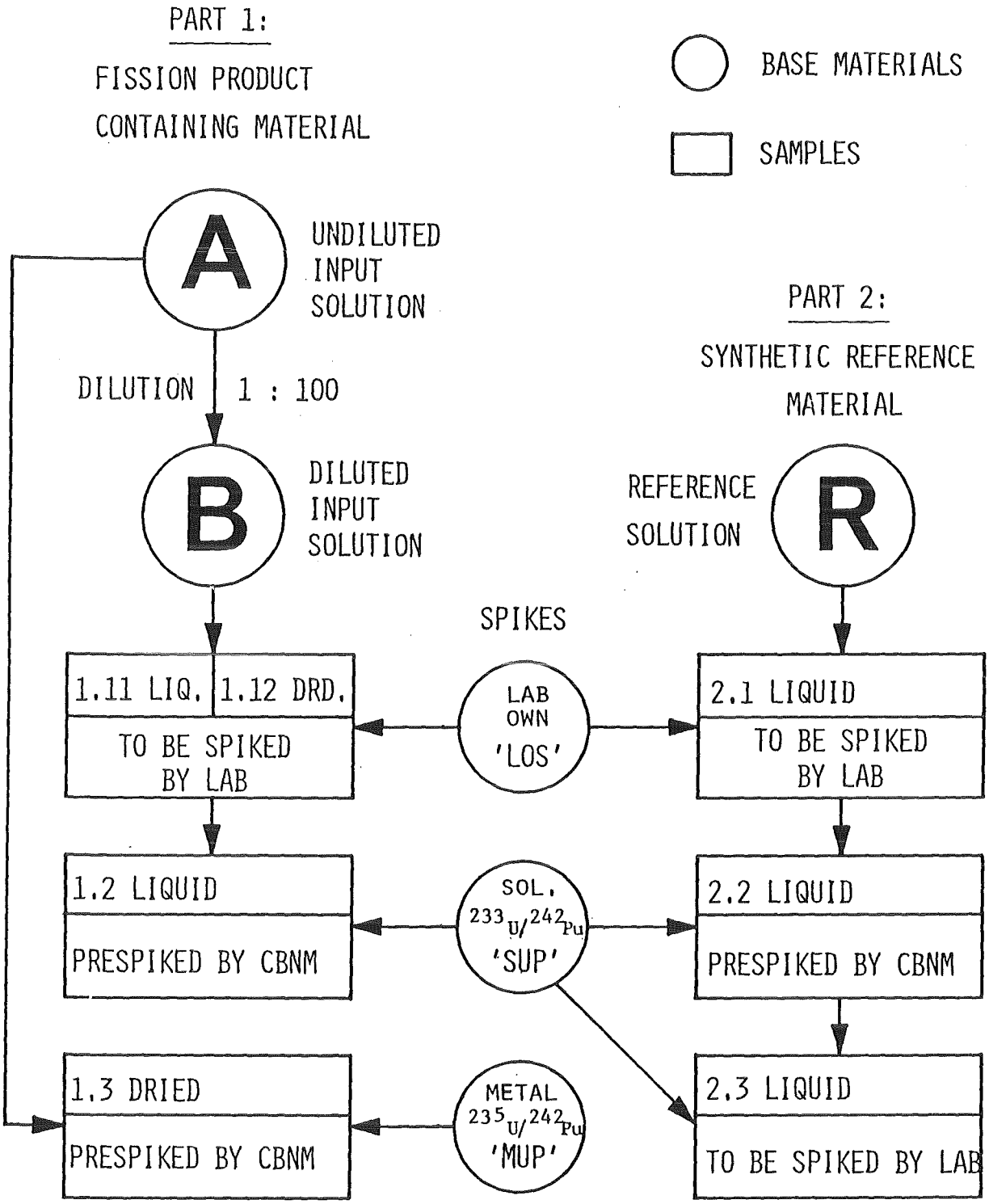

Table 2.1 General lay-out

Programme part	Code number of the prepared samples	Characterization at	
		C B N M	N B S
1.11 Diluted input solution "BU"	111001-111050	001 020 039	002 010 030
1.12 Diluted input solution BU-dried	I 112001-112043 II 112051-112093 III 112101-112143	010 020 030 140 141 142	
1.2 Spiked diluted input solution "BS"	121001-121006 120001-120050	001 002 003 004 005 006	001 010 020 030 040 050
1.3 Spiked input solution "AS"	I 130101-130150	101 110 120 130 140 150	111 121 131
	II 130201-130235	201 210 220 230 235	211 221 231
	III 130301-130335	301 310 320 330 335	311 321 331
	IV 130401-130435	401 410 420 430 435	411 421 431
	V 130501-130535	501 503 505	502 504 506
	VI 130601-130635	601 603 605 611 640	602 604 606
2.1 Reference Solution "RU"	213001-213050	001 020 040	010 030 050
2.2 Spiked Reference Solution "RU"	221001-221006 220001-220050	001 002 003 004 005 006	001 010 020 030 040 050
2.3 Mixed Spiked Solution "SUP"	230001-230050	001 050	002 025 049

Table 2.2 Samples of the test materials characterized by CBNM and NBS

3. Characterization of IDA-80 test samples at CBNM

3.1 Mass spectrometric equipment

The isotopic measurements were performed on a 30 cm - 90° magnetic sector Teledyne type mass spectrometer, equipped with a triple filament rhenium ion source. Ion collection was done on a deep bucket Faraday cage collector. Prior to analysis the filaments were outgassed at about 2000°C.

Purified Uranium and Plutonium solutions containing 10 mg U/ml and 1 mg Pu/ml were used for the filament loading. One μl of Uranium or Plutonium solution was placed on each of the two sample filaments and dried before putting them into the sample insert.

During the measurements, a liquid nitrogen cold trap was used to get an extra clean vacuum in the source region.

Data collection was done by peak switching (magnetic field), vibrating reed amplifier, voltage to frequency convertion and HP 9830 calculator, or manual scanning of the magnetic field, vibrating reed amplifier and strip-chart recorder equipped with an expanded scale device. All results were corrected for bias using the NBS isotopic SRM's for Uranium and Plutonium.

3.2 Chemical sample purification prior to isotopic measurements

Both separation and purification of U and Pu fractions were performed using an anion exchange procedure.

a) Separation of U-Pu

The following procedure was applied for U amounts up to 25 mg and Pu amounts up to 500 μg.

First the U/Pu sample was evaporated to dryness and dissolved in 200 μl 2M HNO_3.

To achieve valency homogeneization in the Pu^{IV} state, one drop of 1.25 M $FeCl_2$ was added to convert all Pu to the III valent state. Subsequently, 100 μl 1 M $NH_2OH.HCl$ and 100 μl 1 M $NaNO_2$ were added. This mixture has a slightly oxidizing effect and also buffers the solution to prevent oxidation to Pu^{V} and Pu^{VI} state. Finally, 430 μl concentrated nitric acid was added to obtain Pu^{IV} 8 M HNO_3 solution.

This solution was brought on an anion exchange column with an inner diameter of 0.4 cm and loaded with Dowex resin 1 X 4 100 - 200 mesh to about 10 cm height. The resin was preconditioned with distilled water and with 8 M HNO_3. After loading the sample onto the column, 4 ml of 8 M HNO_3 were added. The Pu fraction remains on the column, the U fraction descends slowly. Another 5 ml of 8 M HNO_3 eluted the U fraction from the column. This fraction was collected, evaporated to dryness and dissolved in 1 M HNO_3 for MS measurement.
The column was then washed with ~ 20 ml 8 M HNO_3 to elute any remaining U. It is very difficult to elute all the U in one separation. It is therefore necessary to perform a purification step of the Pu fraction. Consequently after washing with ~ 20 ml 8 M HNO_3, the Pu fraction was eluted with 0.35 M HNO_3, evaporated to dryness and prepared for the next purification step.

b) Second purification step of Pu

A procedure analogous to the one described above, was performed on solutions containing Pu only.
The only difference with the procedure described above, is that the column used for the Pu-purification was shorter (5 cm anion exchange resin column). After purification and evaporation the Pu fraction was dissolved in 1 M HNO_3 and collected for MS measurement.

3.3 Aliquotation and preparation of the samples for the characterization measurements

a. RU and BU samples were received as solutions, three ampoules of each (see Table 2.2). Weighed aliquots of the samples were transferred to glass vials and all samples were spiked with weighed aliquots of CBNM-spike.
The blended solutions were evaporated and dissolved in 200 μl 2 M HNO_3 for the purification steps.

b. BU-dried samples, see Chapter I, section 3.2.2.

c. RS and BS samples were received as solutions (six glass vials of each). The sampling was done directly from the bulk spiked solution. The solutions were evaporated to dryness and dissolved in 200 μl 2 M HNO_3 for the purification steps.

d. The SUP material was received as solution in two sealed glass ampoules (see Table 2.2). Three weighed aliquots of each ampoule were spiked with NBS SRM 960 and SRM 944. After homogenization the solution was evaporated to dryness and dissolved in 200 μl 2 M HNO_3 for the purification steps.

e. The metal spike was received in sealed glass ampoules. Weighed fractions of either NBS SRM 950a or SRM 960 (for U) and SRM 944 (for Pu) were added to the metal spike. The metal spike was dissolved in 7 M HNO_3/0.1 M HF and evaporated to dryness. The residue was again dissolved in 200 μl 2 M HNO_3 for the purification steps.

f. Six independently in situ spiked solutions were received as evaporated samples in 27 glass vials (see Table 2.2). The residues were dissolved in 200 μl 2 M HNO_3 for the purification steps.

g. Measurements on NBS SRM U-500 and NBS SRM-947 were performed to provide isotope fractionation corrections.

4. Characterization of IDA-80 test samples at NBS

4.1 Mass Spectrometric equipment

Plutonium

Plutonium isotopic measurements were made on a 12", 90° magnetic sector mass spectrometer of NBS design (Ref. A), equipped with a pulse-counting detector. The NBS instrument possesses a thin lens ion source and utilizes magnetic field peak switching for isotope ratio measurement. The instrument is interfaced to a programmable calculator which controls the magnetic field and data acquisition and analysis.

The mass spectrometric sample loading procedure consisted of the electroplating of nominally 2 ng of Pu from solution directly onto the sample filament. A heating pattern that culminated in a temperature of 1350°C produced a total Pu^+ ion beam of 100,000 ions per second that gradually decayed in the course of the 30 minute data acquisition period.

The correction factor for isotopic fractionation was determined from analyses of SRM 947. Multiple determinations were made throughout the analytical programme to guarantee control of the measurement process and to provide an indication of measurement precision. The correction factor determined from the average of these measurements was applied to the experimental ratios. The correction factor for $^{240}Pu/^{239}Pu$, 1.00195 $\pm$ 0.00070 (1s, 28 determinations), agreed with previous measurements by the same procedure.

Uranium

Uranium isotopic measurements were made on a mass spectrometer identical to the instrument used for the plutonium isotopic measurements, except that it was equipped with a deep bucket Faraday cage collector.

Approximately 2 μg of uranium was loaded onto each of two sample filaments of a rhenium triple filament ion source. The samples, in a solution of 0,8 M nitric acid, were dried on the filaments as the uranyl nitrate dihydrate before insertion into the mass spectrometer. Before loading of the samples, the filaments were outgassed in a vacuum and under a potential field at approximately 2200°C for 30 minutes.

A uranium ion current at the collector of 6 x 10^{-11}A was obtained for the isotopic measurements and was sustained at a stable signal intensity throughout the measurement time. All data were corrected for isotopic fractionation by comparison with SRM U-500 run under identical conditions. The measurements on the NBS SRM U-500 $^{235}U/^{238}U$ ratio yielded a value of 1.0025 + 0.00013 (1s, 9 determinations). The certified value for SRM U-500, $^{235}U/^{238}U$ = 0.9997. Consequently the correction factor is 0.9976 and agreed with previous measurements by the same procedure.

4.2 Chemical sample purification prior to isotopic measurements

Both separation and purification of U and Pu fractions were performed using an anion exchange procedure.

(a) Separation of U-Pu

Approximately 0.5 ml of 5 M HNO_3 and 0.01 M HF solution and one drop of 30 % H_2O_2 were added to each U-Pu sample. The solution was evaporated to 1-2 drops and diluted with 5 drops of 8 M HNO_3. (One additional sample from each group was oxidized with $HClO_4$ to provide a check on the equilibration of sample and spike). The sample solution was added to an anion exchange column with an inner diameter of 0.5 cm and loaded with AG1x8, 100-200 mesh, resin to 3 cm height. [The column was cleaned with 8 M HNO_3 and a 0.3 M HNO_3 + 0.15 % H_2O_2 solution and preconditioned with 8 M HNO_3]. After loading the sample onto the column, the sample vial was rinsed with a total of 20 drops of 8 M HNO_3 and the U was then eluted with 165 drops of 4 M HNO_3. An additional 135 drops of 4 M HNO_3 was added to the column and discarded. The Pu was eluted with 150 drops of a 0.3 M HNO_3 + 0.15 % H_2O_2 solution.

(b) Second Purification of Pu

The Pu fraction was evaporated to 1-2 drops of solution, diluted with 5 drops of 4 M HNO_3, and transferred to a column with an inner diameter of 0.5 cm and loaded with AG1x4, 100-200 mesh, resin to a height of 1.3 cm. [The column was cleaned with 8 M HNO_3 and a 0.3 M HNO_3 + 0.15 % H_2O_2 solution and preconditioned with 8 M HNO_3]. Impurities were eluted with 200 drops of 4 M HNO_3 and the Pu was eluted with 100 drops of a 0.3 M HNO_3 + 0.15 % H_2O_2 solution. The Pu fraction was evaporated to 1-2 drops, 8 drops of 8 M HCl were added, the solution was evaporated to 1-2 drops, and the 8 M HCl addition and evaporation was repeated. The Pu fraction was then diluted with 0.1 M HCl to give a solution containing 200 ng Pu/g.

