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Determination of the Isotope U-235 in Uranium Hexafluoride by Gas Mass Spectrometry: Results of an Interlaboratory Experiment Performed in 1975

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Determination of the Isotope U–235 in Uranium Hexafluoride by Gas Mass Spectrometry: Results of an Interlaboratory Experiment Performed in 1975

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Determination of the Isotope U-235 in Uranium Hexafluoride by Gas Mass Spectrometry: Results of an Interlaboratory Experiment Performed in 1975

Abstract

Samples of uranium hexafluoride with a U-235 content of about 0.4, 0.7 and 3 % were measured with 10 gas mass spectrometers in 8 European laboratories. Identical reference materials were used with U-235 abundances deviating less than 6 % from those of the samples and known with an accuracy better than ± 0.15 %.

By statistical evaluation of the data, errors of about 0.1 % were calculated for the determination of the ratio of ratios $(\text{U-235}:\text{U-238})_{\text{sample}} / (\text{U-235}:\text{U-238})_{\text{reference}}$ with increasing tendency for U-235 abundances below the natural range.

The experiment was performed by Messrs. DORNIER-SYSTEM GmbH., Friedrichshafen, on behalf of the Safeguards Project of the Federal Republic of Germany.

Gasmassenspektrometrische Bestimmung des Isotops U–235 in Uranhexafluorid: Ergebnisse eines 1975 durchgeführten Vergleichsexperiments

Zusammenfassung

Uranhexafluoridproben mit einem U-235 Gehalt von etwa 0.4, 0.7 und 3 % wurden mit 10 Gasmassenspektrometern in 8 europäischen Laboratorien gemessen. Es wurden identische Referenzproben verwendet, deren U-235 Häufigkeit jeweils weniger als 6 % von denen der Meßprobe abwich und mit einer Genauigkeit von besser als ± 0.15 % bekannt war.

Die statistische Auswertung der Daten ergab Fehler von etwa 0.1 % für die Bestimmung des Verhältnisses $(\text{U-235}: \text{U-238})_{\text{Meßprobe}} / (\text{U-235}: \text{U-238})_{\text{Referenzprobe}}$ mit ansteigender Tendenz für U-235 Häufigkeiten unterhalb des natürlichen Bereiches.

Das Experiment wurde im Auftrag des Projektes Spaltstoffflußkontrolle der Bundesrepublik Deutschland durch die Firma DORNIER-SYSTEM GmbH., Friedrichshafen, durchgeführt.

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Preface

From the point of view of nuclear safeguards, uranium enrichment plants for the industrial scale production of uranium enriched to about 3 % U-235, play an important role. One of the major safeguards measures in such plants is the establishment of material balances which calls for the precise knowledge of the abundance of the U-235 content in the uranium hexafluoride gas stream at the inlet and outlet of the facility. This U-235 content is determined most suitably with special UF_6 gas mass spectrometers and by the use of reference materials of similar and precisely known composition.

For this reason, it was proposed in 1974 by the Nuclear Safeguards Project of the Federal Republic of Germany that as a continuance of the analytical comparison experiments /1-3/ organized by the Project in the preceding years it should be investigated within the framework of an interlab test which errors have to be expected for such measurements in routine operation and whether these errors are mainly caused by the measurement procedure proper or by the accuracy of the reference materials (standards) available.

The experiment was performed by Messrs. Dornier System GmbH., Friedrichshafen, on behalf of the Nuclear Safeguards Project. In 1974 the sample materials were prepared and distributed among the participating laboratories in which the measurements were carried out in 1975.

After statistical evaluation of the data obtained and their compilation in a "preliminary report" the results were discussed in detail at a final meeting of participants held at Messrs. Dornier System GmbH in Friedrichshafen on May 12/13, 1976. The judgement reached at this meeting forms the basis of the final report, above all of the summary of results given in Chapter 6.

As can be seen from the attached list of participants eight laboratories in all participated in the experiment. Since the Institut für Kernverfahrenstechnik (Institute of Nuclear Engineering) of Gesellschaft für Kernforschung mbH. carried out the measurements at three different mass spectrometers, a total of 10 groups of measured values were available for evaluation; they have been entered under separate codes.

The Project and the authors wish to express their cordial thanks to all participating laboratories for their constructive cooperation both in the performance of the measurements and in the interpretation of the results obtained, thus ensuring successful completion of the experiment. Our thanks are expecially due to the authors of written contributions to this report and to Dr. P. De Bièvre and his coworkers of the EURATOM Central Bureau of Nuclear Measurements, Geel, Belgium, for comprehensive work on the precise determination of the isotopic composition of the sample materials used which will henceforth be available to each laboratory as a reference material in the form of UF_6. This is a significant step for practical work, which allows to reduce the differences in results obtained by the individual laboratories.

Dipak Gupta
Head of the Nuclear Safeguards Project

List of Participants

BAM
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1. Introduction

The layout and objectives of this experiment rely on the concept that in nuclear safeguards the U-235 content in streams of uranium hexafluoride is determined most suitably in the gaseous state by special UF_6 mass spectrometers.

In such instruments, the gas to be measured preferably enters the electron bombardment ion source as a molecular beam and the measured variable proportional to the U-235/U-238 isotope ratio is determined by a double collector system. The proportionality factor is obtained by alternate measurement using a reference material of similar and precisely known composition. This method has to be clearly distinguished from the less accurate thermionic mass spectrometry which requires preceding chemical conversion of the UF_6 implying additional sources of error. 1)

In conformity with the composition of the three gas streams occurring in enrichment plants, UF_6 samples from the "tails", "feed", and "product" gas streams with a U-235 abundance of about 0.4, 0.7 and 3.0 %, respectively, were investigated. While the samples having a U-235 content in the natural range were measured by all participating laboratories, only 5 and 6 laboratories, respectively, participated in the measurement of the enriched and depleted materials.2)

It did not appear meaningful to include into the investigation discrepancies between the results of laboratories which may occur by use of different reference materials, since this difficulty can be avoided, in principle, by the use if identical reference materials at all laboratories subjected to nuclear safeguards. Also, the measurement and comparison with laboratory owned reference materials of different origins would have made it impossible to recognize whether still other causes have to be assumed for the "interlab deviation", i.e. the error component characterizing the scattering of laboratory mean values.

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1) In this context, reference is made to the comparison experiment for the determination of the U-235 content in UF_6, performed recently by the New Brunswick Laboratory, Energy Research and Development Administration, New Brunswick, New Jersey, USA. In this experiment the laboratories were free to decide upon the method they wanted to apply [4].

2) Some laboratories feared a negative effect upon the measurement conditions by the introduction of UF_6 samples of such different isotopic compositions into one single mass spectrometer.

For this reason and since the suppliers of UF_6 mass spectrometers specify that the isotopic abundance of U-235 in the reference standard must lie within ± 6 % (relative) of the U-235 abundance in the sample to be measured in order to ensure the accuracy of measurement and reproducibility, a sample pair of similar isotopic composition was prepared for each of the three interesting U-235 concentration ranges. One sample each of this pair was considered as the "test material ", i.e. the sample to be measured, the other as the "reference material ".

The single measurement of such a sample pair consisting of one single introduction of the test material and of the reference material was termed a "measurement cycle," the immediate repetition of six measurement cycles was called "measurement period". To be able to observe also any long-term effects, the laboratories were requested to perform six measurement periods in total at roughly monthly intervals. This schedule was not strictly observed by all laboratories.1)

With a view of the objectives pursued by the experiment the laboratories were asked to carry out the measurements under the usual routine conditions, avoiding additional expenditure in terms of measurement technology and to submit brief statements about the type of mass spectrometer used and about the procedure of the measurement. Unfortunately, this information cannot be presented in detail here since this would largely imply disclosure of the laboratory code. Generally, it can be said that – as far as information is available – mass spectrometers with electron bombardment ion sources were used in all cases, which had been well adapted to the special requirements for UF_6 measurements (molecular beam, cooling finger, etc.) and had been provided with double collector systems for the detection of ions. In some instruments the whole measurements process was controlled by a computer.

With the exception of laboratory 4 the measured data of the ratio were not corrected by the laboratories to take into account possible memory effects, since this is not done anyhow in routine operation by the majority of participants. In all cases the mass spectrometer inlet system was carefully pumped when the samples were exchanged and sometimes scavenged in addition with the new sample material, above all in cases where samples of a different U-235 concentration range were introduced 2). In some of the sample ampules extraordinarily high amounts of HF were found.

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1) Cf. Annexes I and VII.

2) Cf. also Annex VIII

According to the measurement procedure applied, two problems must be distinguished as regards the objectives of the experiment:

i) Determination of the errors to be expected in routine operation from the mass spectrometric assay of the ratio of ratios
a = $(\text{U-235: U-238})_{\text{test material}}/(\text{U-235: U-238})_{\text{reference material}}$,
which defines the difference between the unknown U-235/U-238 ratio of the sample to be measured ("test material") and the (within given limits) known U-235/U-238 ratio of the reference material.

ii) Comparison of errors in determining the ratio a with the uncertainty concerning the knowledge of the exact U-235 abundance in the reference material, which is required to calculate the U-235 abundance in the sample to be measured.

Interlaboratory comparison proper related to the first complex of problems, i.e., the estimate of the total error to be expected in the measurement of the ratio a and, as far as possible, its subdivision into single components.

Considering the second objective of the experiment mentioned above, it was of interest to determine also the complete isotopic composition of UF_6 samples used in the test. These measurements were performed for all 6 sample materials at Capenhurst Works of British Nuclear Fuels Ltd. (BNFL), Great Britain, and at the EURATOM Central Bureau of Nuclear Measurements (CBNM), Geel, Belgium. Besides, the U-235 abundance in some measured samples was determined by the Institute of Nuclear Engineering (IKVT) of Gesellschaft für Kernforschung mbH (GfK), Karlsruhe, and by URENCO Nederland Operations B.V. (UOBV), Almelo, Netherlands.1).

However, the objectives of the experiment did not include the evaluation of the isotopic compositions of the UF_6 samples on the basis of the measurements of all the participating laboratories with a view to the future use of these samples as reference materials. The comprehensive work done by CBNM in this respect2) is an independent contribution rendered by CBNM as a service to all laboratories in this field which was initiated by this experiment.

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1) These investigations have been described in Annexes V to VII.

2) Cf. Annex V

2. Preparation of the Sample Material

The sample material (UF_6) used in the interlab test came from the Almelo enrichment plant and was delivered with the three levels of U-235 abundance already mentioned:

about 0.4 % ("tails")
about 0.7 % ("feed")
about 3 % ("product").

Prior to further use the total amount of a U-235 level of abundance was filled into a UF_6 purification facility by resublimation. Subsequently, the material was cleaned by rectification in the packed column of the facility.