(c) Second Purification of U

The uranium fraction was evaporated to dryness, a few drops of 8 M HCl were added and the solution was again evaporated to dryness. The U was dissolved in 5 drops of 8 M HCl and transferred to a column with an inner diameter of 0.5 cm and loaded with AG1x8, 100-200 mesh, resin to a height of 5 cm. [The column was cleaned with 8 M HNO_3 and 0.1 M HNO_3 and preconditioned with 8 M HCl]. Impurities were eluted with 100 drops of 8 M HCl and then with 45 drops of 8 M HNO_3. The uranium was eluted with 120 drops of 0.1 M HNO_3. The uranium fraction was evaporated to dryness and diluted with 0.3 M HNO_3 to give a solution containing 1 mg U/g.

4.3 Aliquotation and preparation of the samples for characterization measurement

(a) The RU and BU samples were received as solutions in three sealed ampoules of each. Weighed aliquots (1.5 - 2 g) of the samples were transferred to glass vials and two aliquots from each ampoule were spiked with weighed aliquots from three ampoules of SRM 993 (^{235}U spike) and from three bottles of SRM 996 (^{244}Pu spike). The SUP spike was used to spike two additional aliquots from each of the three RU samples. Isotopic composition samples were also taken from each ampoule. The spikes and samples were equilibrated by adding diluted HNO_3 and 0.01 M HF and 30 % H_2O_2 and heating. Subsamples containing approximately 200 μg of U and 0.7 μg of Pu were taken for the separation and purification steps.

(b) The RS and BS samples were received as solutions in six sealed ampoules of each. Aliquots containing approximately 200 μg of U and 0.7 μg of Pu were taken for the separation and purification steps.

(c) The SUP spike was received as a solution in three sealed ampoules. In addition to the aliquots taken for step 4.3.a, one aliquot from each ampoule was spiked with SRM's 993 and 996 and treated as the spiked samples in 4.3.(a). Aliquots from each sample were also taken for isotopic composition.

(d) The AS samples were received as dried residues in glass vials. The samples were dissolved by adding approximately 4 g of 5 M HNO_3 + 0.01 M HF solution and warming. An aliquot containing approximately 200 μg of U and 0.7 μg of Pu was taken from each sample for the separation and purification steps.

(e) Measurements on NBS SRM U-500 and NBS SRM-S47 were performed to provide isotopic fractionation corrections to the IDA-80 data.

Reference

A) W.R. Shields, editor, NBS, Technical Note 546 (1970).

5. Characterization of the spike material used in the IDA-80 programme

5.1 Agreed certified values for the SUP mixed spike solution - Uranium

	Results	Uncertainty expressed as 3 $\bar{s}$
Atom Ratios		
$^{234}U/^{233}U$	0.002368	± 0.000024
$^{235}U/^{233}U$	0.000127	± 0.000007
$^{238}U/^{233}U$	0.000496	± 0.000012
Atom %		
^{233}U	99.7017	± 0.0030
^{234}U	0.2361	± 0.0025
^{235}U	0.0127	± 0.0007
^{238}U	0.0495	± 0.0012
Weight %		
^{233}U	99.6996	± 0.0030
^{234}U	0.2371	± 0.0025
^{235}U	0.0128	± 0.0007
^{238}U	0.0505	± 0.0012
Concentration		
atoms		
U/g sol.	4.5426×10^{18}	
^{233}U/g sol.	4.5291×10^{18}	
^{235}U/g sol.	5.7692×10^{14}	
moles		
U/g sol.	7.5433×10^{-6}	$\pm 0.0062 \times 10^{-6}$
^{233}U/g sol.	7.5208×10^{-6}	
^{235}U/g sol.	9.5800×10^{-10}	
gram		
U/g sol.	1.7579×10^{-3}	
^{233}U/g sol.	1.7527×10^{-3}	
^{235}U/g sol.	2.2517×10^{-7}	

5.2 Agreed certified values for the SUP mixed spike solution - Plutonium

Pu reference date 1980.02.09

	Results	Uncertainty expressed as 3 $\bar{s}$
Atom Ratios		
$^{238}Pu/^{242}Pu$	0.01142	± 0.00003
$^{239}Pu/^{242}Pu$	0.00298	± 0.00003
$^{240}Pu/^{242}Pu$	0.09798	± 0.00012
$^{241}Pu/^{242}Pu$	0.02497	± 0.00003
$^{244}Pu/^{242}Pu$	0.00022	± 0.00001
Atom %		
^{238}Pu	1.0039	± 0.0028
^{239}Pu	0.2620	± 0.0025
^{240}Pu	8.6131	± 0.0098
^{241}Pu	2.1950	± 0.0023
^{242}Pu	87.9067	± 0.0127
^{244}Pu	0.0193	± 0.0008
Weight %		
^{238}Pu	0.9883	± 0.0027
^{239}Pu	0.2590	± 0.0025
^{240}Pu	8.5503	± 0.0098
^{241}Pu	2.1881	± 0.0023
^{242}Pu	87.9948	± 0.0126
^{244}Pu	0.0195	± 0.0008
Concentration		
atoms		
Pu/g sol.	1.5626×10^{16}	
^{239}Pu/g sol.	4.0941×10^{13}	
^{241}Pu/g sol.	3.4299×10^{14}	
^{242}Pu/g sol.	1.3736×10^{16}	
moles		
Pu/g sol.	2.5948×10^{-8}	$\pm 0.0053 \times 10^{-8}$
^{239}Pu/g sol.	6.798×10^{-11}	
^{241}Pu/g sol.	5.696×10^{-10}	
^{242}Pu/g sol.	2.2810×10^{-8}	
gram		
Pu/g sol.	6.275×10^{-6}	
^{239}Pu/g sol.	1.6252×10^{-8}	
^{241}Pu/g sol.	1.3730×10^{-7}	
^{242}Pu/g sol.	5.521×10^{-6}	

5.3 Certified values for the metal mixed spike determined by CBNM - Uranium

	Results	Uncertainty expressed as 3 $\bar{s}$ Atom ratios n = 3 Element concentration n = 12
Atom Ratios		
$^{234}U/^{233}U$	0.12781	± 0.00039
$^{235}U/^{238}U$	15.154	± 0.014
$^{236}U/^{238}U$	0.03691	± 0.00014
Atom %		
^{234}U	0.7832	± 0.0031
^{235}U	92.8626	± 0.0059
^{236}U	0.2262	± 0.0008
^{238}U	6.1280	± 0.0053
Weight %		
^{234}U	0.7793	± 0.0032
^{235}U	92.7920	± 0.0050
^{236}U	0.2270	± 0.0008
^{238}U	6.2017	± 0.0053
Concentration		
atoms		
U/g spike	2.5410×10^{21}	
^{235}U/g spike	2.3596×10^{21}	± 0.0014×10^{21}
moles		
U/g spike	4.2194×10^{-3}	
^{235}U/g spike	3.9182×10^{-3}	
gram		
U/g spike	9.9250×10^{-1}	
^{235}U/g spike	9.2095×10^{-1}	

5.4 Certified values for the metal mixed spike determined by CBNM - Plutonium Pu reference date 1980.02.09

	Results	Uncertainty expressed as 3 $\bar{s}$ Atom ratios n = 3 Element concentration n = 10
Atom Ratios		
$^{238}Pu/^{242}Pu$	0.011637	± 0.000043
$^{239}Pu/^{242}Pu$	0.006392	± 0.000020
$^{240}Pu/^{242}Pu$	0.09811	± 0.00011
$^{241}Pu/^{242}Pu$	0.024919	± 0.000026
$^{244}Pu/^{242}Pu$	0.000215	± 0.000007
Atom %		
^{238}Pu	1.0197	± 0.0036
^{239}Pu	0.5601	± 0.0017
^{240}Pu	8.5969	± 0.0089
^{241}Pu	2.1834	± 0.0021
^{242}Pu	87.6211	± 0.0133
^{244}Pu	0.0188	± 0.0006
Weight %		
^{238}Pu	1.0038	± 0.0036
^{239}Pu	0.5537	± 0.0017
^{240}Pu	8.5346	± 0.0088
^{241}Pu	2.1767	± 0.0021
^{242}Pu	87.7122	± 0.0132
^{244}Pu	0.0190	± 0.0006
Concentration		
atoms		
$^{}Pu$/g spike	7.2494×10^{18}	
^{239}Pu/g spike	4.0604×10^{16}	
^{241}Pu/g spike	1.5828×10^{17}	
^{242}Pu/g spike	6.3520×10^{18}	$\pm 0.0171 \times 10^{18}$
moles		
$^{}Pu$/g spike	1.2038×10^{-5}	
^{239}Pu/g spike	6.7425×10^{-8}	
^{241}Pu/g spike	2.6283×10^{-7}	
^{242}Pu/g spike	1.0548×10^{-5}	
gram		
$^{}Pu$/g spike	2.9109×10^{-3}	
^{239}Pu/g spike	1.6118×10^{-5}	
^{241}Pu/g spike	6.3357×10^{-5}	
^{242}Pu/g spike	2.5532×10^{-3}	

6. Characterization of the test samples

6.1 Agreed certified values for the B solution (BU) - Uranium Prog. part 1.11

	Results	Uncertainty expressed as 3 $\bar{s}$	
Atom Ratios			
$^{233}U/^{238}U$	---		
$^{234}U/^{238}U$	0.000089	± 0.000001	
$^{235}U/^{238}U$	0.005748	± 0.000005	
$^{236}U/^{238}U$	0.001812	± 0.000013	
Atom %			
^{233}U	---		
^{234}U	0.0088	± 0.0001	
^{235}U	0.5704	± 0.0004	
^{236}U	0.1798	± 0.0014	
^{238}U	99.2410	± 0.0015	
Weight %			
^{233}U	---		
^{234}U	0.0087	± 0.0001	± 1.1 %
^{235}U	0.5633	± 0.0004	± 0.071 %
^{236}U	0.1783	± 0.0014	± 0.79 %
^{238}U	99.2497	± 0.0015	± 0.0015 %
Concentration			
atoms			
U/g sol.	5.1842×10^{18}		
^{235}U/g sol.	2.9570×10^{16}		
moles			
U/g sol.	8.6086×10^{-6}	± 0.0080×10^{-6}	± 0.93 %
^{235}U/g sol.	4.9103×10^{-8}		
gram			
U/g sol.	2.0491×10^{-3}		
^{235}U/g sol.	1.1541×10^{-5}		

6.2 Agreed certified values for the B solution (BU) - Plutonium

Prog. part 1.11 - Pu reference date: 1980.02.09

	Results	Uncertainty expressed as 3 $\bar{s}$	
Atom Ratios			
$^{238}Pu/^{239}Pu$	0.00301	± 0.00003	
$^{240}Pu/^{239}Pu$	0.37011	± 0.00039	
$^{241}Pu/^{239}Pu$	0.04789	± 0.00008	
$^{242}Pu/^{239}Pu$	0.02469	± 0.00007	
Atom %			
^{238}Pu	0.2082	± 0.0017	
^{239}Pu	69.1707	± 0.0254	
^{240}Pu	25.6007	± 0.0203	
^{241}Pu	3.3126	± 0.0053	
^{242}Pu	1.7078	± 0.0044	
Weight %			
^{238}Pu	0.2070	± 0.0017	± 0.82 %
^{239}Pu	69.0631	± 0.0254	± 0.037 %
^{240}Pu	25.6681	± 0.0203	± 0.079 %
^{241}Pu	3.3352	± 0.0053	± 0.16 %
^{242}Pu	1.7266	± 0.0044	± 0.25 %
Concentration			
atoms			
Pu/g sol.	1.8092×10^{16}		
^{239}Pu/g sol.	1.2504×10^{16}		
^{241}Pu/g sol.	5.993×10^{14}		
moles			
Pu/g sol.	3.0042×10^{-8}	$\pm 0.0075 \times 10^{-8}$	± 0.25 %
^{239}Pu/g sol.	2.0780×10^{-8}		
^{241}Pu/g sol.	9.952×10^{-10}		
gram			
Pu/g sol.	7.193×10^{-6}		
^{239}Pu/g sol.	4.967×10^{-6}		
^{241}Pu/g sol.	2.3970×10^{-7}		

6.3 Agreed certified values for the BS dilution ratio - Uranium Prog. part 1.2

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 6)
$^{233}U/^{238}U$	0.85242	± 0.00057

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{233}U/^{238}U$	0.8523	± 0.0011
$^{234}U/^{238}U$	0.002107	± 0.000022
$^{235}U/^{238}U$	0.005854	± 0.000007
$^{236}U/^{238}U$	0.001811	± 0.000014

6.4 Agreed certified values for the BS dilution ratio - Plutonium

Prog. part 1.2 - Pu reference date: 1980.02.09

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 6)
$^{242}Pu/^{239}Pu$	1.0854	± 0.0013

Calculated Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$
$^{238}Pu/^{239}Pu$	0.00151	± 0.00006
$^{240}Pu/^{239}Pu$	0.4728	± 0.0006
$^{241}Pu/^{239}Pu$	0.07420	± 0.00014
$^{242}Pu/^{239}Pu$	1.0844	± 0.0035
$^{244}Pu/^{239}Pu$	0.000233	± 0.000010

6.5 Agreed certified values for the reference solution (RU) - Uranium

Prog. part 2.1

	Results	Uncertainty expressed as 3 $\bar{s}$	
Atom Ratios			
$^{234}U/^{238}U$	0.000092	± 0.000001	
$^{235}U/^{238}U$	0.012353	± 0.000011	
$^{236}U/^{238}U$	0.000068	± 0.000001	
Atom %			
^{234}U	0.0091	± 0.0001	
^{235}U	1.2200	± 0.0011	
^{236}U	0.0067	± 0.0001	
^{238}U	98.7642	± 0.0011	
Weight %			
^{234}U	0.0089	± 0.0001	± 1.1 %
^{235}U	1.2048	± 0.0011	± 0.091 %
^{236}U	0.0067	± 0.0001	± 1.5 %
^{238}U	98.7796	± 0.0011	± 0.0011 %
Concentration			
atoms			
U/g sol.	4.3401×10^{18}		
^{235}U/g sol.	5.2949×10^{16}		
moles			
U/g sol.	7.2070×10^{-6}	± 0.0072×10^{-6}	± 0.10 %
^{235}U/g sol.	8.7925×10^{-8}		
gram			
U/g sol.	1.7154×10^{-3}		
^{235}U/g sol.	2.0666×10^{-5}		