After rectification the purified and homogenized UF_6 was transferred directly from the liquid phase of the column into the sample ampoules as a condensate.

To avoid recontamination and falsification, respectively, of the U-235 abundance during the filling process, caused by possible contaminations present in the fork-shaped manifold between the facility and the ampoules, this manifold was scavenged with UF_6 having the concentration of the U-235 to be filled in. The scavenging gas was discarded.

In order to prepare adequate reference samples for this experiment[1)], precalculated amounts of sample material of different U-235 abundances had to be mixed. The reference material so prepared was again purified, homogenized and transferred from the liquid phase into ampoules as a condensate, as described above.

To obtain representative results from the measurements of ratios supplied by the individual laboratories and to be able to draw conclusions, it had to be ensured that the U-235 abundance is the same in all sample ampoules distributed among the laboratories. So, to minotor the U-235 abundance the first filled ampoule of each batch was measured by mass spectrometry and compared with the last, although not delivered as a sample ampoule. Within the measurement accuracy of the mass spectrometer used (VARIAN, CH5-UF) significant deviations were not found in these measurements.

[1)] Cf. Chapter 1

The sample material (UF_6) was distributed among the participating laboratories in the sample ampoules shown in Fig. 2-1. These sample ampoules, completely made of stainless steel, consist of a sample cylinder (50 ccm), a welded spring bellows valve with an elastomer gasket and an NW-10 mini-flange [1].

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[1] Supplier of the sample cylinders and valves:
NUPRO COMPANY, 15635 Savanac Road, Cleveland, Ohio 44110, USA.
The spring bellows valve can be disassembled for purification.
By replacement of the elastomer gasket by a metallic cone it can be used as a metallic joint as well.

Fig. 2—1: Sample ampoule with cylinder and mini - flange

3. Determination of the Isotopic Composition of the Sample Material

As already mentioned in the Introduction, the complete isotopic composition of the six sample materials used in the test was determined at Capenhurst Works of British Nuclear Fuels Ltd. (BNFL) and at the Central Bureau of Nuclear Measurements (CBNM), Geel, Belgium. The results obtained have been compiled in Table 3-1.

BNFL carried out these measurements incurring expenditure as usual for a commercial analysis performed on behalf of a third party and indicates error bars of about ± 0.15 %. They relate to a 95 % confidence level and include the error of the reference standards used (see Annex VI).

On account of the particular objective pursued by CBNM of making suitable part of the UF_6 samples for use as reference materials, considerable expenditure in terms of measurement technology was spent on these investigations (see Annex V). The error bars for the values given in Table 3-1 were given as maximum errors of about ± 0.15 %.

The difference of the U-235 abundances determined by the BNFL and CBNM laboratories is calculated to be 0.13 % at the maximum (test material "feed"). In all cases the measured value for the U-235 isotope determined by one of the laboratories lies within the error bar stated by the other laboratory.

The data contained in the footnotes 2 and 3 of Table 3-1 show the results obtained at the laboratory of UOBV while assaying the U-235 content using reference materials which had been received earlier from CBNM (see Annex VII). They are in excellent agreement with the values determined by CBNM.

Also footnote 4 shows the result of the determination of the U-235 content of the test material "feed" performed by the GfK Institute of Nuclear Engineering (IKVT). The reference material used was a working standard prepared at the Institute by mixing an NBS isotope standard with natural uranium supplied by Messrs. Merck [10] (cf. also Annex VIII). This value ranges between the results obtained by BNFL and CBNM.

Table 3–1: Results of Isotopic Composition Analyses of the Sample Materials

1		2	3	4	5
Sample		Laboratory	Isotopic Abundances[1] [at. %]		
			U-234	U-235	U-238
"Tails"	Test[2] Material	BNFL	0.0030 ± .0003	0.4379 ± .0007	99.559
		CBNM	0.0025 ± .0002	0.4383 ± .0007	99.5592 ± .0009
	Reference Material	BNFL	0.0030 ± .0003	0.4611 ± .0007	99.536
		CBNM	0.0027 ± .0002	0.4608 ± .0007	99.5365 ± .0009
"Feed"	Test[3] [4] Material	BNFL	0.0045 ± .0005	0.7192 ± .001	99.277
		CBNM	0.0053 ± .0003	0.7201 ± .0011	99.2746 ± .0014
	Reference Material	BNFL	0.0045 ± .0005	0.6838 ± .001	99.312
		CBNM	0.0050 ± .0003	0.6840 ± .0011	99.3110 ± .0014
"Product"	Test Material	BNFL	0.032 ± .003	3.273 ± .005	96.695
		CBNM	0.0327 ± .0007	3.2714 ± .0049	96.6959 ± .0056
	Reference Material	BNFL	0.030 ± .003	3.086 ± .005	96.884
		CBNM	0.0307 ± .0007	3.0841 ± .0049	96.8852 ± .0056

1) In all cases, the U-236 isotopic abundance was reported to be below the detection limits (BNFL: < 0.001 %; CBNM: < 0.0001 %)

2) UOBV, Almelo, U-235 abundance for "tails" test material determined using CBNM standard: 0.4383 ± 0.001 [at. %] (Cf. Annex VII)

3) UOBV, Almelo, U-235 abundance for "feed" test material determined using CBNM standard: 0.7203 ± 0.001 [at. %] (Cf. Annex VII)

4) GfK, IKVT, Karlsruhe, U-235 abundance for "feed" test material determined using NBS-MERCK standard: 0.7194 ± 0.001 [at. %] ([10], Cf. also Annex VIII)

4. Determination of the Ratio of Ratios $a = (\text{U-235: U-238})_{\text{test material}} / (\text{U-235: U-238})_{\text{reference material}}$

The original data of measurements submitted by the participating laboratories have been compiled in Annex I. They served as a basis for the calculation of the ratio of ratios

$$a = \frac{(\text{U-235:U-238})_{\text{test material}}}{(\text{U-235:U-238})_{\text{reference material}}}$$

from which all further evaluations were derived and which have been indicated in Annex II for the different sample materials ("tails"; "feed" and "product").

In the following Tables 4-1 to 4-3 the mean values per period of measurement calculated from these data have been compiled together with the standard deviation (SD) of the individual measurement cycle and its relative value (RSD) calculated for each period of measurement[1)].

A comparative review of the mean values of the individual periods yields in some cases the occurrence of "outliers". For example, the value of 0.94956 determined by laboratory 7 from tails measurements in the first period of measurement is distinctly higher than all subsequent evaluations this laboratory had performed with this sample material. The application to this data group of the criterion for outliers proposed by Dixon [5, 6] allows to reject this value with an error probability of less than 2 %. Nevertheless, both in this and in similar other cases all the mean values of a period were taken into account in the following evaluation of results so as to comply with practical conditions: Generally, the required comparison data are not available in routine operation to identify a mean value of a period as an "outlier". Besides, it should be mentioned that in the example quoted above that measured value of the group would be rejected which comes closest to the data determined by the other laboratories.

As to data obtained from this experiment, the scattering of values measured during a cycle (columns 3 and 4 of Tables 4-1 through 4-3) does not even allow a conclusion to be drawn about the quality of the related mean value of the period: In all three cases in which the relative standard deviation of the individual value measured in a cycle is 1 ^{o}/oo or more[2)], the corresponding mean value of the period lies within the data group belonging to the laboratory.

[1)] For a definition and calculation of the terms used here and in the following chapters we refer to Annexes III and IV.

[2)] Tails, lab. 7, 3rd period; feed and product each, lab. 2, 4th period.

Columns 5 of Tables 4-1 to 4-3 show in addition the mean values of a laboratory in order to illustrate the transition to the laboratory specific values represented in Tables 4-4 to 4-6:

Besides the mean values of laboratories repeated in column 2, columns 3 and 4 state the standard deviations of the individual value measured in a cycle and their relative values, respectively, determined on the basis of all measurements performed by a laboratory. Columns 5 and 6 exhibit the respective data for the period of measurement [1]. On the bottom of each table the total mean value ("grand mean") calculated from the laboratory mean values is indicated together with its standard deviation and the relative value of this standard deviation.

The relative deviations of the laboratory means from this grand mean have been represented in Figs. 4-1 to 4-3. The error bars denote the relative standard deviation of the laboratory means for which a comparative survey has been included in Fig. 4-4 [2]. In addition, in Figs. 4-1 to 4-3 the values from the determination of the ratio a given in Annex V by CBNM have been entered [3] with an error bar of ± 0.5 ‰ as indicated by CBNM (maximum error).

It strikes that almost all the laboratory mean values are inferior to the CBNM data. In the case of the feed and product measurements (Figs. 4-2 and 4-3) this could be explained by memory effects in the mass spectrometers since they cause too low a measured value for ratios of a greater than 1.0. It is very improbable that the CBNM data themselves are influenced by the memory effect because in the determination of U-235 abundances in the samples this laboratory did not even obtain significant differences when the ratio a was increased to about 1.5 by use of reference materials of greatly differing isotopic compositions (see Annex V, Chapter 6). Also UOBV, in their determination of U-235 abundance, observed this large degree of independence of results of the ratio a in feed and tails samples, even up to a value of about 2.2 (see Annex VII).

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[1] These standard deviations and their relative values were calculated from estimated values obtained by means of a variance analysis for the variances of these two error components (see Chapter 4 and Annex III, formulae 15 and 16).

[2] For definition see Annex III, formula 8.

[3] The mean values obtained by the two first measurement methods given in Chapter 4, Fig. V-3 were used.

In the case of tails measurements (Fig. 4-1) scattering of laboratory mean values cannot simply be explained by the memory effect. Since this ratio a has a value lower than 1.0, the memory effect would yield too high measured values, which is not confirmed, since also in this case the CBNM value, with one exception, is maximum. The higher scattering of laboratory mean values as compared to the feed and product measurements suggests that in the measurement of the depleted tails samples other sources of error - e.g., calibration errors of the measurement bridges - are in the foreground.