6.6 Agreed certified values for the reference solution (RU) - Plutonium

Prog. part 2.1 - Pu reference date: 1980.02.09

	Results	Uncertainty expressed as 3 $\bar{s}$	
Atom Ratios			
$^{238}Pu/^{239}Pu$	0.00151	± 0.00004	
$^{240}Pu/^{239}Pu$	0.25841	± 0.00022	
$^{241}Pu/^{239}Pu$	0.03550	± 0.00012	
$^{242}Pu/^{239}Pu$	0.00767	± 0.00002	
Atom %			
^{238}Pu	0.1159	± 0.0028	
^{239}Pu	76.7406	± 0.0161	
^{240}Pu	19.8306	± 0.0134	
^{241}Pu	2.7243	± 0.0091	
^{242}Pu	0.5886	± 0.0012	
Weight %			
^{238}Pu	0.1153	± 0.0028	± 2.4 %
^{239}Pu	76.6542	± 0.0161	± 0.021 %
^{240}Pu	19.8912	± 0.0134	± 0.067 %
^{241}Pu	2.7440	± 0.0091	± 0.33 %
^{242}Pu	0.5953	± 0.0012	± 0.20 %
Concentration			
atoms			
Pu/g sol.	2.0086×10^{16}		
^{239}Pu/g sol.	1.5414×10^{16}		
^{241}Pu/g sol.	5.472×10^{14}		
moles			
Pu/g sol.	3.3354×10^{-8}	$\pm 0.0081 \times 10^{-8}$	(± 0.24 %)
^{239}Pu/g sol.	2.5596×10^{-8}		
^{241}Pu/g sol.	9.087×10^{-10}		
gram			
Pu/g sol.	7.982×10^{-6}		
^{239}Pu/g sol.	6.119×10^{-6}		
^{241}Pu/g sol.	2.1904×10^{-7}		

6.7 Agreed certified values for the RS dilution ratio - Uranium Prog. part 2.2

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 6)
$^{233}U/^{238}U$	1.05590	± 0.00089

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{233}U/^{238}U$	1.0564	± 0.0014
$^{234}U/^{238}U$	0.002594	± 0.000027
$^{235}U/^{238}U$	0.012481	± 0.000013
$^{236}U/^{238}U$	0.000068	± 0.000001

6.8 Agreed certified values for the RS dilution ratio - Plutonium

Prog. part 2.2 - Pu reference date: 1980.02.09

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 6)
$^{242}Pu/^{239}Pu$	0.8979	± 0.0010

Calculated Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$
$^{238}Pu/^{239}Pu$	0.011660	± 0.000059
$^{240}Pu/^{239}Pu$	0.34484	± 0.00041
$^{241}Pu/^{239}Pu$	0.05761	± 0.00016
$^{242}Pu/^{239}Pu$	0.8968	± 0.0029
$^{244}Pu/^{239}Pu$	0.000195	± 0.000008

6.9 Agreed certified values for the AS dilution ratio - Uranium Prog. part 1.3

6.9.1 AS I

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 6)
$^{235}U/^{238}U$	0.92759	± 0.00024

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{234}U/^{238}U$	0.007859	± 0.000023
$^{235}U/^{238}U$	0.9274	± 0.0011
$^{236}U/^{238}U$	0.003947	± 0.000015

6.9.2 AS II

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 5)
$^{235}U/^{238}U$	0.90039	± 0.00044

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{234}U/^{238}U$	0.007651	± 0.000025
$^{235}U/^{238}U$	0.9027	± 0.0012
$^{236}U/^{238}U$	0.003890	± 0.000016

6.9.3 AS IV

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 5)
$^{235}U/^{238}U$	0.91032	$\pm$ 0.00043
Calculated Dilution Ratios	**Result**	**Uncertainty expressed as 3 $\bar{s}$**
$^{234}U/^{238}U$	0.007716	$\pm$ 0.000025
$^{235}U/^{238}U$	0.9104	$\pm$ 0.0012
$^{236}U/^{238}U$	0.003908	$\pm$ 0.000016

6.9.4 AS VI

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 5)
$^{235}U/^{238}U$	0.91626	± 0.00034

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{234}U/^{238}U$	0.007769	± 0.000025
$^{235}U/^{238}U$	0.9167	± 0.0012
$^{236}U/^{238}U$	0.003922	± 0.000016

6.10 Agreed certified values for the AS dilution ratio - Plutonium

Prog. part 1.3 - Pu-reference date: 1980.02.09

6.10.1 AS I

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 6)
$^{242}Pu/^{239}Pu$	1.1022	$\pm$ 0.0010

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{238}Pu/^{239}Pu$	0.01555	$\pm$ 0.00008
$^{240}Pu/^{239}Pu$	0.4734	$\pm$ 0.0007
$^{241}Pu/^{239}Pu$	0.07445	$\pm$ 0.00018
$^{242}Pu/^{239}Pu$	1.1036	$\pm$ 0.0057
$^{244}Pu/^{239}Pu$	0.0002315	$\pm$ 0.0000061

6.10.2 AS II

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 5)
$^{242}Pu/^{239}Pu$	1.0701	± 0.0022

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{238}Pu/^{239}Pu$	0.01516	± 0.00009
$^{240}Pu/^{239}Pu$	0.4703	± 0.0008
$^{241}Pu/^{239}Pu$	0.07363	± 0.00020
$^{242}Pu/^{239}Pu$	1.0705	± 0.0061
$^{244}Pu/^{239}Pu$	0.0002244	± 0.0000065

6.10.3 AS IV

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 5)
$^{242}Pu/^{239}Pu$	1.0771	± 0.0014

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{238}Pu/^{239}Pu$	0.01530	± 0.00009
$^{240}Pu/^{239}Pu$	0.4714	± 0.0008
$^{241}Pu/^{239}Pu$	0.07393	± 0.00019
$^{242}Pu/^{239}Pu$	1.0826	± 0.0060
$^{244}Pu/^{239}Pu$	0.0002270	± 0.0000066

6.10.4 AS VI

Measured Dilution Ratio	Result	Uncertainty expressed as 3 $\bar{s}$ (n = 5)
$^{242}Pu/^{239}Pu$	1.0839	± 0.0014

Calculated Dilution Ratios	Result	Uncertainty expressed as 3 $\bar{s}$
$^{238}Pu/^{239}Pu$	0.01539	± 0.00009
$^{240}Pu/^{239}Pu$	0.4721	± 0.0008
$^{241}Pu/^{239}Pu$	0.07412	± 0.00020
$^{242}Pu/^{239}Pu$	1.0904	± 0.0061
$^{244}Pu/^{239}Pu$	0.0002287	± 0.0000066

6.11 Certified values for the elemental concentrations of the A-solution

Progr. part 1.3

	Results	Uncertainties expressed as 3 $\bar{s}$
U-concentration		
atoms		
U/g sol.	4.2922×10^{20}	
^{235}U/g sol.	2.4483×10^{18}	
moles		
U/g sol.	7.1275×10^{-4}	
^{235}U/g sol.	4.0655×10^{-6}	
gram		
U/g sol.	1.6966×10^{-1}	$\pm 0.0012 \times 10^{-1}$
^{235}U/g sol.	9.5557×10^{-4}	
Pu-concentration		
atoms		
Pu/g sol.	1.5023×10^{18}	
^{239}Pu/g sol.	1.0391×10^{18}	
^{241}Pu/g sol.	4.976×10^{16}	
moles		
Pu/g sol.	2.4946×10^{-6}	
^{239}Pu/g sol.	1.7255×10^{-6}	
^{241}Pu/g sol.	8.264×10^{-8}	
gram		
Pu/g sol.	5.973×10^{-4}	$\pm 0.018 \times 10^{-4}$
^{239}Pu/g sol.	4.125×10^{-4}	
^{241}Pu/g sol.	1.9920×10^{-5}	

Appendix 1

C B N M
Mass Spectrometry

GE/NI/MS/02/83
4th January 1982

Physical Constants for use in IDA-80, Safeguard Verification Measurements, etc.

Atomic Masses* and half-lives**

Uranium		
Isotopes	Mass Number	Half-lives (a = years)
232	232.03714	$(6.98 \pm 0.10) \cdot 10^{1}$ a
233	233.03963	$(1.592 \pm 0.002) \cdot 10^{5}$ a
234	234.04095	$(2.454 \pm 0.006) \cdot 10^{5}$ a
235	235.04393	$(7.037 \pm 0.011) \cdot 10^{8}$ a
236	236.94556	$(2.342 \pm 0.003) \cdot 10^{7}$ a
238	238.05079	$(4.468 \pm 0.005) \cdot 10^{9}$ a

Plutonium		
Isotopes	Mass Number	Half-lives (a = years)
238	238.04956	$(8.774 \pm 0.009) \cdot 10^{1}$ a
239	239.05216	$(2.411 \pm 0.003) \cdot 10^{4}$ a
240	240.05381	$(6.55 \pm 0.02) \cdot 10^{3}$ a
241	241.05685	$(1.44 \pm 0.02) \cdot 10^{1}$ a
242	242.05874	$(3.76 \pm 0.02) \cdot 10^{5}$ a
244	244.06420	$(8.2 \pm 0.1) \cdot 10^{7}$ a

* Based on $^{12}C = 12.000000$
taken from A.H. Wapstra, K. Bos: The 1977 Atomic Mass Evaluation, Nucl. Data Tables 19, 175 (1977
Avogadro's number: 6.0220943×10^{23} mol^{-1}

** INDC(NDS)-127/NE to be issued by IAEA 1981 (Lorenz) (for Plutonium) and ongoing evaluation at BNL (for Uranium)

P. DE BIEVRE

Appendix 2

Alpha-Spectrometric Measurements of ^{238}Pu

G. Bortels and A. Verbruggen (CBNM)

he ^{238}Pu has been measured in the following materials

a) diluted input solution, B, progr. part 1.11
b) reference solution, R, progr. part 2.1
c) SUP mixed spike solution

1. Source preparation and measurement conditions

B solution

Two samples of the B solution have been purified separately to remove the ^{241}Am and any ^{228}Th present. The chemical procedure used is described in Appendix 3 of the present report. The purified plutonium was redissolved in 1 M nitric acid at a concentration of about 0.5 $\mu g/\mu l$. Subsequently, one drop-deposition source has been prepared from each of the purified solutions using tetraethylene glycol (TEG) as a spreading agent [1]. Stainless-steel discs were used as source backings. The source activities were about 9370 Bq for both sources. The sources have been measured using a surface barrier detector of 100 mm^2 active area. The energy scale was 0.74 keV/channel in all spectra.

One of the sources was measured in a geometry of about 1.3 %, corresponding to a count rate of 120 s^{-1}. With that source, about $2x10^6$ counts were collected in each spectrum. The energy resolution was 35 keV FWHM. The correction for ^{238}Pu tailing underneath the $^{239+240}Pu$ peaks was less than 0.1 % of the latter.

The second source was measured in a geometry of 0.8 %, corresponding to a count rate of 76 s^{-1}. In this case, a diaphragm of 7.8 mm diameter was placed in front of the detector and a magnetic field was used to prevent conversion electrons from reaching the detector. The elimination of the conversion electrons was needed in an additional study for the fitting of alpha spectra by a computer code. The use of a magnetic field, however, may produce a small effect on the measured alpha activity ratio of $^{238}Pu/^{239+240}Pu$.

These spectra contained each 1.4×10^6 counts. The energy resolution here was 31 keV FWHM. The ^{238}Pu tail correction to the $^{239+240}Pu$ group was only 0.03 % of the latter. The background correction to the ^{238}Pu group was less than 0.01 % of that group.

The measurements on the B solution started 1984.06.14 and were finished on 1984.06.19; 12 spectra have been measured.

R solution

Two samples of the R solution were purified separately as in the case of the B solution. Also, one TEG drop-deposition source was subsequently prepared for each sample; the source activities were 11700 and 13670 Bq. The same 100 mm^2 SB detector and diaphragm were also used here. The source-to-detector geometry of 0.8 % corresponded to count rates of 95 and 110 s^{-1} for these sources. With one of the sources, a magnetic field has been used during the measurements to eliminate conversion electrons. A total number of 15 spectra with 0.74 keV/channel were measured containing each from 2×10^6 to 2.4×10^6 counts. The energy resolution was 18 keV FWHM for all spectra. The correction to the $^{239+240}Pu$ group as a result of ^{238}Pu tailing amounted to only 0.03 %. The background correction at the ^{238}Pu peak was less than 0.01 % of the latter peak. The measurements on the R solution started 1984.06.22 and finished 1984.06.26.

SUP solution

One sample of the SUP solution was purified as for the preceding solutions. One TEG drop-deposition source of 2140 Bq activity was prepared. The source was measured in a geometry of 2.8 % which corresponds to a count rate of 60 s^{-1}. 13 spectra, with 074 keV/channel and containing each 10^6 counts, were measured. The energy resolution was 19.6 keV FWHM. The same 100 mm^2 SB detector was used but without diaphragm. The correction to the $^{239+240}Pu$ group, resulting from the ^{238}Pu tailing, was 1.9 % of the former. This relatively high figure is due to the high ^{238}Pu abundance in the SUP sample. As in all previous cases, the correction for background counts was completely negligible. The measurements on the SUP sample started 1984.02.10 and were finished 1984.02.14.

2. Results

The spectra have been analysed by visual interpretation as published earlier [2]. The ^{238}Pu group has been corrected for ^{241}Am grown into the source since the time of purification. For this correction, the ^{241}Pu abundance from the mass spectrometric measurements was used. As the energy range considered for the $^{239+240}Pu$ group contains 3.1 % of the ^{241}Pu alpha activity this was corrected for. The $(^{238}Pu/^{239}Pu)_o$ abundance ratio at the reference date of 1980.02.09 (t = o) is obtained from the alpha-activity ratio $R_t = (^{238}Pu/^{239+240}Pu)$ at the time of measurement t from the expression

$$\left(\frac{A_{38}}{A_{39}}\right)_o = \frac{R_t}{\lambda_{38}\, e^{-\lambda_{38}t}} \left[\lambda_{39}\, e^{-\lambda_{39}t} + \left(\frac{A_{40}}{A_{39}}\right)_o \lambda_{40}\, e^{-\lambda_{40}t}\right] \qquad (1)$$

Here, $(A_{38})_o$, $(A_{39})_o$ and $(A_{40})_o$ denote, respectively, the ^{238}Pu, ^{239}Pu and ^{240}Pu abundances at the reference date. The decay constants ($\lambda = \ln 2/T_{1/2}$) are indicated in a similar way by λ_{38}, λ_{39} and λ_{40}. The following half-life values and overall uncertainties (standard deviation for the mean), were used [3]

^{238}Pu $T_{1/2}$ = (87.74 ± 0.09) a
^{239}Pu $T_{1/2}$ = (24110 ± 30) a
^{240}Pu $T_{1/2}$ = (6550 ± 20) a
^{241}Pu $T_{1/2}$ = (14.4 ± 0.2) a.

The results of R_t for the B, R and SUP solutions and the corresponding uncertainty components are given in Table 1. The overall uncertainty is obtained from the quadratic sum of the uncertainty components.

Using tne results for R_t from Table 1 and the abundances $(A_{39})_o$ and $(A_{40})_o$ (see sections 5.2, 6.2 and 6.6) the expression 1 provides the results which are given in column 2 of the Table 2. Here, the quoted uncertainties are three times the uncertainties obtained from error propagation following the expression 1 in which overall uncertainties of one standard deviation for the mean were introduced for all parameters. Hence, quoted uncertainties correspond to a 99.7 % confidence level. The results from mass-spectrometric measurements are also shown for comparison.

Table 1. Measured alpha-activity ratios $R_t = {}^{238}Pu/{}^{239+240}Pu$ and overall uncertainties (st. dev. mean) at the date of measurement

	B	R	SUP
R_t	0.3374	0.2077	8.3673
Overall uncertainty (st.dev.mean)	± 0.0003	± 0.0002	± 0.015
Number of measurements	12	15	13
Uncertainty components			
Random (st.dev.mean)	± 0.0002	± 0.0001	± 0.007
Tailing	---	---	± 0.012
^{241}Am separation	± 0.0002	± 0.0001	± 0.003
Ratio distortion due to magnetic field	± 0.0001	± 0.0001	--
Overall uncertainty	± 0.0003	± 0.0002	± 0.015

Table 2. Comparison of $^{238}Pu/^{239}Pu$ abundance ratios as of 1980.02.09, measured by alpha particle spectrometry and MS-measurements

	CBNM MS measurements $^{238}Pu/^{239}Pu$	CBNM α-spectrometric measurements $^{238}Pu/^{239}Pu$	NBS MS measurements $^{238}Pu/^{239}Pu$
B solution	0.003009 ± 0.000019	0.003000 ± 0.000020	0.003014 ± 0.000030
R solution	0.001517 ± 0.000003	0.001526 ± 0.000010	0.001502 ± 0.000052
SUP solution	3.856 ± 0.020	3.857 ± 0.050	3.809 ± 0.048

References

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Appendix 3

Purification of plutonium for alpha spectrometric measurements

A. Verbruggen (CBNM)

The alpha spectrometric determination of ^{238}Pu with high precision and accuracy requires a purification procedure for plutonium.

A method for the purification of microgram amounts of plutonium using anion exchange techniques has been developped for synthetic input solutions containing fission products and in particular for the IDA-80 test samples. Special care has been taken to avoid thorium and americium contamination of the plutonium sample.

1. Outline of the method

The ability of the lighter actinides to form anions in mineral acids has led to the use of anion exchange techniques for their separation from other elements.

The purification method is based on the strong adsorption of Pu(IV) as $[Pu(NO_3)_6]^{2-}$ by anion exchangé resins in nitric acid medium. Distribution coefficients are less than 10^3 for acid concentrations of 6 M and higher.

On the other hand, the hexa- and pentavalent oxidation states are adsorbed with a distribution coefficient of 20 or less, while plutonium in the trivalent state shows only negligible or no adsorption.

Transplutonium elements are not adsorbed, or only adsorbed to a negligible extent at acid concentrations from 1 to 14 M.

Metallic impurities are not or very slightly adsorbed on anion exchange resin.

Nitrate complexes of UO_2^{2+} are only weakly adsorbed by strongly basic anion exchangers in nitric acid solutions. Distribution coefficient value is smaller than 2 in 1 M HNO_3 and increases up to approximately 20 in 6-8 M HNO_3. Further increase in nitric acid concentration leads to decrease in adsorption of the complex (Dg is approximately 6 in 14 M HNO_3).

Thorium in the tetravalent state forms a stable anionic nitrate complex in nitric acid solutions corresponding to the formula $[Th(NO_3)_6]^{2-}$. This hexanitrato complex is very well adsorbed on an anion exchanger with distribution coefficient of about 300 in 6 M HNO_3. The failure of thorium to

form anionic chloride complexes to an appreciable extent can be utilised to separate thorium from plutonium because it forms anionic chloride complexes which are readily retained by anion exchange resins. Distribution coefficient for Pu(IV) larger than 10^3 for acid concentrations higher than 8 M HCl.

Plutonium can readily be eluted by means of nitric acid or hydrochloric acid solutions of concentrations such as 0.35 M HNO_3 or 1 M HCl in which the distribution coefficient is sufficiently low.

2. Reagents and Laboratory Ware

- Anion exchange resin: DOWEX 1-X4, 100-200 mesh in chloride form. The original resin in the chloride form was converted to the nitrate form with 8 M HNO_3.
- Water: purified by a Milli-Q milipore water treatment installation including reversed-osmose, carbon filter, mixed ion exchange and 0.22 μm membrane filter.
- Nitric acid: Merck grade suprapure.
- Column: Two sizes made of borosilicate glass, length about 125 mm and 65 mm with inside and outside diameters of 4 mm and 6 mm. The last 15 mm were tapered to an opening of 0.5 to 1 mm. The column had a 25 ml reservoir at the top and was fitted with a quartz wool plug at the bottom to retain the resin. Volume of the largest is 1.4 ml and of the smaller one is 0.7 ml.
- Iron(II)chloride, Hydroxylammoniumchloride, Sodiumnititre: Merck reagent grade.

Eppendorf pipettes were used for sample transfers during the procedure.

3. Procedure

1. Evaporate the sample to dryness, plutonium is oxidized into the hexavalent state Pu(VI).
2. Redissolve residue in 200 μl 2 M HNO_3.
3. Reduce Pu(VI) to Pu(III) by adding 25 μl 1.25 M $FeCl_2$; mix and wait about 5 min.

4. Add 100 μl 1 M $NH_2OH.HCl$ which will stabilise plutonium as Pu(IV) after the oxidation in the next step; mix and wait 5 min.
5. Add 100 μl 1 M $NaNO_2$ to oxidize Pu(III) to Pu(IV); mix and wait 5 min.
6. Add 430 μl concentrated HNO_3 to obtain 8 M HNO_3 solution.
7. Transfer to large preconditioned anion exchange column.
8. Pass 3 ml 8 M HNO_3 to the column in 1 ml increments. Reservoir walls have to be rinsed with the 1 ml increment. Wait between increments until reservoir is empty. Fission products, transplutonium elements and metallic impurities are eluted.
9. Add 30 ml 8 M HNO_3 to elute uranium fraction.
10. Add 10 ml 0.35 M HNO_3 to elute plutonium fraction.
11. Repeat steps 1 to 6.
12. Transfer to small preconditioned anion exchanger column.
13. Add 15 ml 8 M HNO_3 to the column in 5 ml increments to wash residues of previous steps.
14. Add 15 ml 9 M HCl to the column in 5 ml increments to elute thorium.
15. Add 6 ml 0.35 M HNO_3 to elute plutonium.

Recovery of plutonium is about 100 %. Efficiency of separation is better than 99 %. This method is applicable for plutonium amounts up to 500 μg. The U/Pu ratio may vary between 1 and 500 without having any influence on the separation efficiency.

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List of Tables

Chapter III: Compilation of technical and administrative requirements for the shipment of the test samples to the participating laboratories

by

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1. Introduction

Because of the large variety of national and international regulations and the size of IDA-80, the transport of the small fissile test samples required a considerable amount of labor and expense. This Chapter reports on the shipment of these small amounts of radioactive material and on all the different formalities which had to be fulfilled.

Samples totalling about 100 mg of U and 0.4 mg of Pu, were sent to 36 laboratories in 13 countries. Total cost of this operation reached US $ 40.000 (1980 dollars).

The preparation for packaging and transportation has been done by CBNM. Transnubel Co., Dessel, Belgium was charged with the actual transport to the different laboratories.

We report on our experiences with the IDA-80 samples for the following reasons:

1. We offer it as a 1980 picture of what the real situation is.
2. We hope to encourage regulating Authorities to allow a quick(er) and simple(r) movement of small samples across borders since such a movement is essential for good and fast verification measurements in international Safeguards.

2. Shipping containers

The shipment of these samples was performed by air, sea or by surface transport in certified Type B containers. The DOT 6M-15 gallon container presented the most favourable ratio total weight/total price.

For shipment to the National Bureau of Standards (NBS) Washington, 1 DOT 6M-55 gallon and 1 DOT 6M-110 gallon container were used due to the large number of samples shipped for joint characterization.

The DOT 6M containers were approved by the US competent authorities: see an example of the required certificate USA/0002/B()F in Appendix 3. The approvals were recognized by all other countries concerned. In total 109 DOT 6M containers were shipped.

3. Distribution of the IDA-80 test samples over the transport containers

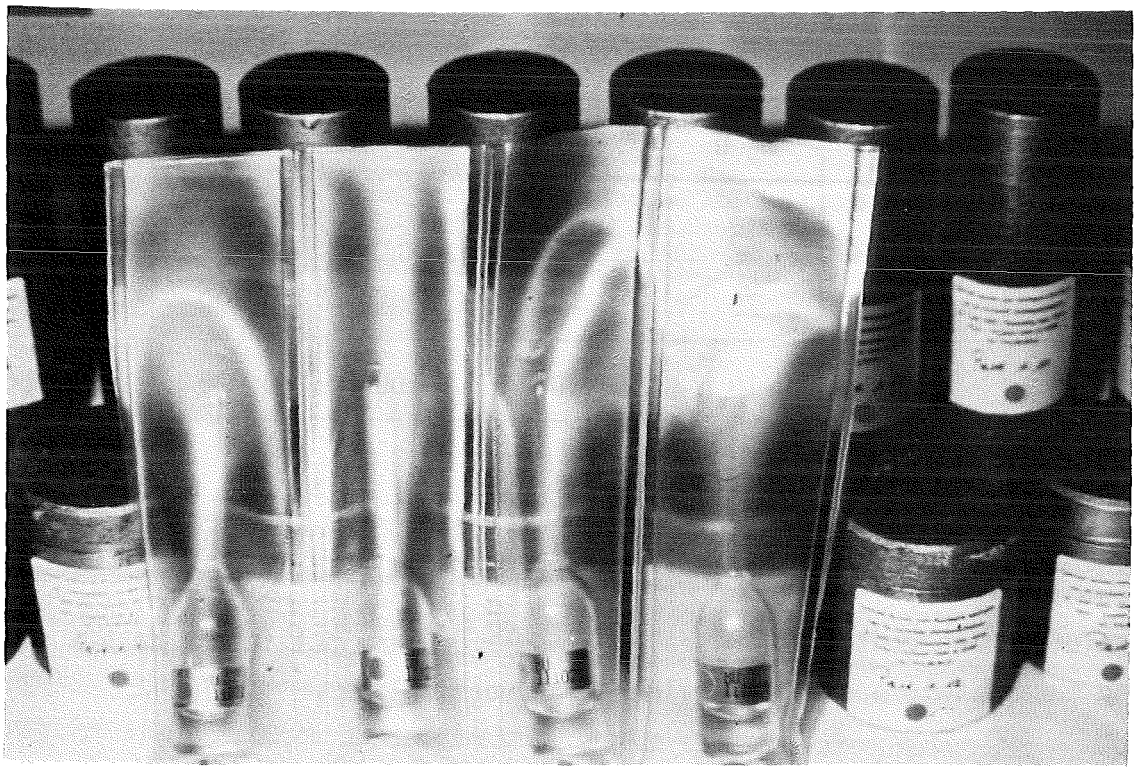
For each participating laboratory 11 test samples were dispatched containing a total amount of 97.65 mg U and 0.395 mg Pu.

They were shipped in three DOT 6M transport containers. The origin and isotopic composition of the different samples in the three containers were the following:

Samples	Origin	mg U	$^{233}U/U$	$^{235}U/U$	µg Pu	$^{239}Pu/Pu$
Bu-sol.	US	12.5	-	0.006	48.75	0.69
Bu dried	US	7.5	-	0.006	29.25	0.69

Table 3.1 First sample container (see content in Photogr. 3.1)

See schematic drawing "Container 1" in Appendix 1.



Photogr. 3.1: Liquid test samples in sealed glass ampoules as well as lead container

The solutions were contained in sealed glass ampoules (see Photogr. 3.1) and the dried samples in glass vials with plastic stoppers. Each closed vial was put into a single plastic bag. Because of the γ-radiation (diluted input solution) the samples were contained in two lead containers. These were packed into a steel container with vermiculite. See Photogr. 3.2 and schematic drawing "Container 1" in Appendix 1.

Samples	Origin	mg U	$^{233}U/U$	$^{235}U/U$	μg Pu	$^{239}Pu/Pu$
BS	US	6.25	-	0.006	22.7	0.69
	EC	5.0	0.996	-	20.8	0.003
AS dried	UC	12.4	-	0.006	45.0	0.69
	EC	12.4	-	0.88	37.5	0.006

Table 3.2 Second sample container

See schematic drawing "Container 2" in Appendix 1.