Table 4–1: Period Means for "Tails" Measurements and Related Data *)

1 Lab. Code	2 Period Mean $Y_{ij.}$	3 SD of Cycle $\pm\ \sigma\ (i,j)$	4 RSD of Cycle $\pm\ s\ (i,j)$ [$^{o}/oo$]	5 Lab. Mean $Y_{i..}$
4	0.95022	0.00015	0.15	0.95006
	0.95018	0.00017	0.18	
	0.94983	0.00008	0.09	
	0.94982	0.00023	0.24	
	0.95028	0.00012	0.12	
	0.95003	0.00023	0.24	
6	0.95088	0.00013	0.14	0.95082
	0.95081	0.00014	0.15	
	0.95078	0.00019	0.20	
	0.95085	0.00010	0.11	
	0.95080	0.00013	0.14	
	0.95080	0.00007	0.08	
7	0.94956	0.00036	0.38	0.94690
	0.94681	0.00081	0.86	
	0.94611	0.00099	1.04	
	0.94613	0.00048	0.51	
	0.94592	0.00048	0.50	
	0.94688	0.00055	0.58	
8	0.95282	0.00075	0.79	0.95155
	0.95118	0.00009	0.09	
	0.95171	0.00032	0.34	
	0.95125	0.00026	0.28	
	0.95123	0.00032	0.34	
	0.95109	0.00026	0.28	
9	0.95028	0.00031	0.33	0.95052
	0.95039	0.00019	0.20	
	0.95052	0.00083	0.87	
	0.95055	0.00033	0.34	
	0.95073	0.00013	0.14	
	0.95063	0.00009	0.09	
10	0.95021	0.00024	0.25	0.95003
	0.94866	0.00067	0.71	
	0.95091	0.00042	0.44	
	0.95019	0.00014	0.15	
	0.94973	0.00037	0.39	
	0.95050	0.00017	0.18	

*) The "EINS" program was used to calculate these data (Annex IV)

Table 4–2: Period Means for "Feed" Measurements and Related Data *)

1 Lab. Code	2 Period Mean $Y_{ij.}$	3 SD of Cycle $\pm\ \sigma$ (i,j)	4 RSD of Cycle $\pm$ s (i,j) [^{0}/oo]	5 Lab. Mean $Y_{i..}$
1	1.05278	0.00012	0.12	1.05291
	1.05281	0.00071	0.68	
	1.05300	0.00010	0.10	
	1.05299	0.00013	0.13	
	1.05300	0.00018	0.17	
	1.05288	0.00016	0.15	
2	1.05219	0.00047	0.44	1.05180
	1.05167	0.00016	0.15	
	1.05142	0.00014	0.14	
	1.05176	0.00105	1.00	
	1.05152	0.00073	0.69	
	1.05221	0.00095	0.90	
3	1.05252	0.00033	0.31	1.05260
	1.05269	0.00020	0.19	
	1.05293	0.00012	0.11	
	1.05253	0.00038	0.36	
	1.05240	0.00047	0.45	
	1.05252	0.00030	0.28	
4	1.05297	0.00018	0.17	1.05303
	1.05333	0.00020	0.19	
	1.05308	0.00015	0.14	
	1.05305	0.00010	0.10	
	1.05273	0.00010	0.10	
	1.05302	0.00015	0.14	
5	1.05271	0.00007	0.07	1.05275
	1.05274	0.00015	0.14	
	1.05281	0.00015	0.15	
	1.05275	0.00005	0.04	
	1.05276	0.00007	0.06	
	1.05274	0.00014	0.13	
6	1.05324	0.00007	0.07	1.05319
	1.05323	0.00011	0.11	
	1.05325	0.00003	0.03	
	1.05317	0.00026	0.25	
	1.05315	0.00008	0.08	
	1.05313	0.00006	0.06	

(continued)

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*) The "EINS" program was used to calculate these data (Annex IV)

(continued)

1 Lab. Code	2 Period Mean $Y_{ij.}$	3 SD of Cycle $\pm\ \sigma$ (i,j)	4 RSD of Cycle $\pm$ s (i,j) [$^{0}/oo$]	5 Lab. Mean $Y_{i..}$
7	1.05226	0.00031	0.29	1.05202
	1.05229	0.00034	0.32	
	1.05194	0.00050	0.48	
	1.05156	0.00044	0.42	
	1.05178	0.00074	0.70	
	1.05230	0.00015	0.14	
8	1.05098	0.00017	0.16	1.05197
	1.05199	0.00014	0.13	
	1.05236	0.00067	0.64	
	1.05187	0.00035	0.33	
	1.05236	0.00020	0.19	
	1.05228	0.00018	0.17	
9	1.05249	0.00029	0.27	1.05220
	1.05194	0.00044	0.42	
	1.05226	0.00050	0.48	
	1.05210	0.00008	0.07	
	1.05224	0.00005	0.05	
	1.05220	0.00009	0.08	
10	1.05218	0.00037	0.35	1.05298
	1.05332	0.00060	0.57	
	1.05460	0.00101	0.95	
	1.05248	0.00013	0.13	
	1.05268	0.00033	0.31	
	1.05260	0.00024	0.33	

Table 4–3: Period Means of "Product" Measurements and Related Data *)

1	2	3	4	5
Lab. Code	Period Mean	SD of Cycle	RSD of Cycle	Lab. Mean
	$Y_{ij.}$	$\pm\ \sigma\ (i,j)$	$\pm$ s (i,j) [$^{o}/oo$]	$Y_{i..}$
2	1.06110	0.00066	0.62	1.06258
	1.06257	0.00045	0.42	
	1.06307	0.00037	0.35	
	1.06250	0.00116	1.09	
	1.06345	0.00065	0.61	
	1.06277	0.00030	0.28	
4	1.06207	0.00010	0.10	1.06236
	1.06237	0.00016	0.15	
	1.06270	0.00011	0.10	
	1.06213	0.00027	0.26	
	1.06240	0.00021	0.20	
	1.06252	0.00010	0.09	
6	1.06294	0.00015	0.14	1.06286
	1.06283	0.00007	0.06	
	1.06287	0.00014	0.13	
	1.06292	0.00006	0.05	
	1.06275	0.00008	0.08	
	1.06287	0.00007	0.07	
8	1.06244	0.00006	0.05	1.06222
	1.06229	0.00020	0.19	
	1.06215	0.00028	0.27	
	1.06233	0.00039	0.36	
	1.06284	0.00036	0.34	
	1.06227	0.00047	0.44	
9	1.06259	0.00014	0.13	1.06259
	1.06250	0.00009	0.08	
	1.06256	0.00014	0.13	
	1.06267	0.00012	0.12	
	1.06254	0.00018	0.17	
	1.06265	0.00019	0.18	

*) The "EINS" program was used to calculate these data (Annex IV)

Table 4–4: Laboratory Means for "Tails" Measurements and Related Data *)

1 Lab. Code	2 Lab. Mean $Y_{i..}$	3 SD of Cycle $\pm\ \sigma_C$	4 RSD of Cycle $\pm\ S_C$ [^{o}/oo]	5 SD of Period $\pm\ \sigma_P$	6 RSD of Period $\pm\ S_P$ [^{o}/oo]
4	0.95006	0.00017	0.18	0.00019	0.20
6	0.95082	0.00013	0.14	not significant	
7	0.94690	0.00065	0.69	0.00133	1.41
8	0.95155	0.00039	0.41	0.00064	0.67
9	0.95052	0.00040	0.42	not siginificant	
10	0.95003	0.00038	0.40	0.00076	0.80

Grand Mean: $Y_{...} = 0.94998 \pm 0.00066$ (± 0.69 ^{o}/oo)

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*) See Annex IV, "ZWEI" program for calculation of the data in the table and Annex III, formulae 9 - 11 for calculation of the data in the bottom line.

Table 4–5: Laboratory Means for "Feed" Measurements and Related Data *)

1	2	3	4	5	6
Lab. Code	Lab. Mean $Y_{i..}$	SD of Cycle $\pm \sigma_C$	RSD of Cycle $\pm S_C$ [^{o}/oo]	SD of Period $\pm \sigma_P$	RSD of Period $\pm S_P$ [^{o}/oo]
1	1.05291	0.00032	0.30	not significant	
2	1.05180	0.00068	0.65	0.00020	0.18
3	1.05260	0.00032	0.30	0.00013	0.12
4	1.05303	0.00015	0.14	0.00018	0.17
5	1.05275	0.00011	0.11	not significant	
6	1.05319	0.00013	0.12	0.00001	0.01
7	1.05202	0.00045	0.43	0.00025	0.24
8	1.05197	0.00034	0.32	0.00051	0.49
9	1.05220	0.00030	0.29	0.00013	0.13
10	1.05298	0.00054	0.51	0.00085	0.81

Grand Mean: $Y_{\cdots} = 1.05255 \pm 0.00016$ ($\pm$ 0.15 ^{o}/oo)

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*) See Annex IV, "ZWEI" program for calculation of the data in the table and Annex III, formulae 9 - 11 for calculation of the data in the bottom line.

Table 4–6: Laboratory Means for "Product" Measurements and Related Data *)

1 Lab. Code	2 Lab. Mean $Y_{i..}$	3 SD of Cycle $\pm\ \sigma_C$	4 RSD of Cycle $\pm S_C$ [^{o}/oo]	5 SD of Period $\pm\ \sigma_P$	6 RSD of Period $\pm S_P$ [^{o}/oo]
2	1.06258	0.0006	0.62	0.00076	0.71
4	1.06236	0.00017	0.16	0.00023	0.21
6	1.06286	0.00010	0.09	0.00006	0.05
8	1.06222	0.00032	0.30	0.00016	0.15
9	1.06259	0.00015	0.14	0.00009	0.03

Grand Mean: $Y_{\cdots} = 1.06252 \pm 0.00011$ ($\pm$ 0.10 ^{o}/oo)

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*) See Annex IV, "ZWEI" program for calculation of the data in the table and Annex III, formulae 9 - 11 for calculation of the data in the bottom line.

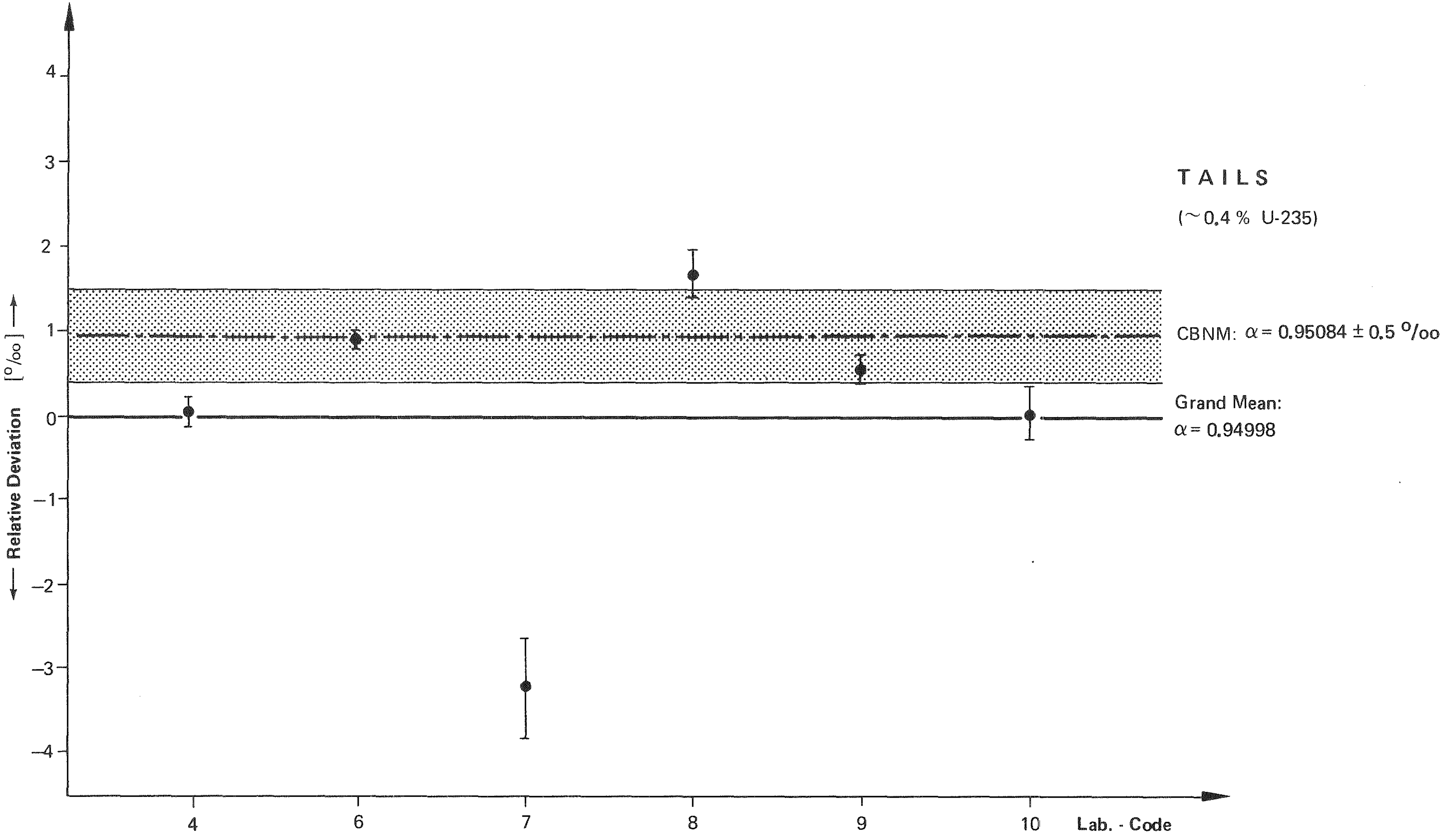


Fig. 4–1: Determination of the ratio α : deviations of laboratory means from grand mean. (Error bars indicate the relative standard deviations of the laboratory means).

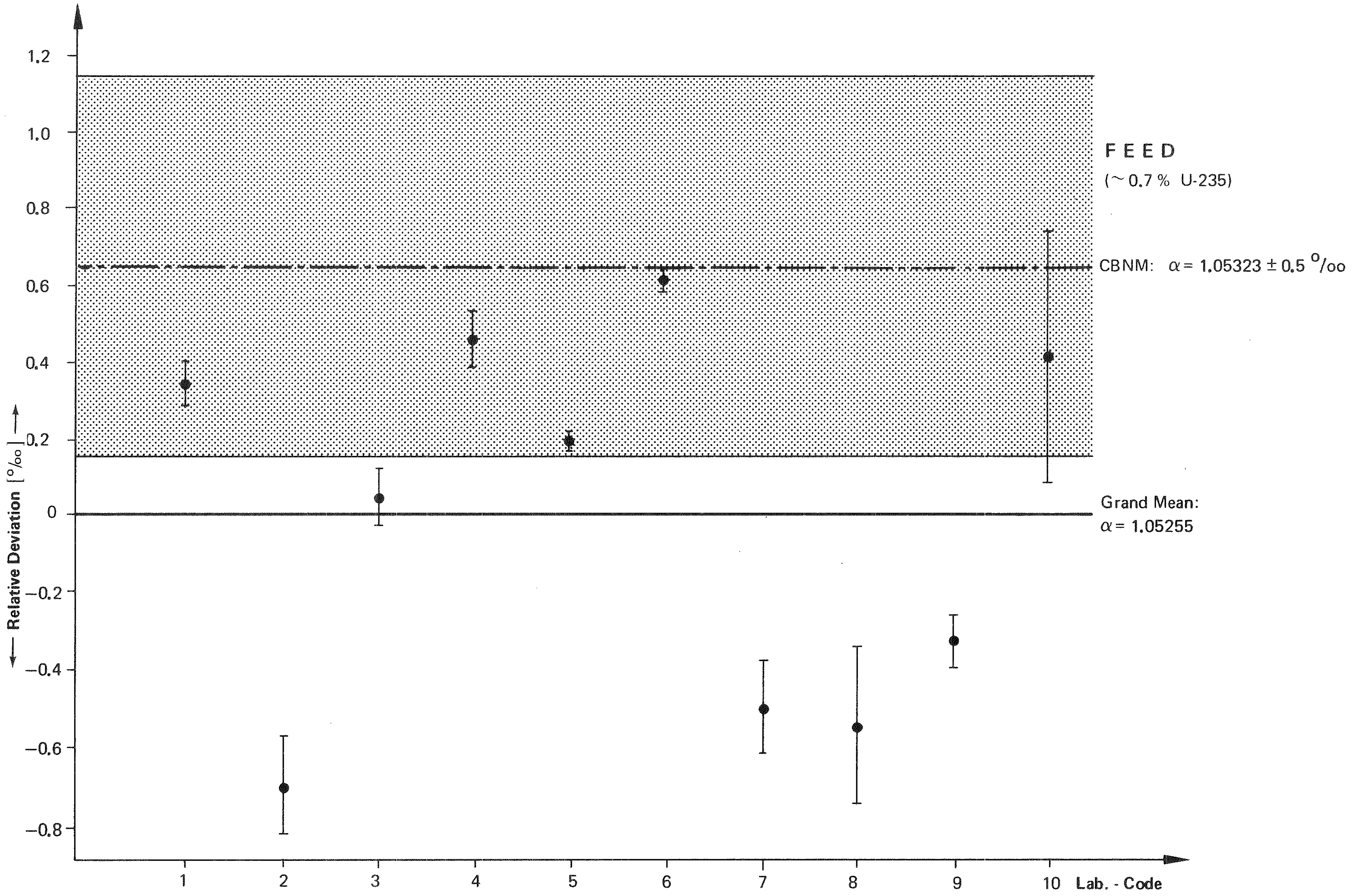


Fig. 4–2: Determination of the ratio α : deviations of laboratory means from grand mean. (Error bars indicate the relative standard deviations of the laboratory means).

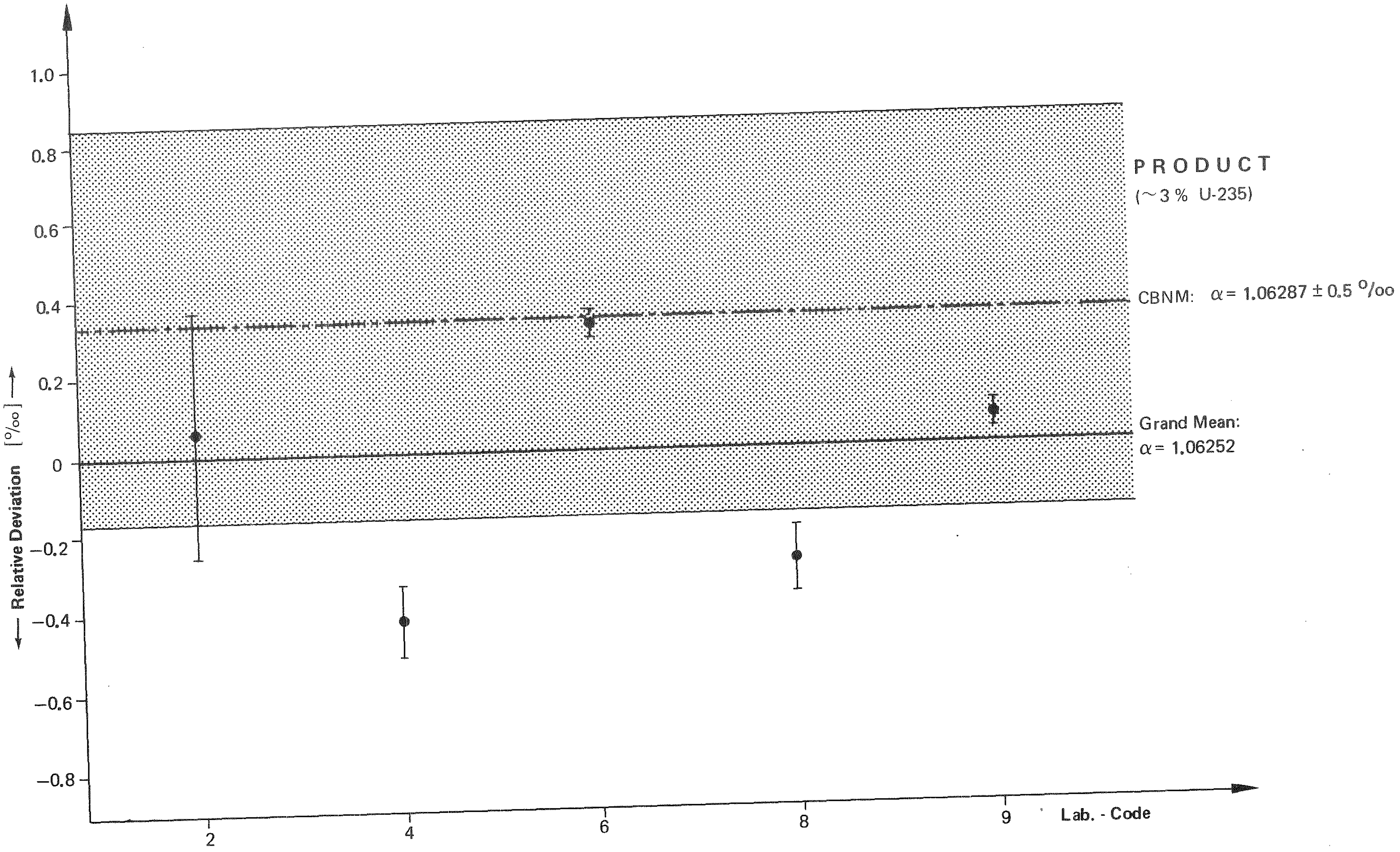


Fig. 4–3: Determination of the ratio α : deviations of laboratory means from grand mean. (Error bars indicate the relative standard deviations of the laboratory means).