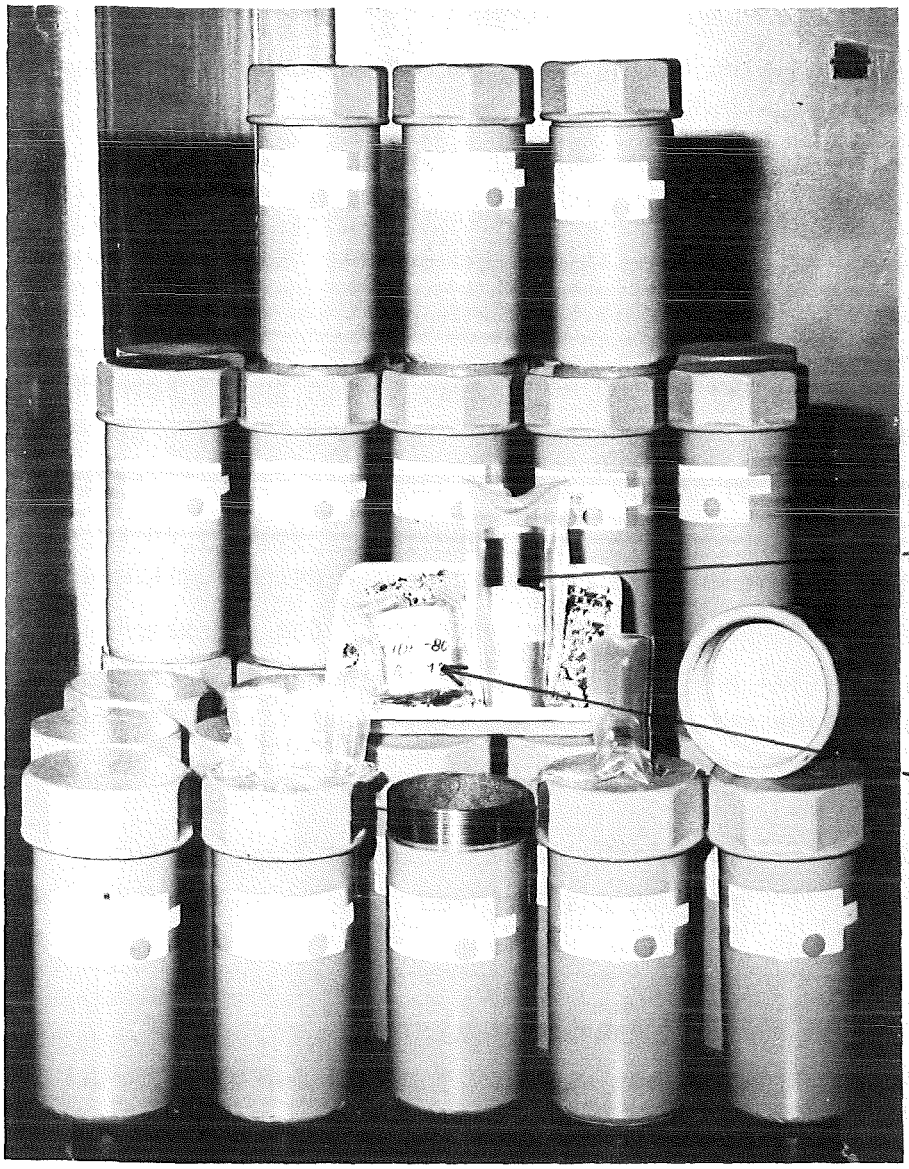
Solution	Origin	mg U	$^{233}U/U$	$^{235}U/U$	mg Pu	$^{239}Pu/Pu$
SUP	EC	10	0.996	-	0.040	0.003
RU	EC	20	-	0.012	0.100	0.767
RS	EC	10	0.49	0.006	0.045	0.386

Table 3.3 Third sample container

See schematic drawing "Container 3" in Appendix 1.

Each solution was contained in sealed glass ampoules, put into a double plastic bag and packed into a steel container with vermiculite.

A survey of all samples supplied per laboratory is given in Appendix 2.



Photogr. 3.2 Steel containers

4. Formalities for material of US origin

The total amount of material dispatched to each laboratory was less than 100 mg U and less than 0.5 mg Pu. The "US origin" part of this material was maximum 39.1 mg U containing 0.6 % ^{235}U and maximum 147 μg Pu. It corresponded to the input solution spiked with SUP and solid spike material.

For this part approval of the Department of Energy (DOE) Washington was required. The request for approval was initiated by CBNM and handled by the EC Supply Agency. The formalities required from "US-DOE" for sample shipment can be classified in four categories:

4.1 For participants in countries having an agreement for cooperation in the peaceful use of atomic energy with the US Government:
an approval by DOE on an MB 10 Form, called "Retransfer of Special Nuclear Material of United States Origin" was required. This was the case for Japan, Denmark, Austria, Finland and Brasil. See an example in Appendix 4.

4.2 For participants in countries having no agreement with the US Government:
the IAEA in Vienna was prepared to take over the responsibility of the material under the provision of the USA-IAEA agreement for collaboration. This was the case for USSR and CSSR (time required: 2 years): an approval on a MB 10 form had to be requested by IAEA from the US-DOE with special indication of the laboratory and country concerned. See an example in Appendix 4a. The material was shipped to IAEA in Vienna and forwarded to the participants after this approval was obtained.

4.3 For all US participants:
a MB-4 Form "Division of International Affairs Import Approval for Special Nuclear Material" from DOE was required. See an example in Appendix 5.

4.4 For all laboratories in the European Community:
no prior authorization from DOE was required for transfer of material of US Origin within the European Community.

In the cases 4.1 through to 4.4 DOE had to be notified after each shipment through a SER-1 Form and receipt signed by the EC Supply Agency. See an example in Appendix 6.

5. Practical organization and execution of the transport

The shipments were divided into four groups:

5.1 The first group concerned the shipments to the Federal Republic of Germany.
The shipments to the following consignees were performed on 17th July 1980 by air from Brussels to Frankfurt Airport and subsequently by road through Transnuklear Hanau, subcontractor for Transnubel Dessel

- Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen mbH, Eggenstein-Leopoldshafen (WAK Karlsruhe)
- Euratom-Europäisches Institut für Transurane, Karlsruhe (TUI)
- Kernforschungszentrum Karlsruhe GmbH, Karlsruhe (KfK)
- Kraftwerk Union, Karlstein (KWU).

The air transport was performed in accordance with IATA Restricted Regulations Part 2A Class 7 and to the ADR Regulations of Dangerous Goods for Surface Transportation. The transport authorization was granted by the "Physikalisch Technische Bundesanstalt (PTB) Braunschweig. See an example in Appendix 15.

Furthermore, the transport documents had to be accompanied by an Import Notification (§ 12 Abs. 1 Strahlenschutz-Verordnung) required by the Bundesanstalt für gewerbliche Wirtschaft, Frankfurterstrasse 29-31, Postfach 5171, D-6236 Eschborn/TS 1. See an example in Appendix 14.

The shipment to the Institut für Chemische Technologie, Kernforschungsanlage GmbH in JÜlich was performed by road on 25th July 1981 in accordance with Belgian and German authority regulations based on the ADR regulations. See an example in Appendix 15.

The shipment to the Bundesanstalt für Materialprüfung (BAM) in Berlin was delayed until 15th September 1981 on request of the consignee and was performed in accordance with

a) IATA regulations
b) Federal Republic of Germany regulations
c) with the transport permit of 'Der Senator für Wirtschaft und Verkehr' in Berlin.

See an example in Appendices 16 and 17.

All required formalities for air and surface transportations are summarized in Point 7.

5.2 The second group concerned the shipments to the following consignees in the U.S.A.:

- Vallecitos Nuclear Center, Pleasanton CA 94566;
- E.I. du Pont de Nemours & Co., Aiken SC 29801;
- Los Alamos National Laboratory, Los Alamos NM 87545;
- Oak Ridge National Laboratory, Oak Ridge TN 37830;
- Westinghouse Handford Co., Richland WA 99352;

- Analytical Labs.-Atomic Int. Div. - Rocky Flats Plant, Golden CO 8049;
- Analytical Chemistry Lab., Argonne IL 60439;
- Exxon Nuclear Idaho, Idaho Falls ID 83401;
- US Department of Energy-New Brunswick Laboratory, Argonne IL 60439;
- Allied General Nuclear Services, Barnwell SC 29812;
- Argonne National Laboratory, Idaho Falls ID 83401;
- National Bureau of Standards, Washington DC 20234.

All packages for USA were loaded in one 20 feet sea container and addressed to Transnuclear Inc. Skyline Centre, 5205 Leesburg Pike, Falls Church, Virginia 22041, USA, subcontractor of Transnubel.

The transport was performed on 14th September 1980 immediately after the issuance of transport permits in the USA and MB-4 Forms, covering the transport of fissile materials under an agreement for cooperation between the USA and the European Atomic Energy Community.

The shipment of the sea container was performed in accordance with Belgian and Dutch regulations for the road transportation from CBNM to Rotterdam Harbour. (Belgium Regulations' Law of 29.3.58; Royal Decree 28.2.63., Royal Decree 27.6.66. Netherlands Regulations, Kernenergie Wet - 1963, Besluit vervoer van splijtstoffen 1969).

From Rotterdam to Portsmouth VA (USA), the sea transport was performed in accordance with IMCO (Inter-Governmental Maritime Consultative Organization) regulations.

The distribution to the US consignees was performed by the above-mentioned subcontractor of Transnubel Dessel.

5.3 The third group concerned five shipments performed by surface transportation in accordance with ADR and the regulations in force in France, this transport was arranged by the subcontractor Transnucléaire S.A., rue C. Colomb 11, F-75008 Paris, Netherland and Belgium:

- on 18th July 1981 to SCK/CEN Mol, Belgium;
- on 22nd July 1980 to the Netherlands Energy Research Foundation (ECN) Petten, Netherland;
- on 31st July 1980 to
  Centre d'Etudes Nucléaires (CEN) Saclay, Gif-sur-Yvette, France
  Centre de l'Energie Atomique (CEA) Montrouge, France;
  Centre d'Etudes de Bruyères-le-Châtel, Montrouge, France.

5.4 The fourth group is related to the air shipment, according to the IATA regulations, namely to the following consignees:

- on 16th July 1980 to
  the Power Reactor and Nuclear Fuel Development Corporation, Tokyo, Japan;
  the British Nuclear Fuel Ltd (BNFL), Windscale, UK;
  the AERE, Harwell, UK;
  Cogema, Marcoule, France;
  the "Österreichische Studiengesellschaft für Atomenergie GmbH, Vienna, Austria;
  the AWRE, Aldermaston, UK;
  Tokai Research, Japan;
  the Risø National Laboratory, Roskilde, Denmark;
- on 1st October 1980 to
  the University of Helsinki, Helsinki, Finland;
- on 15th January 1980 to
  the IAEA, Vienna, Austria
  the Instituto de Pequioas Energéticas e Nucleares, Sao Paolo, Brasil.

6. Third party nuclear liability insurance

Transport of special nuclear materials, as well as non-fissile radioactive materials, has to be covered by an appropriate insurance.

For transports to the United States the Third Party Nuclear Liability was covered up to 3 miles from the continental coast by the Insurance Boels Begault, rue des Chevaliers 13, B.1050 Brussel.

Transports to Österreich, Brasil, Denmark, Suomi, France, Japan, Netherland, United Kingdom were insured until the airport by the same insurance.

In all countries, the carriage of special nuclear materials can only be performed when they are covered in accordance with the local national insurance regulations. For example, in the United States the insurance must be subscribed by the operator of a nuclear facility and the additional governmental indemnity is granted by the "Price Anderson Amendment" (Nuclear Safety Vol. 16, No. 5, Oct. 75). For Germany and West Berlin transports of special nuclear materials can only be performed when they are covered by the "Deutsche Reaktorversicherungsgemeinschaft", Sedanstrasse 8, Köln.

It is to be noted that the insurance premiums depend upon the transport means. Indeed, the size of the premium increases heavily from surface to air to sea transportation.

Section 7 - see over.

7. General View of Required Formalities

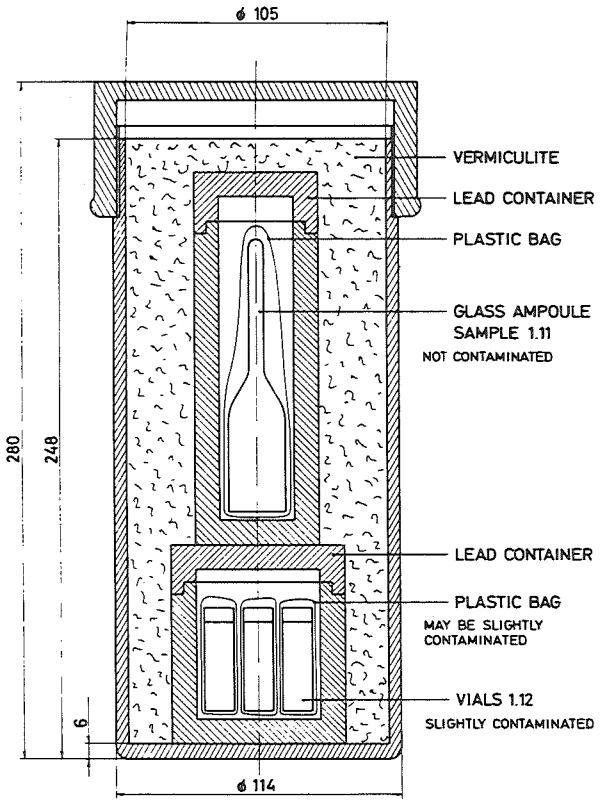
	Austria	Brazil	Belg.	Denm.	FRG	Finl.	France	Japan	Nether land	UK	USA
	App.	App.	App.	App.	App.	App.	App.	App.	App.	App.	App.
Transport Authorisation	•	•	•	•	•	•	•	•	•	•	•
Transport Authorisation destination countries		sub contr.			sub contr.			sub contr.			sub contr.
Export Declaration	13	13		13	13	13	13	13	13	13	13
Import Declaration					14						
Liability Insurance	•	•	•	•	•	•	•	•	•	•	•
Eur. Safeguards Decl. Luxemburg	7	7	7	7	7	7	7	7	7	7	7
Note for Transmission	8	8	8	8	8	8	8	8	8	8	8
Proforma Invoice and Consignment Note	9	9	9	9	9	9	9	9	9	9	9
Schematic Packing	10	10	10	10	10	10	10	10	10	10	10
Shippers Note Sabena	11	11		11		11		11		11	11

In the Table all documents required by CBNM, national and international authorities are summarized.

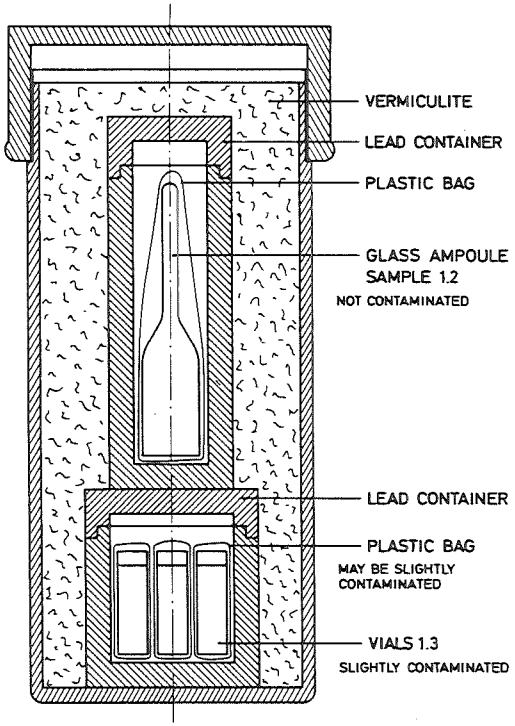
For the internal CBNM Safeguards' Declarations, App. 7 and 8, in total 300 forms had to be prepared.

7. General View of Required Formalities (Continuation)

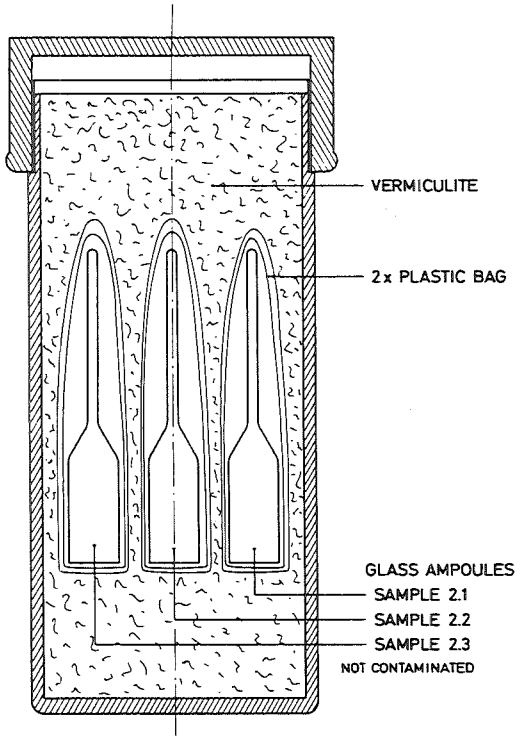
	Austria	Brazil	Belg.	Denm.	FRG	Finl.	France	Japan	Nether land	UK	USA
	App.	App.	App.	App.	App.	App.	App.	App.	App.	App.	App.
Surface Transport ADR Regulation			•		•		•		•		
Air IATA Restricted Regulations	•	•		•	•	•	•	•		•	
Sea IMCO Regulations											•
Shipper Certification	12	12		12	12	12	12	12		12	12
MB 10	4	4				4		4			
MB 4											5
SER - 1	6	6				6		6			6
Certificate Type B Container	3	3	3	3	3	3	3	3	3	3	3
Notification to Transferee before Shipment	•	•	•	•	•	•	•	•	•	•	•



CONTAINER 1



CONTAINER 2



CONTAINER 3

	AFGELEID	EURATOM C.B.N.M. GEEL
DATER	BEHANDELING	BEW.
Wolters	IDA-80 SCHEMATIC PACKING	T 1867
De Bièvre		AANT. BL. 1 / BLAD Nr 1 / SCHAAL
GOT. DAENS-LUYTS.ENTS	DAT. 12.5.1980	VERV.

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16			17	18	19
Part of program	Sample description	Physical state	Approx. amount per sample			Approximate isotope abundances (%)									Approx. dose rate[a] (mrem/h) distance (cm)			Color code[b]	Identification numbers	Transport container color code
			Volume (ml)	U (mg)	Pu (µg)	Uranium 233	Uranium 235	Uranium 236	Uranium 238	Plutonium 238	Plutonium 239	Plutonium 240	Plutonium 241	Plutonium 242	0	10	50			
1.11	BU	liquid	5	12	40	-	0.6	0.2	99	0.2	70	25	3	2	60	10	1	yellow	111 001 to 111 050	
1.12	BU I, BU II BU III	dried	-	3	10	-	0.6	0.2	99	0.2	70	25	3	2	120	20	1	yellow	112 001 to 112 150	yellow
1.2	BS	liquid	5	11	40(20)[c]	44	0.3	0.1	55	0.6	38	17	3	42	30	5	0.5	yellow	120 001 to 120 050	
1.3	3 vials out of AS I to AS VI	dried	-	8	25(13)[c]	-	44	0.1	56	0.6	38	17	3	42	180	28	1.5	red	130 101 to 130 650	red / yellow
2.1 [d]	RU	liquid	10	20	100	-	1	-	99	0.1	75	20	3	1	-	-	-	blue	213 001 to 213 050	
2.2	RS	liquid	5	10	45	50	0.5	-	49.5	0.5	38	14	3	44	-	-	-	blue	220 001 to 220 050	blue
2.3 [e]	SUP	liquid	5	10	40	100	-	-	0.3	1	1	8	2	87	-	-	-	white	230 001 to 230 050	

IDA-80 : Survey of samples supplied per laboratory

a) Upper limits measured in direct contact with the sample and at a distance of 10 and 50 cm

b) Caution! All yellow and red labeled samples contain fission products of high γ-activity!

c) Values in brackets indicate the amounts of Pu originating from sample solutions A or B from which fission product contents can be derived (see Table II)

d) Fraction of the RU-sample material supplied with part 2.1 is used in part 2.3

e) For the RU-material needed for this part, refer to the material mentioned in part 2.1

Note: all amounts and abundances given in this table should be considered as rough approximations only.

CAUTION! All yellow and red labeled samples contain fission products of high γ-activity!

U.S. Department of Transportation
Research and Special Programs Administration

400 Seventh Street, S.W.
Washington, D.C. 20590

(subsequent pages have been omitted)

IAEA CERTIFICATE OF COMPETENT AUTHORITY

Type B Fissile Radioactive Material Package Design

Certificate Number USA/0002/B()F
(Revision 2)

This establishes that the packaging design described herein, when loaded with the authorized radioactive contents, has been certified by the National Competent Authority of the United States as meeting the regulatory requirements for Type B packaging for radioactive materials, including fissile radioactive materials, as prescribed in IAEA[1] Regulations and in accordance with §§ 49 CFR 173.393b of the USA[2] Regulations for the transport of radioactive materials.

I. Package Identification - DOT Specification 6M.

II. Packaging Description - The packaging authorized by this certificate consists of a metal, drum-type birdcage which utilizes a sealed DOT Specification 2R (Appendix A) metal inner containment vessel, centered and supported within a removable head type metal outer drum by means of machined discs and rings of specified solid materials which provide thermal and impact protection. Packaging must comply with the provisions of 49 CFR 178.104, attached as Appendix B hereto. Typical assembly detail for a 10-gallon configuration DOT-6M is illustrated on Appendix C hereto.

III. Authorized Radioactive Contents - The authorized contents consist of fissile or non-fissile radioactive materials in special or non-special form which will not decompose at temperatures up to 250°F (121°C). Contents exceeding the Type B limits as specified in 49 CFR 173.389(1) must be additionally contained within one or more sealed and leak-tight metal cans or polyethylene bottles within the DOT Spec. 2R inner vessel. The radioactive thermal decay energy output of the contents shall not exceed 10 watts.

Additional limitations for fissile radioactive material contents are as follows:

(i) Fissile Class-I Packages. The following quantities of fissile radioactive material are authorized for Fissile Class I packages: 1.6 kilograms uranium-235; 0.9 kilograms of plutonium (See Note); 0.5 kilograms of uranium-233. The maximum ratio of hydrogen to fissile materials must not exceed three, all sources of hydrogen within the Spec. 2 R containment vessel being considered.

(NOTE: Because of the 10-watt thermal decay heat limitation, the limit for plutonium-238 is only 0.02 kilograms.)

App. 4

MB/10

Number RTD/TA(EU)-19
AG/1459/7

APPROVAL FOR RETRANSFER OF SPECIAL NUCLEAR MATERIAL
OF UNITED STATES ORIGIN

The approval of the United States Department of Energy is hereby requested to the transfer

from EURATOM SUPPLY AGENCY (for BCMN Geel)
(Transferor)

to Government of Japan (for the Power Reactor and Nuclear Fuel Development Corporation)
(Transferee)

of United States supplied special nuclear material in the quantity and meeting the specifications described below (hereinafter called "specified material") which the transferor obtained pursuant to its Agreement for Cooperation for Civil Uses with the United States Government. Material was originally obtained by transferor from UES/EU/4, under Contract or Order Number AGT/4.

SPECIFIED MATERIAL
(Fill in where applicable)

Identification				
Fuel Type	Marking, No., etc.	Total U (In Grams)	U-235, U-233 or Pu (In Grams)	Isotopic Percent U-235, U-233, or I
test samples		100 mg U 0.5 mg Pu		3 %

The specified material, which is now located at Geel, will upon approval hereby by the United States Department of Energy be transferre on or about 1979-1980 for use at see below (*) and will be accepted for the following specified purpose:

Several dispatches in the microgram range during the ESARDA IDA-78 programme : to determine the uncertainty of the analytical measurements of U and Pu isotopes

The transferor, with the concurrence of the transferee, will notify within 30 days after the aforesaid date the United States Department of Energy of the actual date and quantity of material transferred. It is agreed by the transferor and transferee that as of that date the specified material will cease to be subject to the Agreement for Cooperation and contract indicated above and will be subject to the transferee's Agreement for Cooperation for Civil Uses with the United States Government.

EURATOM SUPPLY AGENCY 11. 1. 1980
p. J.B. MENNICKEN, Director General
(Transferor) (Date)

The Government of Japan
Masayasu Miyabayashi March 12, 1980
(Transferee) (Date)

Above requested transfer under Article IV A of transferee's Agreement for Cooperation for Civil Uses with the United States Government approved provided physical transfer is consummated by May 5, 1981

x Harold M. Kouts May 5, 1980
(For the United States Department of Energy)

(*) Dr. Susumu Suzuki, Manager, Nuclear Fuels Division, Power Reactor and Nuclear Fuel Development Corporation, 9-13, 1-chome, Akasaka, Minato-ku, Tokyo/Japan

App. 4a

NB#10

Number RTD/IAEA(EU)-15

AG/1459/9

APPROVAL FOR RETRANSFER OF SPECIAL NUCLEAR MATERIAL OF UNITED STATES ORIGIN

The approval of the United States Department of Energy is hereby requested to the transfer

from EURATOM SUPPLY AGENCY (for BCMN Geel)
(Transferor)

to International Atomic Energy Agency (IAEA), Vienna, Austria (for V.G. Khlopin
(Transferee) Radium Institute, Leningrad)

of United States supplied special nuclear material in the quantity and meeting the specifications described below (hereinafter called "specified material") which the transferor obtained pursuant to its Agreement for Cooperation for Civil Uses with the United States Government. Material was originally obtained by transferor from UES/EU/4, under Contract or Order Number AGT/4.

SPECIFIED MATERIAL
(Fill in where applicable)

Identification				
Fuel Type	Marking, No., etc.	Total U (In Grams)	U-235, U-233 or Pu (In Grams)	Isotopic Percent U-235, U-233, or Pu
test samples		100 mg U 0.5 mg Pu		3 %

The specified material, which is now located at Geel, will upon approval hereby by the United States Department of Energy be transferred on or about 1979-1980 for use at see below (*) and will be accepted for the following specified purpose:

Several dispatches in the microgram range during the ESARDA IDA-78 programme : to determine the uncertainty of the analytical measurements of U and Pu isotopes

The transferor, with the concurrence of the transferee, will notify within 30 days after the aforesaid date the United States Department of Energy the actual date and quantity of material transferred. It is agreed by the transferor and transferee that as of that date the specified material will cease to be subject to the Agreement for Cooperation and contract indicated above and will be subject to the transferee's Agreement for Cooperation for Civil Uses with the United States Government.

EURATOM SUPPLY AGENCY 22 April, 1981 DEVELOPMENT DIV'N, IAEA

for J.B. MENNICKEN, Director General — R.E. Kerrebrock, Acting Director 27 May 1981

(Transferor) (Date) (Transferee) (Date)

Above requested transfer under Article II D of transferee's Agreement for Cooperation for Civil Uses with the United States Government approved, provided physical transfer is consummated by November 17, 1982

Harold D Bengelsdorf November 17, 1981

(For the United States Department of Energy)

(*) Mr. A.A. Lipovsky, V.G. Khlopin Radium Institute, Roentgen 1, 197022 Leningrad/USSR

MB-4

AG/1459/10 (13)

DIVISION OF INTERNATIONAL AFFAIRS IMPORT APPROVAL
FOR SPECIAL NUCLEAR MATERIAL

1. Shipper: - Transnubel / Europalaan 20 / B-2480 DESSEL, Belgium
   - Mr. J. Van Audenhove, CBNM, Geel, Belgium
   - Material shipped from CBNM

2. U.S. consignee: Mr. Ernest Garner
   Chief, Inorganic Analytical Research Division
   A-219 Chemistry
   U.S. Department of Commerce
   National Bureau of Standards
   Washington, D.C. 20234 - tel. (301) 921-3674

3. Quantity of special nuclear material (SNM) and enrichment: 450. mg of uranium containing approximately 58.5 milligrams of U-235 (enrichment ranging from 0.5% to 90.0%) 72 mg of uranium 233 2.25 mg of plutonium

4. Origin of SNM: Toll enriching contract UES/EU/4

5. Reason for import: Determination of uncertainty of analytical measurements of U and Pu isotopes; Framework: Nuclear Safeguards ESARDA IDA 78 Interlaboratory Measurement Evaluation Program.