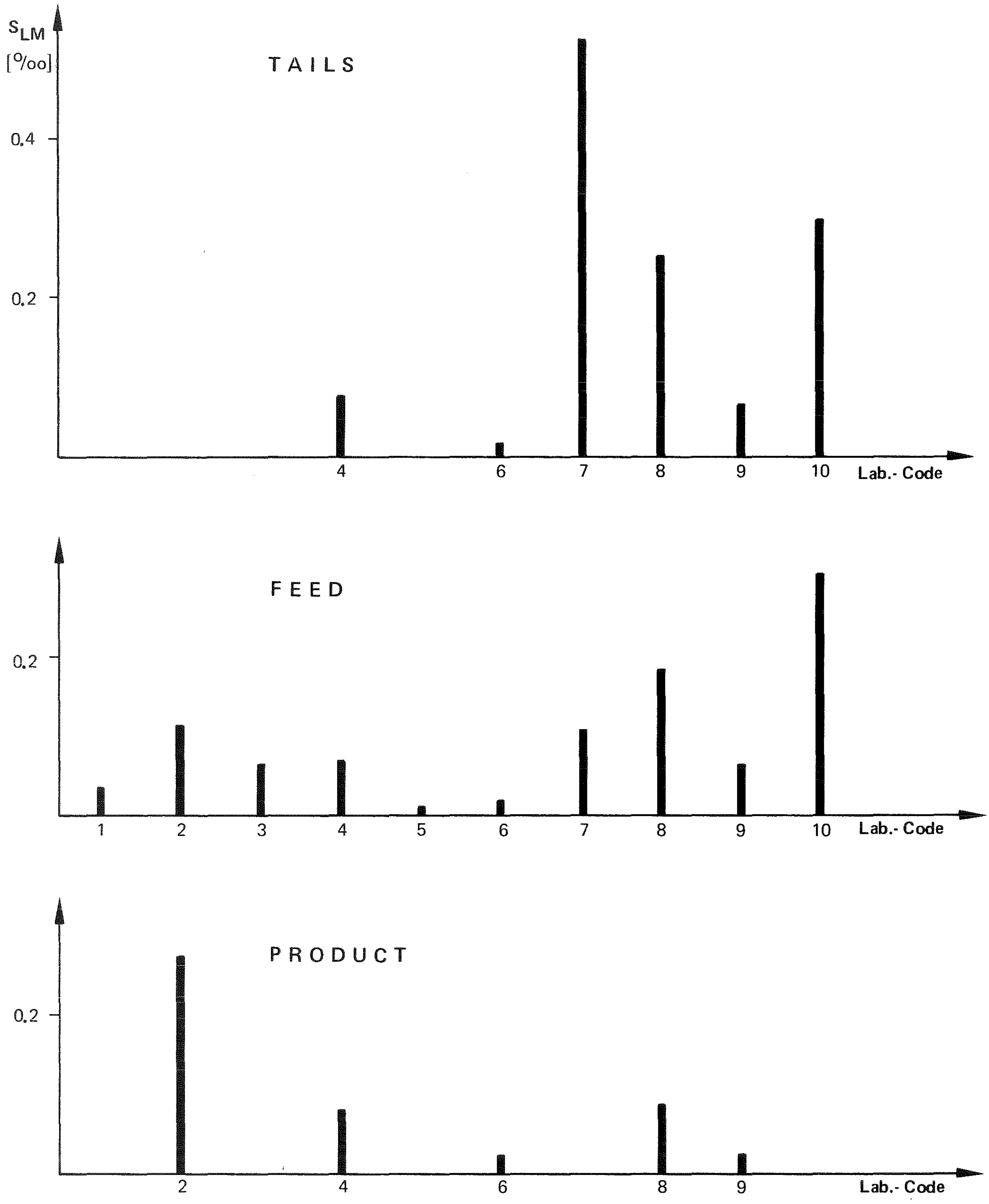


Fig. 4–4: Relative standard deviations of laboratory means

5. Calculation of Estimated Values of the Error Components of Ratio Determinations and of the Total Measurement Errors to be Expected in Routine Operation

As explained in more detail in Annex III, Chapter 5, the structure of the experiment allows to break down into three single components the total error obtained in the determination of the ratio a and to calculate estimated values of their variances using the method of variance analysis. They are mean values of the scattering of values measured during a cycle and during a period as well as of laboratory means, all of them based on the measurements of all laboratories.

The results of these calculations have been compiled in columns 4 to 6 of Table 5-1. Instead of variances, the corresponding relative standard deviations have been indicated because of their better illustrative capacity. In column 2 those laboratories have been mentioned which have provided the data for the particular evaluation while in column 3 the total number of individual measured values are given which, consequently, had been available for calculation.

In principle, the data of all laboratories were used for the calculations, which had participated in the measurements of the respective sample material. However, to be able to compare the error components calculated for the different sample materials on the basis of auniform group of laboratories, additional calculations were made with respect to the group of laboratories 4, 6, 8 and 9 participating in all the measurements.

Since on account of their narrow basis these results do not seem appropriate to describe the absolute values of the error components to be expected in routine operation, they have been put in brackets.

The dependence on the U-235 abundance of the measurement accuracy achieved by mass spectrometry gets clearly visible: Leaving apart a minor irregularity, all error components decrease with increasing U-235 content.

The influence of the individual error components upon the total error occurring in the determination of the ratio a gets particularly evident from the formula 13 given in Annex III according to which the relative standard deviation of a mean value of measured values is calculated from the error components given in Table 5-1 to be

Table 5–1: Estimates of Error Components Calculated by Analysis of Variances *)

1	2	3	4	5	6
Sample	Group of Laboratories	Number of Measured Values	RSD of Error Components [$^o/oo$]		
			"Cycle" S_C	"Period" S_P	"Laboratory" S_L
"Tails" (~ 0.4 % U-235)	4, 6, 7, 8, 9, 10 (4, 6, 8, 9)	216 (144)	0.41 (0.31)	0.72 (0.35)	1.67 (0.64)
"Feed" (~ 0.7 % U-235)	1 to 10 (4, 6, 8, 9)	360 (144)	0.36 (0.23)	0.32 (0.27)	0.46 (0.56)
"Product" (~ 3 % U-235)	2, 4, 6, 8, 9 (4, 6, 8, 9)	180 (144)	0.33 (0.19)	0.34 (0.13)	0.18 (0.26)

*) Calculated using "HP3" program, Annex IV

$$S_{total} = \sqrt{\frac{S_L^2}{l} + \frac{S_P^2}{l \cdot p} + \frac{S_C^2}{l \cdot p \cdot c}}$$

where l is the number of laboratories, each of them performing the measurements within p periods comprising c cycles each. Intercomparison of the error components makes visible the governing influence of the "laboratory" component on the total error if one considers that, in general, at least the contribution of the "cycle" component gets reduced by repetition measurements.

When checking the suitability of the available data material for the application of the method of variance analysis, it was examined whether the scattering of the mean values of periods belonging to the measurements of one single laboratory can be considered "random" in the statistical meaning or whether a significant systematic dependence on time can be recognized. It appeared that the existence of such an effect can in no case be the conclusion, if the statistical error bars are taken into account. Some examples have been shown in Fig. 5-1.

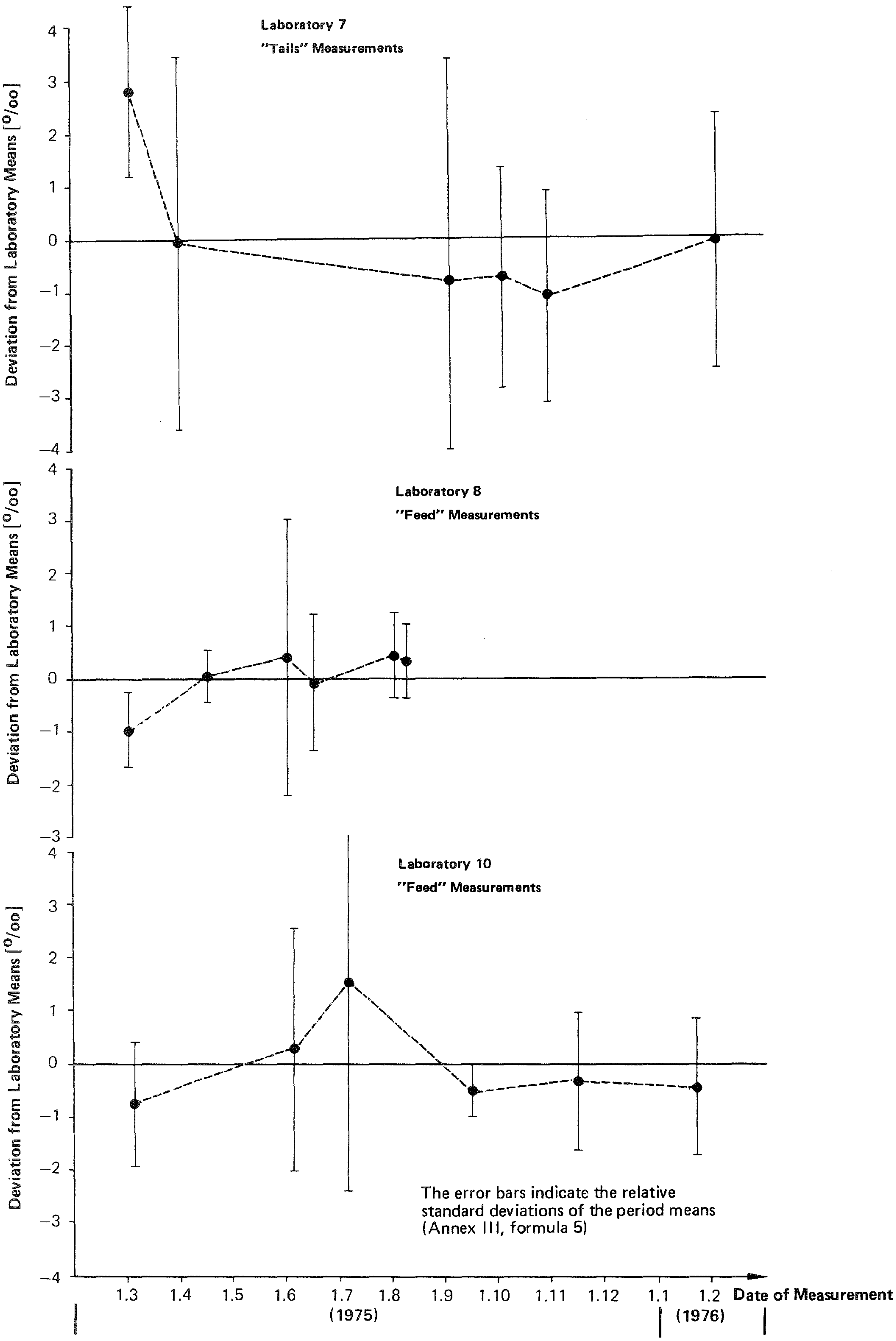
It seems meaningful under these circumstances to combine the "period" error component S_P with the "laboratory" error S_L since in routine operation only one period of measurements is usually performed (p = 1).