6. Shipping container approved by USDOE: (see footnote)
   Interstate Commerce Commission: (see footnote)

7. Have the following requirements been met: (yes or no)

   a) Port Clearance: _____ b) IATA Clearance: _____ c) FAA Clearance: _____

   d) ICC Clearance: _____ e) U.S. Customs Clearance: _____

8. Shipping details:

   a) Name of carrier: _____
   b) Flight or ship number: _____
   c) Date of departure: JUNE 1980
   d) U.S. port of arrival: _____
   e) Date of arrival: _____
   f) Airway bill or bill of lading no.: _____
   g) Name of freight forwarder or shipping agent responsible for handling of shipment in the U.S.: Transnuclear, Inc. / Falls Church, Virginia
   h) Customs broker: Transnuclear, Inc. / White Plains, New York

Footnote: Shipping container: IAEA certificate of competent authority type B fissile radioactive material package. Design: certificate # USA/0002/B( )F. Packaging description: the packaging consists of a metal, drum-type birdcage which utilizes a sealed D.O.T. specification 2R metal inner containment vessel, centered and supported within a removable head type metal outer 10 gallon K (DOE 6M) drum by means of machined discs and rings of solid materials which provide thermal and impact protection. Packaging complies with the provisions of 49 CFR 178.104.

APPROVED BY UNITED STATES DEPARTMENT OF ENERGY

Michael O'Donnell, ONA/IA — 6/3/80

Name Title Date

SER-1

App. 6

NOTIFICATION OF MATERIAL TRANSFER

CONTRACT NO. _____

RETRANSFER NO. _____

OTHER REFERENCE _____

The special nuclear material described hereunder has been transferred from _____ (Transferor) to _____ (Transferee) under the provision of applicable agreements in effect with the Government of the United States of America and in accordance with the terms of the above referenced transfer documents.

SPECIFIED MATERIAL

Description, Markings, Type of Material, Form, Etc.	Total U (Nearest Gram)	U-235, U-233 or Pu* (Nearest Gram)	Enrichment Percent %	Other Materia Specify: State Unit:

Other pertinent information: (Use attachment if necessary)

*If any of this is produced material, please state.

This is to certify that the above material was shipped on behalf of:

_____ (Transferor)

DATE SHIPPED: _____

NAME: _____

TITLE: _____

DATE: _____

This is to certify that the above material was received on behalf of

_____ (Transferee)

DATE RECEIVED: _____

NAME: _____

TITLE: _____

DATE: _____

(See Instructions on Back of This Form)

CCR
BCMN
Steenweg op Retie
2440 Geel (B)

Code : WBCM

SPECIALE SPLIJTSTOFFEN of SPLIJTGRONDSTOFFEN
MATIERES FISSILES SPECIALES ou BRUTES

App. 7 — N° 000.11

№ 1483

Doc. **A1c**

(adresser les trois premiers exemplaires au service de gestion compable du B.C.M.N.)
(de drie eerste exemplaren aan de dienst boekhouding van splijtstoffen van het C.B.N.M. zenden.)

Date de la variation / Datum van verandering ...../...../.....
Local No/Lokaal Nr .....
Place/ Plaats .....
Zône: ..... Nom du déclarant/Naam v.d. verklarende .....

(réservé à la comptabilité)
Local 2 5
Date 6 11
P.M.P. 12

Aard v/d meting / Nature de la mesure ..... (1)
Mesure 13

DECLARATION DE / AANGIFTE VAN

- Reception/Ontvangst ☐
- Expedition/Verzending ☐
- Dechets/Afval ☐
- Pertes/Verliezen ☐
- Changement de catégorie / Verandering van categorie ☐
- Nouvelle mesure/Nieuwe meting ☐
- Autre/Andere ☐
- ..... ☐

Lid-staat v/d EG ⇄ Pays de la CE ☐
Staat buiten de EG ⇄ Pays hors CE ☐
Intern / Interne ☐

Type de variation
14 15

No LOT
BC.....

(Expédition/Verzending) Destination/Bestemming
.....
.....
.....
(2) ZBM ☐☐☐☐

(Réception/Ontvangst) Expéditeur/Afzender
.....
.....
.....
(2) ZBM ☐☐☐☐
- No lot expéditeur
- Lot Nr afzender
.....

ZBM Corresp.
16 17 18 19

Désignation du lot
20 21 22 23 24 25 26 27

DESCRIPTION DU LOT / PARTIJOMSCHRIJVING

Aantal artikelen / Nombre d'articles ☐☐☐☐

Nombre d'articles
28 29 30 31

Materiaalbeschrijving / Description des matières

(3) Forme/Vorm ..... ☐☐
(3) Nature du récipient / Aard v/d verpakkingen ..... ☐
(3) Etat des matières / Toestand v/d materialen .....
Composition chimique / Chemische samenstelling
.....

Description des matières
32 33 34 35

Matière / Stof

Element

- U. Nat./Nat. U ☐ N
- U Appauvri/Verarmd U ☐ D
- U. enrichi à ⩽20 % / Verrijkt U ⩽ 20 % ☐ L
- U. enrichi à > 20 % / Verrijkt U > 20 % ☐ H
- Plutonium ☐ P
- Thorium ☐ T
- Eau lourde / Zwaar water ☐

- Poids net / Netto gewicht ..... g.
- Poids l'élément / Gewicht v/h element ..... g.
- Poids d'U-235 / Gewicht U-235 ..... g.
- Poids d'U-233 / Gewicht U-233 ..... g.
- Enrichissemennt / Verrijking ..... %

Elément ☐ 36
Poids 37 45
Unité ☐ 46
Isotope ☐ 47
Poids 48 56
Unité ☐ 57
Engagement ☐ 58
Usage 59 60

Isotope / Isotoop

- U-235 ☐ G
- U-233 ☐ K
- U-235 + 233 ☐ J

- Usage de l'échantillon (4) / Gebruik v/h monster ☐
- Engagement/Verbintenis (5) ☐

Date d'envoi à la comptabilité
Datum van verzending aan de boekhouding
.....
Handtekening / Signature

Réception comptable
Date : .....
Visa : .....

Numéro de lot
BC .....

Exemplaire N 1 → Gestion comptable
Exemplaar — Boekhouding

App. 8

Form A_{2_b}

FISSILE MATERIALS

NOTE FOR TRANSMISSION

N° 1999

Commission of the European Communities

JOINT RESEARCH CENTRE

Geel Establishment

RESPONSIBLE :

ADDRESS : Central Bureau for Nuclear Measurements
Steenweg op Retie, 2440 Geel, Belgium
Tel. (014) 589421 - Telex 33589 EURAT B

MBA CODE : WBCM

To

ORGANIZATION :

RESPONSIBLE :

ADDRESS :

MBA CODE :

* Measurement (5)	Batch designation (8)	Number of items (9)	Material description * (10)	* Element (11)	Weight of element (12)	Unit (13)	* Isotope (14)	Weight of fissile isotopes (15)	Unit (16)	* Obligation (17)	DESCRIPTION OF SAMPLE ACTIVITY AND WEIGHT : Remarks

REMARKS

PRO FORMA Nr.....

TYPE OF PACKAGE AND CATEGORY

DATE OF DISPATCH	NAME AND SIGNATURE FOR DISPATCH	NAME AND SIGNATURE FOR RECEIPT	DATE OF RECEIPT
DATE OF ACCOUNTANCY			DATE OF ACCOUNTANCY

CBNM - B-2440 GEEL BELGIUM

copy n° 1 (dispatch)

* (Euratom) Regulation n° 3227/76 - Annex II

App. 9

COMMISSIE DER EUROPESE GEMEENSCHAPPEN
DIREKTORAAT – GENERAAL
Gemeenschappelijk Centrum voor Onderzoek

COMMISSION DES COMMUNAUTES EUROPEENNES
DIRECTION GENERALE
Centre Commun de Recherche

CENTRAAL BUREAU VOOR NUCLEAIRE METINGEN
BUREAU CENTRAL DE MESURES NUCLEAIRES
Steenweg naar Retie – B-2440 GEEL (België)
Tel. (014) 58.94.21
Telex : EURATOM GEEL B33589

VERZENDINGSNOTA
NOTE D'ENVOI

No :

Date :

PRO-FORMA

Voor rekening van : Pour compte de : Verzonden per : Expédié par :	Uw ref. : Vos Onze ref. : Nos
Reden van verzending : Motif de l'expédition :	

Aantal Nombre	Omschrijving / Désignation

BESTEMMELING :

Waarde :
Valeur :

Commission of the European Communities

JOINT RESEARCH CENTRE

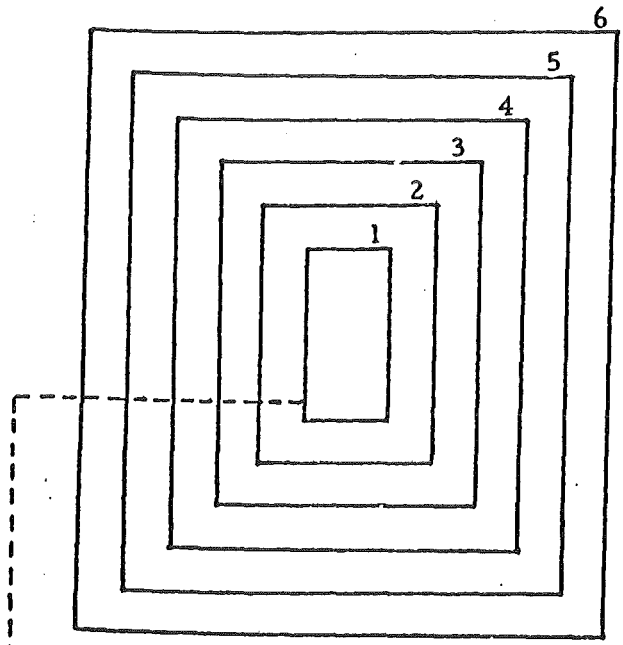
App. 10

Geel Establishment

Central Bureau for Nuclear Measurements
Steenweg op Retie, 2440 Geel, Belgium
Tel. (014) 589421 - Telex 33589 EURAT B

SCHEMATIC PACKING

Details for .....



6. _____

5. _____

4. _____

3. _____

2. _____

1. _____

When packed for shipment, the inside of this container (N°. ..... in schematic) was contaminated with radioactive material.
Therefore this container should be opened using standard techniques of your plant for the handling of contaminated items (in glove box, etc.).

By : .....

Date : .....

App. 11
(rear has been omitted)

LUCHTHAVEN VAN VERTREK EN GEVRAAGDE ROUTE | LUCHTHAVEN VAN BESTEMMING. | VLUCHT-DAG | VLUCHT-DAG | GERESERVEERD

1/ ROUTE EN BESTEMMING
NAAR | DOOR EERSTE VERVOERDER | NAAR | DOOR | NAAR | DOOR | NAAR | DOOR

2/ REKENINGNUMMER VAN DE BESTEMMELING | NAAM EN ADRES VAN DE BESTEMMELING

3/ REKENINGNUMMER VAN AFZENDER | NAAM EN ADRES VAN DE AFZENDER

4/ REKENINGNUMMER VAN DE AGENT VAN VERVOERDER | NAAM EN ADRES VAN DE AGENT VAN VERVOERDER

IATA CODE VAN AGENT

VERZENDINGSAANGIFTE

SABENA

MET DRUKLETTERS INVULLEN

Ondergetekende verklaart uitdrukkelijk dat de gegevens in deze verzendingsaangifte opgegeven met de waarheid stroken. Hierbij geeft de afzender, of zijn agent, opdracht aan Sabena volgens deze gegevens de luchtvrachtbrief op te stellen. Hij verklaart tevens kennis te hebben genomen der "Algemene Voorwaarden voor het Vervoer van Goederen" van de luchtvervoerder en aanvaardt deze volledig; zij zullen ook toegepast worden op alle verhandelingen en grondvervoer welke door Sabena uitgevoerd worden, vóór de luchtvrachtbrief opgemaakt is.
Indien het vervoer een eindbestemming of een tussenlanding medebrengt in een ander land dan het land van vertrek, kan dit vervoer onder de bepalingen vallen van de Conventie van Warschau dewelke in het algemeen de aansprakelijkheid van de vervoerders in geval van verlies of beschadiging van goederen beperkt.

Handtekening:

Datum:

5/ KOSTEN TE BETALEN DOOR Afzender | Bestemmeling | WAARDE AANGEGEVEN VOOR VERVOER | WAARDE AANGEGEVEN VOOR TOLDIENST | VERZEKERING - BEDRAG (Verzekering door bemiddeling van SABENA naar luid van paragraaf 8 op keerzijde: voorwaarden | ...% gewoon tarief | ...% tarief oorlog,enz...) | TERUGBETALING - BEDRAG

6/ NIET IN DEZE VAKKEN SCHRIJVEN

AANTAL COLLI RCP	WERKELIJK BRUTO-GEWICHT	Kg/lb.	RATE CLASS	COMMODITY ITEM NO.	CHARGEABLE WEIGHT	RATE	AARD EN HOEVEELHEID DER GOEDEREN (AFMETINGEN OF OMVANG INBEGREPEN)

BIJZONDERE INLICHTINGEN VOOR DE BEHANDELING (MERKEN, NUMMERS EN AARD DER VERPAKKING INBEGREPEN)

NIET IN DEZE VAKKEN SCHRIJVEN

PREPAID 7/ PREPAID WEIGHT CHARGE | V | PREPAID VALUATION CHARGE | C | TOTAL OTHER PREPAID CHARGES: DUE CARRIER | A | DUE AGENT | P | TOTAL PREPAID

R OTHER CHARGES (EXCEPT WEIGHT CHARGE AND VALUATION CHARGE) | FOR CARRIER'S USE ONLY AT DESTINATION

S | COLLECT CHARGES IN DESTINATION CURRENCY

T | Z COD AMOUNT | COD AMOUNT

COLLECT 8/ COLLECT WEIGHT CHARGE | V | COLLECT VALUATION CHARGE | C | TOTAL OTHER COLLECT CHARGES: DUE CARRIER | A | DUE AGENT | TOTAL CHARGES

9/ HANDLING INFORMATION | TOTAL COLLECT

SABENA
belgian world airlines

SHIPPER'S CERTIFCATION FOR RADIOACTIVE MATERIALS
Two completed and signed copies of this certification shall be handed to the carrier. App. 12
(USE BLOCK LETTERS)

WARNING : Failure to comply in all respects with the IATA Restricted Articles Regulations may be a breach of the applicable law, subject to legal penalties. This Certification shall in no circumstances be signed by an IATA Cargo Agent, consolidator or a forwarder.