The numerical value of the term S_{LP} (p = 1) = $\sqrt{S_L^2 + S_P^2}$ can be calculated from the data of Table 5-1 (columns 5 and 6) for the sample materials used in the test.

If the measurement is performed by one laboratory only (l = 1), the relative standard deviation

$$S_{total}\,(l = p = 1) = \sqrt{S_{LP}^2 + \frac{S_C^2}{c}}$$

obtained in accordance with the formula above has to be assigned as total statistical error to the mean value derived from c cycles. For the case c = 6 this expression was calculated using the values given in Table 5-1. Its amount is almost exclusively determined by the first term of the sum S_{LP}^2, i.e., the error bar can practically not be reduced by increasing the number of measurement cycles.



The error bars indicate the relative standard deviations of the period means (Annex III, formula 5)

Fig. 5—1: Determination of the ratio α : period means as a function of time

In Fig. 5-2 the double value ("2σ-value") of this expression has been represented as a function of the U-235 content of the sample. It shows the rise in the error of measurement with decreasing U-235 content.

Since the calculation of the U-235 abundance in a measured sample always requires the knowledge of the U-235 content in the reference sample used ("standard"), the expected error bar of this variable of ± 1.5 $^{0}/oo$ was indicated by the dotted line in the figure[1]. Accordingly, the uncertainty in the isotopic composition of the standard makes a greater contribution to the total error in the determination of U-235 concentrations performed in the natural range and above. For the depleted material the statistical errors in the measurement of the ratio a become a more determinant factor. However, the accuracy of these statements has to be verified if also in the determination of the U-235 abundance in the standard a relative error has to be assumed which depends on the U-235 content itself.

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[1] This value has been derived from the errors indicated in Chapter 3.

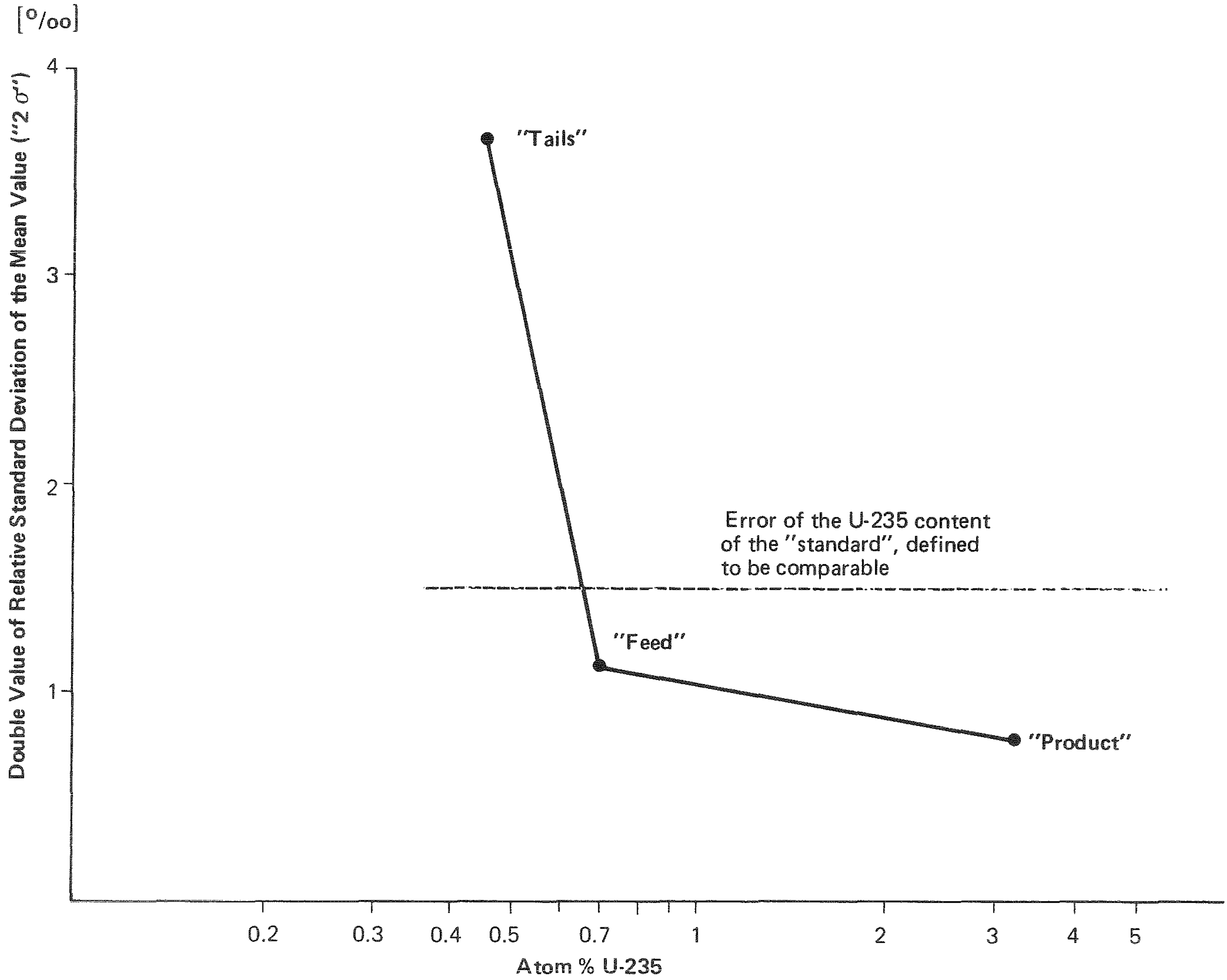


Fig. 5–2: Double value of relative standard deviation ("2 σ")of a determination of the ratio comprising 6 cycles of measurements as a function of the U-235 content

6. Summary of Results

As far as the laboratory group participating in the experiment and the expenditure in terms of measurement technology are considered as representative of routine operation and assuming that all laboratories use the same reference materials (standards), the following statements can be made:

6.1 In the range under consideration the statistical error in the determination of the ratio of ratios $a = (\text{U-235:U-238})_{\text{measured sample}} / (\text{U-235:238})_{\text{reference material}}$ increases with decreasing U-235 abundance. For samples having a natural or enriched U-235 content the statistical error amounts to about 0.1 % (double relative standard deviation from the mean value) if the measurement is performed by one single laboratory (see Chapter 5).

6.2 Of the error components considered the "interlab deviation" contributes most to the total error resulting from the determination of the ratios. It can, e.g., be caused by the memory effect (in the ion source part and in the inlet system of the mass spectrometer) (see Chapters 4 and 5 as well as Annex VIII).

6.3 A significant systematic long-term change in the determination of the ratio a cannot be detected (see Chapter 5).

6.4 In the determination of the U-235 abundance within the natural range and for enriched material the uncertainty in the isotopic composition of the reference material (standard) makes a higher contribution to the total error of the result than the measurement of the ratio a. In case of the depleted material the reverse was found in this test.

This statement is valid assuming a concentration independent uncertainty of about ± 0.15 % of the U-235 content in the reference material (standard) (see Chapter 5).

6.5 The improvement of the absolute determination of U-235 abundance in UF_6 measurements presently calls preferably for a reduction in the uncertainty of the isotopic composition of the reference materials available (standards).

With respect to the depleted material special attention has also to be paid to the measurement technology.

6.6 Investigations performed by two laboratories yielded a large independence of the determined U-235 abundance of the ratio of U-235 abundances in the sample and in the reference material (standard), approximately up to the ratio 2. So, in these cases, there is no indication whatsoever of a significant "memory effect" (see Annexes V and VII).

Annex I:

Data Reported by the Laboratories

Data - Laboratory 1

Feed:

Period of Measurement		Measurement Cycle						Date of Measurement
		1	2	3	4	5	6	
Test Material	1	69052	69046	69038	69062	69035	69033	6.3.1975
	2	68244	68233	68161	68213	68171	68183	14.4.1975
	3	68867	68858	68860	68853	68855	68855	5.5.1975
	4	68856	68852	68836	68833	68823	68834	22.5.1975
	5	68709	68702	68700	68706	68694	68684	3.7.1975
	6	68797	68774	68754	68741	68727	68735	22.7.1975
Reference Material	1	65592	65592	65575	65586	65580	65574	6.3.1975
	2	64767	64785	64767	64756	64804	64801	14.4.1975
	3	65402	65392	65403	65382	65381	65395	5.5.1975
	4	65405	65388	65375	65362	65356	65362	22.5.1975
	5	65262	65250	65240	65229	65230	65234	3.7.1975
	6	65351	65314	65311	65295	65272	65268	22.7.1975

Data - Laboratory 2

Feed:

Period of Measurement		Measurement Cycle						Date of Measurement
		1	2	3	4	5	6	
Test Material	1	7106	7107	7109	7106	7107	7105	11.4.1975
	2	7110	7112	7113	7114	7116	7115	14.5.1975
	3	7116	7114	7112	7113	7112	7111	12.6.1975
	4	7138	7132	7136	7135	7140	7131	6.8.1975
	5	7057	7057	7054	7052	7052	7056	13.11.1975
	6	7047	7043	7041	7029	7031	7028	25.11.1975
Reference Material	1	6755	6759	6755	6753	6750	6753	11.4.1975
	2	6760	6764	6764	6765	6766	6764	14.5.1975
	3	6767	6765	6765	6766	6764	6764	12.6.1975
	4	6791	6792	6783	6782	6780	6777	6.8.1975
	5	6715	6710	6706	6712	6708	6703	13.11.1975
	6	6695	6691	6683	6688	6688	6679	25.11.1975

Data - Laboratory 2

Product:

Period of Measurement		Measurement Cycle						Date of Measurement
		1	2	3	4	5	6	
Test Material	1	32857	32858	32873	32864	32839	32839	14.8.1975
	2	33248	33246	33234	33228	33252	33244	10.11.1975
	3	33536	33537	33544	33542	33552	33538	24.11.1975
	4	33084	33100	33105	33103	33099	33112	27.11.1975
	5	33117	33110	33104	33121	33118	33130	27.11.1975
	6	33124	33144	33150	33148	33155	33160	27.11.1975
Reference Material	1	30966	30955	30964	30952	30968	30974	14.8.1975
	2	31290	31286	31289	31286	31292	31264	10.11.1975
	3	31540	31545	31539	31561	31560	31564	24.11.1975
	4	31193	31181	31144	31137	31135	31131	27.11.1975
	5	31150	31145	31148	31152	31127	31122	27.11.1975
	6	31182	31176	31194	31185	31201	31196	27.11.1975

Data - Laboratory 3

Feed:

Period of Measurement		**Measurement Cycle**						**Date of Measurement**
		1	2	3	4	5	6	
Test Material	**1**	98002	97962	97918	97974	97966	97954	5.3.1975
	2	98084	98060	98070	98020	97982	98016	14.4.1975
	3	98118	98055	98058	98030	98030	98028	4.5.1975
	4	98143	98108	98062	98087	98085	98082	16.5.1975
	5	97740	97805	97777	97825	97797	97847	26.6.1975
	6	97925	97862	97865	97872	97897	97875	4.8.1975
Reference Material	**1**	93120	93100	93070	93068	93050	93036	5.3.1975
	2	93168	93146	93136	93116	93106	93116	14.4.1975
	3	93178	93125	93113	93115	93108	93108	4.5.1975
	4	93198	93175	93177	93208	93230	93202	16.5.1975
	5	92840	92908	92980	92953	92952	92942	26.6.1975
	6	93007	92973	92955	93023	93033	92998	4.8.1975

Data - Laboratory 4 *)

Tails:

Period of Measurement		**Measurement Cycle**						**Date of Measurement**
		1	2	3	4	5	6	
Test Material	**1**	9504	9503	9502	9503	9500	9501	30.9.1975
	2	9501	9502	9505	9502	9500	9501	27.10.1975
	3	9498	9499	9499	9497	9498	9499	11.11.1975
	4	9497	9500	9496	9497	9497	9502	5.12.1975
	5	9503	9501	9502	9504	9503	9504	11.12.1975
	6	9502	9499	9498	9500	9504	9499	15.12.1975
Reference Material	**1**	10000	10000	10000	10000	10000	10000	30.9.1975
	2	10000	10000	10000	10000	10000	10000	27.10.1975
	3	10000	10000	10000	10000	10000	10000	11.11.1975
	4	10000	10000	10000	10000	10000	10000	5.12.1975
	5	10000	10000	10000	10000	10000	10000	11.12.1975
	6	10000	10000	10000	10000	10000	10000	15.12.1975

Data - Laboratory 4 *)

Feed:

Period of Measurement		**Measurement Cycle**						**Date of Measurement**
		1	2	3	4	5	6	
Test Material	1	10530	10529	10528	10529	10533	10529	30.9.1975
	2	10533	10533	10533	10531	10537	10533	27.10.1975
	3	10530	10529	10533	10532	10531	10530	11.11.1975
	4	10532	10531	10530	10531	10530	10529	5.12.1975
	5	10527	10528	10526	10527	10527	10529	11.12.1975
	6	10529	10529	10529	10530	10532	10532	15.12.1975
Reference Material	1	10000	10000	10000	10000	10000	10000	30.9.1975
	2	10000	10000	10000	10000	10000	10000	27.10.1975
	3	10000	10000	10000	10000	10000	10000	11.11.1975
	4	10000	10000	10000	10000	10000	10000	5.12.1975
	5	10000	10000	10000	10000	10000	10000	11.12.1975
	6	10000	10000	10000	10000	10000	10000	15.12.1975

*) The laboratories 4 and 6 reported only the ratio $\alpha = (\text{U-235: U-238})_{\text{test material}} / (\text{U-235: U-238})_{\text{reference material}}$.
The values 10.000 and 1.000.000, respectively, had to be entered for reasons of programming.

Data - Laboratory 4 *)

Product:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	10621	10621	10621	10622	10619	10620	30.9.1975
	2	10623	10621	10625	10625	10623	10625	27.10.1975
	3	10628	10627	10627	10627	10625	10628	11.11.1975
	4	10622	10625	10620	10617	10621	10623	5.12.1975
	5	10625	10623	10623	10622	10624	10628	11.12.1975
	6	10625	10625	10625	10624	10627	10625	15.12.1975
Reference Material	1	10000	10000	10000	10000	10000	10000	30.9.1975
	2	10000	10000	10000	10000	10000	10000	27.10.1975
	3	10000	10000	10000	10000	10000	10000	11.11.1975
	4	10000	10000	10000	10000	10000	10000	5.12.1975
	5	10000	10000	10000	10000	10000	10000	11.12.1975
	6	10000	10000	10000	10000	10000	10000	15.12.1975

Data - Laboratory 5

Feed:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	62680	62775	62775	62775	62780	62775	26.2.1975
	2	62810	62810	62800	62790	62790	62795	18.3.1975
	3	62600	62610	62600	62600	62610	62600	6.5.1975
	4	62515	62525	62515	62515	62515	62520	27.5.1975
	5	62240	62240	62235	62235	62245	62235	23.6.1975
	6	62110	62100	62100	62095	62090	62085	7.8.1975
Reference Material	1	59640	59635	59630	59630	59630	59635	26.2.1975
	2	59665	59665	59650	59660	59645	59645	18.3.1975
	3	59460	59460	59470	59460	59460	59470	6.5.1975
	4	59380	59390	59385	59385	59385	59385	27.5.1975
	5	59120	59120	59110	59120	59125	59120	23.6.1975
	6	58990	58980	58990	58985	58990	58980	7.8.1975

Data - Laboratory 6 *)

Tails:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	1000000	1000000	1000000	1000000	1000000	1000000	9.9.1975
	2	1000000	1000000	1000000	1000000	1000000	1000000	9.9.1975
	3	1000000	1000000	1000000	1000000	1000000	1000000	9.9.1975
	4	1000000	1000000	1000000	1000000	1000000	1000000	9.9.1975
	5	1000000	1000000	1000000	1000000	1000000	1000000	9.9.1975
	6	1000000	1000000	1000000	1000000	1000000	1000000	9.9.1975
Reference Material	1	1051461	1051716	1051660	1051755	1051530	1051848	9.9.1975
	2	1051632	1051579	1051863	1051754	1051977	1051622	9.9.1975
	3	1051735	1051509	1051799	1051557	1051939	1052050	9.9.1975
	4	1051696	1051579	1051768	1051813	1051518	1051737	9.9.1975
	5	1051784	1051839	1051608	1051946	1051731	1051565	9.9.1975
	6	1051684	1051646	1051874	1051770	1051757	1051765	9.9.1975

*) The laboratories 4 and 6 reported only the ratio $\alpha = (\text{U-235: U-238})_{\text{test material}} / (\text{U-235: U-238})_{\text{reference material}}$. The values 10.000 and 1.000.000, respectively, had to be entered for reasons of programming.

Data - Laboratory 6 *)

Feed:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	1000000	1000000	1000000	1000000	1000000	1000000	27.8.1975
	2	1000000	1000000	1000000	1000000	1000000	1000000	27.8.1975
	3	1000000	1000000	1000000	1000000	1000000	1000000	27.8.1975
	4	1000000	1000000	1000000	1000000	1000000	1000000	27.8.1975
	5	1000000	1000000	1000000	1000000	1000000	1000000	27.8.1975
	6	1000000	1000000	1000000	1000000	1000000	1000000	27.8.1976
Reference Material	1	949554	949378	949420	949486	949489	949391	27.8.1975
	2	949554	949347	949424	949611	949384	949427	27.8.1975
	3	949428	949394	949458	949439	949458	949481	27.3.1975
	4	949364	949342	949729	949888	949372	949384	27.8.1975
	5	949572	949524	949458	949634	949440	949577	27.8.1975
	6	949553	949575	949569	949520	949636	949475	27.8.1975

Data Laboratory 6 *)

Product:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	1000000	1000000	1000000	1000000	1000000	1000000	11.9.1975
	2	1000000	1000000	1000000	1000000	1000000	1000000	11.9.1975
	3	1000000	1000000	1000000	1000000	1000000	1000000	11.9.1975
	4	1000000	1000000	1000000	1000000	1000000	1000000	11.9.1975
	5	1000000	1000000	1000000	1000000	1000000	1000000	11.9.1975
	6	1000000	1000000	1000000	1000000	1000000	1000000	11.9.1975
Reference Material	1	940606	940787	940847	940958	940675	940857	11.9.1975
	2	940947	940854	940941	940824	940934	940819	11.9.1975
	3	940683	940810	941001	940992	940798	940827	11.9.1975
	4	940744	940789	940834	940886	940762	940797	11.9.1975
	5	940861	940963	940970	941086	940913	940954	11.9.1975
	6	940766	940828	940927	940904	940805	940876	11.9.1975

Data - Laboratory 7

Tails:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	43189	43187	43247	43277	43214	43258	6.3.1975
	2	43192	42304	42120	42132	42443	42272	1.4.1975
	3	42294	42280	42535	42543	42619	42561	5.9.1975
	4	42586	42621	42598	42594	42650	42634	3.10.1975
	5	42637	42663	51143	51116	51289	51379	22.10.1975
	6	51374	50974	51957	42798	42793	42799	30.1.1976
Reference Material	1	45476	45490	45519	45571	45511	45583	6.3.1975
	2	45614	44740	44479	44512	44826	44587	1.4.1975
	3	44795	44689	44937	44934	45036	44955	5.9.1975
	4	45040	45063	45005	45000	45055	45073	3.10.1975
	5	45060	45130	54049	54074	54212	54292	22.10.1975
	6	54269	53826	54931	45176	45180	45185	30.1.1976

*) The laboratories 4 and 6 reported only the ratio $\alpha = (\text{U-235: U-238})_{\text{test material}}/(\text{U-235: U-238})_{\text{reference material}}$.
The values 10.000 and 1.000.000, respectively, had to be entered for reasons of programming.