This consignment is within the limitations prescribed for : (mark one)

☐ both passenger and cargo aircraft ☐ only cargo aircraft

NATURE AND QUANTITY OF CONTENT				PACKAGE			
RADIONUCLIDE	**FORM**		**ACTIVITY**		**CATEGORY**	**TRANSPORT INDEX**	**TYPE**
NAME OR SYMBOL OF PRINCIPAL RADIOACTIVE CONTENT	SPECIAL FORM or OTHER FORMS or LOW LEVEL SOLID or LOW SPECIFIC ACTIVITY	CHEMICAL FORM AND PHYSICAL STATE (GAS/ LIQUID/SOLID)	NUMBER OF CURIES, or MILLI-CURIES	NUMBER OF PACKAGES	I-WHITE or II-YELLOW or III-YELLOW LABEL	FOR YELLOW LABEL CATEGORIES ONLY	INDUSTRIAL or TYPE A. or TYPE B (U) or TYPE B(M)

COMPETENT AUTHORITY INDENTIFICATION MARK
(For Special Form, Package Design and Shipment)

ADDITIONAL INFORMATION REQUIRED FOR FISSILE MATERIALS ONLY

EXEMPTED FROM THE ADDITIONAL REQUIREMENTS FOR FISSILE MATERIALS SPECIFIED IN 8.1 OF PART 2 of the IATA RESTRICTED ARTICLES REGULATIONS ☐

NOT EXEMPTED : FISSILE CLASS I ☐ FISSILE CLASS II ☐ FISSILE CLAS III ☐

Additional certificates obtained by the Shipper when necessary :

Special Form Certificate(s) ☐ Government Approvals/Permits ☐
Type "B" Packaging certificate(s) ☐ Special Arrangements ☐
Certificate(s) for Fissile Material ☐

Special Handling Information

I hereby certify that the contents of this consignment are fully and accurately described above by Proper Shipping Name and are classified, packed, marked, labelled and in proper condition for carriage by air according to the current Edition of the IATA Restricted Articles Regulations and all applicable carrier and governmental regulations. I acknowledge that I may be liable for damages resulting from any misstatement or omission and I further agree that any air carrier involved in the shipment of this consignment may rely upon this Certification.

Name and full address of Shipper	Name and title of person signing Certification

Le texte français du présent document est tenu à la disposition de la clientèle dans les bureaux de la SABENA.

De nederlandse tekst van onderhavig document is ter beschikking van de kliënteel in de kantoren van de SABENA.

Date	Signature of the Shipper **(see WARNING above)**

Air Waybill N° *	Airport of Departure *	Airport of Destination *

*This box is optional for completion by issuing carrier

4936

JULY 77

Ⓑ C 665

EX 61

Raadpleeg de toelichting voor het invullen van het formulier

App. 13

AANGIFTE TEN UITVOER — EXEMPLAAR VOOR CIV — 3

2 Aantal bijgevoegde formulieren EXc 61	**3** Exporteur	**4** BTW - n^{r}	Kantoor N^{r} Datum	
	11 Geadresseerde (*)			
	21 Aangever (*) **DOUANE-AGENTSCHAP N M B S**		**22** Land van herkomst (*)	Datum van ontvangst
				25 Land van bestemming
	26 Plaats van de goederen	**27** Plaats van verzending	**28** Voorafgaande douaneregeling (*)	
			29 Aard van de transactie	
	32 Identiteit van het vervoermiddel	**33** Plaats van lading	**34** Leveringsvoorwaarden	
		36 Wijze van vervoer **SPOOR** / **37** Cont. ja/nee (1)	**38** Factuurprijs en munt	**39** N^{r} Betalingsdocument
1	**41** Merken, nummers, aantal en soort der colli; omschrijving der goederen		**42** Statistieknummer	**43** Brutogewicht / **44** Factuurprijs
	47 Uitvoervergunning	**48** Aanv. stat. eenheden	**49** Nettogewicht	**50** Statistische waarde
2	**41** Merken, nummers, aantal en soort der colli; omschrijving der goederen		**42** Statistieknummer	**43** Brutogewicht / **44** Factuurprijs
	47 Uitvoervergunning	**48** Aanv. stat. eenheden	**49** Nettogewicht	**50** Statistische waarde
	52 Kantoor van uitgang	**53** Bijbehorende stukken	**54** Identiteit van het vervoermiddel (vervolg)	
	Speciale vermeldingen			
			60 Ondergetekende _____ doet aangifte ten uitvoer voor rekening van de in vak 3 vermelde exporteur van de hierboven vermelde goederen Te _____, de _____ 19 __ Handtekening	**61** Douane-expediteur NMBS Inschrijvingsn^{r} : 1 Repertoriumn^{r} : _____

(1) Doorhalen wat niet van toepassing is

MASQUELIER 9313 - 9-78 (100.000)

App. 14
(rear has been omitted)

Anlage V
(zu § 12 abs. 1)

A. Einfuhranzeige/Bezugsanzeige [4])

[§ 12 Abs. 1 der Strahlenschutzverordnung [1])]

Durchschrift für den Einführer/Bezieher

(Der Einführer/Bezieher hat die Einfuhranzeige/Bezugsanzeige unaufgefordert bei der für die Abfertigung der Einfuhr/des Bezuges zuständigen Behörde vorzulegen. Die Einfuhranzeige/Bezugsanzeige ersetzt nicht die nach anderen Rechtsvorschriften – z. B. nach dem Außenwirtschaftsgesetz/dem Recht des innerdeutschen Wirtschaftsverkehrs – erforderlichen Genehmigungen, Erklärungen oder Kontrollpapiere.)

Auftragsnummer: _____

Hiermit zeige/n ich/wir

_____
(Name oder Firma des Einführers/Beziehers)

_____
(Straße, Postfach)

_____
(Postleitzahl, Wohnort oder Sitz der Firma)

vertreten bei der Abfertigung durch _____

_____
(Name und Anschrift des Vertreters: z. B. des Spediteurs)

die Einfuhr/den Bezug der folgenden radioaktiven Stoffe an:

1. Handelsübliche Warenbenennung: _____

2. Bezeichnung der Radionuklide nach Anlage IV Tabelle IV 1, IV 4 Spalten 2 und 3 der Strahlenschutzverordnung [1])

   _____

   _____

3. **Kernbrennstoffe** (§ 2 Abs. 1 Nr. 1 des Atomgesetzes [2])) in g [3])
   sonstige radioaktive Stoffe (§ 2 Abs. 1 Nr. 2 des Atomgesetzes [2]) in reziproken Sekunden (oder in Millicurie) oder g [3]); bei umschlossenen radioaktiven Stoffen Stückzahl; bei Bestrahlungsproben errechnete Aktivität:

   _____

   _____

4. Versendungsland: _____
5. Ausländischer Lieferant/DDR-Lieferant: _____
6. Ursprungsland: _____
7. Wert der Waren in DM/VE: _____ (ungefährer Gesamtbetrag)
8. Empfänger der Sendung: _____

   _____

   _____
   (Name oder Firma, Straße, Postfach, Postleitzahl, Wohnort oder Sitz der Firma)

Dem/Den unter Nr. 8 genannten Empfänger(n) der Sendung ist der Umgang mit den radioaktiven Stoffen der nach Nr. 2 und 3 einzuführenden Art und Menge/Aktivität nach den §§ 6, 7 oder 9 des Atomgesetzes [2]) § 3 Abs. 1 der Strahlenschutzverordnung [1]) genehmigt.

Bemerkungen: _____

_____

_____
(Ort, Datum)

_____
(Unterschrift und Firmenstempel des Einführers/Beziehers)

(subsequent pages have been omitted)

Physikalisch-Technische Bundesanstalt

Beförderungsgenehmigung

Nr. 1997 PTB 1979

nach § 4 des Atomgesetzes in der Fassung
vom 31.10.1976 (BGBl. I S. 3053)

Antragsteller: Commission of the European Communities, Geel Establishment, Central Bureau for Nuclear Measurements, 2440 Geel/Belgien

Antrag: von der Commission of the European Communities vom 12.9.1979 (Zeichen: WHW/GP L. 067949) mit Fernschreiben vom 11.10.1979

Beförderer: Zentralbüro für Kernmessungen (ZBKM), Geel/Belgien

Masse und Art des Kernbrennstoffes:

1) 2 Behälter (No. 46 und 53), enthaltend jeweils:

100 ml unbestrahlte HNO_3-Lösung, enthaltend 127 mg Plutonium (0,24 % Plutonium-239) mit ca. 3,3 mg Plutonium-239 + -241, mit einer Gesamtaktivität von < 400 mCi, und

2) 2 Behälter (No. 5 und 7), enthaltend jeweils:

100 ml unbestrahlte HNO_3-Lösung, enthaltend 121,9 mg Uran mit 121,5 mg Uran-233 und auf 0,0024 % im Uran abgereichertes Uran-235

Art der Beförderung:

in einem Transport mit 4 Behältern

Beförderungsstrecke:

vom Zentralbüro für Kernmessungen (ZBKM) in Geel/Belgien auf der Straße über Grenzübergang Aachen/Eynatten und über die Bundesautobahn zur Gesellschaft zur Wiederaufarbeitung von Kernbrennstoffen mbH in Eggenstein-Leopoldshafen 2

Beförderungsmittel:

VW-Bus mit dem polizeilichen Kennzeichen "EUR 0188" des ZBKM in Geel/Belgien

Beschreibung der Verpackung:

Typ B(U)-Verpackung, belgisches Versandstückmuster Typ DOT 6M (Gewicht: max. 36 kg), bestehend aus einem stählernen Innenbehälter (Wandstärke: 5 mm, Höhe 260 mm, Außendurchmesser 105 mm) und einem Außenbehälter mit den Abmessungen: Außendurchmesser ca. 322 mm, Höhe ca. 425 mm,

(subsequent pages have been omitted)

Physikalisch-Technische Bundesanstalt

Beförderungsgenehmigung

Nr. 1228 PTB 1974

Allgemeine Genehmigung nach § 4 des Atomgesetzes (AtG) vom 23.12.1959 (BGBl. I S. 814), zuletzt geändert durch Gesetz vom 23.6.1970 (BGBl. I S. 805)

Antragsteller: Bundesanstalt für Materialprüfung (BAM), Berlin

Antrag: vom 29.4.1974 (Zeichen: 6.3/Th/fi) mit Schreiben vom 13.6.1974 (Zeichen: 6.3/Th/k) und vom 26.6.1974 (Zeichen: 6.3/Th/fi)

Beförderer:

a) Austrian Airlines

b) British European Airways (BEA)

c) Deutsche Lufthansa

d) Pan American World Airways

e) SABENA Belgian World Airlines

Masse und Art des Kernbrennstoffes je Transport:

unbestrahlte oder bestrahlte Kernbrennstoffproben, enthaltend max. 15 g Plutonium (max. 15 g Plutonium-239) oder 15 g Uran-235 oder insgesamt 15 g Plutonium und Uran-235, in fester, nicht pyrophorer Form mit einer Gesamtaktivität von max. 20 Ci

Beförderungsstrecke:

1) von der Internationalen Atomenergie Organisation in Wien über Frankfurt oder München bzw.

2) von dem Zentralbüro für kerntechnische Messungen in Geel/Belgien über Frankfurt

zur BAM nach Berlin (Flughafen Tempelhof oder Tegel) oder in entgegengesetzter Richtung auf dem Luftwege einschließlich gegebenenfalls notwendiger Zwischentransporte und Zwischenlagerungen beim Umschlag.

Beförderungsmittel:

Flugzeuge der unter a) bis e) genannten Gesellschaften

Beschreibung der Verpackung:

Typ B-Verpackung, unilateraler Behälter (Typ: 6M-2, Volumen: 15 Gallonen, Gewicht: 72,5 kp), bestehend aus Außenbehälter (Stahltrommel) und Innenbehälter, gemäß dem

- 2 -

App. 17

(subsequent pages have been omitted)

Der Senator für Wirtschaft

BERLIN

Senator für Wirtschaft, 1 Berlin 62, Martin-Luther-Straße 105

An die
Bundesanstalt für
Materialprüfung (BAM)
1 B e r l i n 45
Unter den Eichen 87

BAM Tgb.-Nr.: Anl.:
Eing.: 22. JULI 1974

GeschZ.: (Angabe bei Antwort erbeten)
IV G 1 Vd/Wt

Zimmer: 107

Fernruf:
Direktwahl 783 8354
über Zentrale 7831 } Apparat wie oben
im Innendienst 95 }

Berlin, den 18. 7. 74

Allgemeine Beförderungsgenehmigung

Genehmigung nach § 4 des Gesetzes über die friedliche Verwendung der Kernenergie und den Schutz gegen ihre Gefahren (Atomgesetz) vom 23.12.1959 (BGBl. 1959 I S. 814/GVBl. 1961 S. 1493), zuletzt geändert durch das Bundes-Immissionsschutzgesetz vom 15.März 1974 (BGBl. I S. 721/GVBl. 1974 S.648 in Verbindung mit den BK/O (61) 8 (Art. 2 und 3) vom 12.7.1961 (GVBl. 1961 S. 1523) BK/O (67) 6 vom 31.5.1967 (GVBl. 1967 S. 1008), BK/O (72) 8 vom 22.6.1972 (GVBl. 1972 S. 1194) BK/O (73) 6 vom 25.9.1973 (GVBl. 1973 S. 1740) der Alliierten Kommandatura Berlin.

Antragsteller: Bundesanstalt für Materialprüfung (BAM), 1 Berlin 45, Unter den Eichen 87

Antrag: Schreiben der BAM vom 22.3.1974 und 13.6.1974 Zeichen: 6.3/Th/K

Beförderer:

Luftweg

BEA, Pan American

Landweg

Bundesanstalt für Materialprüfung, Schenker & Co.

Masse und Art des Kernbrennstoffes:

15 g Pu in fester Form (max. 20 Ci),
Uran in fester Form mit einem Gehalt von nicht mehr als 15 g U-235 (max. 20 Ci)

Art der Beförderung: in einem Behälter

Beförderungsstrecke: Durch einen der Luftkorridore zum Flughafen Tempelhof oder Tegel einschließlich ggf. notwendiger Zwischenlagerung beim Umschlag. Vom Flughafen Tempelhof oder Tegel zur Bundesanstalt für Materialprüfung, 1 Berlin 45, Unter den Eichen 87

Page

List of Tables

List of Photographs