Data - Laboratory 7

Feed:

Period of Measurement		Measurement Cycle						Date of Measurement
		1	2	3	4	5	6	
Test Material	1	71131	71160	71203	71310	71249	71274	6.3.1975
	2	71079	69726	69458	69412	69952	69709	1.4.1975
	3	69730	69667	70085	70098	70169	70141	5.9.1975
	4	70171	70206	70153	70200	70214	70210	3.10.1975
	5	70242	70236	84205	84283	84671	84701	22.10.1975
	6	84789	83946	85605	70528	70528	70523	29.1.1976
Reference Material	1	67620	67622	67652	67745	67736	67729	6.3.1975
	2	67561	66268	66024	65977	66461	66209	1.4.1975
	3	66267	66184	66605	66664	66738	66700	5.9.1975
	4	66762	66763	66676	66776	66742	66786	3.10.1975
	5	66761	66858	80097	80075	80473	80513	22.10.1975
	6	80582	79768	81360	67007	67022	67027	29.1.1976

Data - Laboratory 8

Tails:

Period of Measurement		Measurement Cycle						Date of Measurement
		1	2	3	4	5	6	
Test Material	1	4255	4251	4245	4239	4240	4234	2.3.1975
	2	4310	4309	4309	4309	4308	4309	15.4.1975
	3	4303	4302	4303	4304	4303	4302	28.5.1975
	4	4300	4298	4299	4300	4299	4299	15.6.1975
	5	4314	4312	4313	4315	4315	4316	28.7.1975
	6	4322	4324	4323	4325	4323	4321	9.8.1975
Reference Material	1	4464	4456	4455	4452	4450	4448	2.3.1975
	2	4531	4530	4531	4530	4529	4530	15.4.1975
	3	4520	4523	4521	4522	4520	4521	28.5.1975
	4	4519	4520	4520	4519	4519	4520	15.6.1975
	5	4536	4535	4535	4536	4534	4536	28.7.1975
	6	4546	4547	4545	4546	4544	4544	9.8.1975

Data - Laboratory 8

Feed:

Period of Measurement		Measurement Cycle						Date of Measurement
		1	2	3	4	5	6	
Test Material	1	6955	6958	6962	6968	6965	6963	1.3.1975
	2	7064	7062	7060	7061	7060	7061	15.4.1975
	3	7045	7043	7042	7043	7045	7047	28.5.1975
	4	7042	7045	7049	7048	7048	7049	15.6.1975
	5	7047	7046	7047	7048	7048	7048	1 .8.1975
	6	7050	7050	7051	7050	7054	7053	7.8.1975
Reference Material	1	6616	6622	6624	6630	6627	6626	1.3.1975
	2	6715	6714	6712	6711	6711	6711	15.4.1975
	3	6687	6695	6695	6696	6695	6694	28.5.1975
	4	6698	6699	6699	6700	6701	6699	15.6.1975
	5	6695	6694	6696	6698	6699	6698	1.8.1975
	6	6699	6700	6700	6702	6703	6702	7.8.1975

Data - Laboratory 8

Product:

Period of Measurement				**Measurement Cycle**				**Date of Measurement**
		1	**2**	**3**	**4**	**5**	**6**	
Test Material	**1**	32628	32635	32633	32638	32639	32634	28.7.1975
	2	32669	32673	32669	32665	32680	32675	11.8.1975
	3	32645	32653	32662	32658	32670	32678	2.10.1975
	4	32642	32643	32650	32651	32648	32649	27.10.1975
	5	32610	32598	32615	32578	32579	32600	26.11.1975
	6	33165	33154	33159	33190	33091	33090	19.12.1975
Reference Material	**1**	30710	30718	30717	30720	30718	30717	28.7.1975
	2	30744	30762	30755	30756	30760	30759	11.8.1975
	3	30722	30742	30750	30760	30758	30767	2.10.1975
	4	30720	30734	30745	30748	30721	30722	27.10.1975
	5	30700	30698	30705	30695	30692	30700	26.11.1975
	6	31210	31225	31235	31232	31150	31141	19.12.1975

Data - Laboratory 9

Tails:

Period of Measurement				**Measurement Cycle**				**Date of Measurement**
		1	2	3	4	5	6	
Test Material	**1**	4159	4156	4154	4153	4151	4151	16.4.1975
	2	4147	4145	4145	4144	4144	4143	21.5.1975
	3	4140	4139	4138	4150	4145	4144	13.6.1975
	4	4147	4141	4142	4142	4141	4141	23.6.1975
	5	4144	4141	4139	4139	4136	4135	23.7.1975
	6	4146	4144	4144	4144	4140	4140	19.8.1975
Reference Material	**1**	4374	4375	4372	4371	4368	4368	16.4.1975
	2	4362	4362	4362	4360	4360	4360	21.5.1975
	3	4354	4355	4360	4361	4361	4359	13.6.1975
	4	4360	4357	4357	4358	4358	4357	23.6.1975
	5	4356	4356	4354	4353	4350	4350	23.7.1975
	6	4361	4360	4359	4359	4355	4355	19.8.1975

Data - Laboratory 9

Feed:

Period of Measurement				**Measurement Cycle**				**Date of Measurement**
		1	2	3	4	5	6	
Test Material	1	6842	6835	6835	6833	6835	6828	15.5.1975
	2	6831	6825	6827	6824	6823	6824	22.5.1975
	3	6833	6826	6813	6822	6814	6809	12.6.1975
	4	6815	6816	6811	6810	6810	6810	24.6.1975
	5	6816	6815	6811	6810	6808	6807	22.7.1975
	6	6806	6808	6806	6806	6807	6808	19.8.1975
Reference Material	1	6498	6495	6496	6492	6493	6489	15.5.1975
	2	6492	6490	6489	6489	6489	6483	22.5.1975
	3	6491	6484	6480	6482	6476	6472	12.6.1975
	4	6478	6478	6473	6473	6473	6473	24.6.1975
	5	6477	6477	6473	6472	6470	6469	22.7.1975
	6	6468	6471	6468	6469	6460	6470	19.8.1975

Data - Laboratory 9

Product:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	31780	31780	31770	31765	31745	31730	20.8.1975
	2	31810	31815	31780	31780	31775	31780	25.9.1975
	3	31810	31810	31800	31800	31810	31800	6.11.1975
	4	31820	31820	31810	31805	31800	31790	2.12.1975
	5	31795	31765	31760	31750	31760	31755	8.12.1975
	6	31770	31740	31750	31755	31770	31760	17.12.1975
Reference Material	1	29915	29910	29897	29890	29873	29860	20.8.1975
	2	29940	29940	29910	29910	29910	29910	25.9.1975
	3	29935	29930	29930	29930	29940	29930	6.11.1975
	4	29945	29940	29935	29930	29920	29920	2.12.1975
	5	29920	29890	29890	29882	29890	29895	8.12.1975
	6	29890	29875	29880	29885	29890	29890	17.12.1975

Data - Laboratory 10

Tails:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	35996	35976	36000	36000	35997	35971	12.3.1975
	2	36072	36039	35998	36025	36026	36010	5.6.1975
	3	36100	36034	36036	36029	36042	36029	9.7.1975
	4	37200	37220	37210	37210	37200	37207	16.9.1975
	5	37040	37020	36972	36980	36943	36955	14.11.1975
	6	36958	36969	36940	36945	36947	36944	15.1.1976
Reference Material	1	37900	37858	37881	37880	37877	37860	12.3.1975
	2	38000	37984	37960	38022	37952	37952	5.6.1975
	3	37992	37900	37882	37892	37886	37882	9.7.1975
	4	39150	39171	39166	39150	39150	39162	16.9.1975
	5	38990	38967	38936	38924	38924	38914	14.11.1975
	6	38889	38883	38861	38872	38870	38875	15.1.1976

Data - Laboratory 10

Feed:

Period of Measurement		Measurement Cycle 1	2	3	4	5	6	Date of Measurement
Test Material	1	59380	59420	59420	59430	59420	59420	7.3.1975
	2	59396	59366	59369	59355	59368	59363	5.6.1975
	3	59376	59350	59263	59340	59398	59390	9.7.1975
	4	61321	61346	61348	61348	61356	61349	16.9.1975
	5	61290	61260	61250	61247	61228	61200	14.11.1975
	6	61120	61160	61160	61106	61142	61105	15.1.1976
Reference Material	1	56470	56480	56470	56470	56450	56470	7.3.1975
	2	56438	56360	56355	56355	56312	56364	5.6.1975
	3	56300	56257	56274	56309	56300	56241	9.7.1975
	4	58270	58286	58290	58295	58297	58276	16.9.1975
	5	58198	58178	58188	58180	58180	58160	14.11.1975
	6	58064	58100	58071	58063	58111	58139	15.1.1976

Annex II:

Individual Values Calculated for the Ratio of Ratios
a = (U-235:U-238)/$_{\text{test material}}$ / (U-235:U-238)$_{\text{reference material}}$

Note:
In this Annex, the ratios are given with 10 decimals independent of the number of digits reported by the laboratories (Annex I). If the last digit is a naught, it is not printed by the computer.

Data of "Tails" Samples (Total: 216)

Lab. Code	Period	Measurement Cycle					
		1	2	3	4	5	6
		$\alpha = (\text{U-235: U-238})_{\text{test material}} / (\text{U-235: U-238})_{\text{reference material}}$					
4	1	0.9504	0.9503	0.9502	0.9503	0.95	0.9501
	2	0.9501	0.9502	0.9505	0.9502	0.95	0.9501
	3	0.9498	0.9499	0.9499	0.9497	0.9498	0.9499
	4	0.9497	0.95	0.9496	0.9497	0.9497	0.9502
	5	0.9503	0.9501	0.9502	0.9504	0.9503	0.9504
	6	0.9502	0.9499	0.9498	0.95	0.9504	0.9499
6	1	0.9510576236	0.9508270294	0.9508776601	0.9507917718	0.9509952165	0.9507077068
	2	0.9509029775	0.9509509034	0.9506941493	0.9507926759	0.9505911251	0.9509120197
	3	0.9508098523	0.9510142091	0.9507519973	0.9509707985	0.950625464	0.9505251652
	4	0.9508451111	0.9509509034	0.9507800199	0.9507393424	0.9510060693	0.9508080442
	5	0.9507655564	0.9507158415	0.9509246792	0.9506191382	0.9508134685	0.9509635638
	6	0.9508559605	0.9508903186	0.9506842074	0.950778212	0.9507899638	0.9507827319
7	1	0.949709737	0.9493734887	0.950086777	0.9496609686	0.9495286854	0.9489941426
	2	0.9469022668	0.9455520787	0.9469637357	0.9465312725	0.9468388881	0.9480790365
	3	0.9441678759	0.9460941171	0.9465473886	0.9467886233	0.9463318234	0.9467467467
	4	0.9455150977	0.9457042691	0.9465170537	0.9465333333	0.9466207968	0.945887782
	5	0.9462272526	0.9453356969	0.9462339729	0.9452971853	0.9460820483	0.9463456863
	6	0.9466546279	0.947014454	0.9458593508	0.9473614308	0.947166888	0.9471948656
8	1	0.9531810036	0.953994614	0.9528619529	0.9521563342	0.9528089888	0.9518884892
	2	0.9512248952	0.951214128	0.9510041933	0.951214128	0.9512033561	0.951214128
	3	0.9519911504	0.9511386248	0.9517805795	0.9517912428	0.9519911504	0.9515593895
	4	0.9515379509	0.9508849558	0.9511061947	0.9515379509	0.951316663	0.9511061947
	5	0.9510582011	0.9508269019	0.951047409	0.9512786596	0.9516982797	0.9514991182
	6	0.9507259129	0.9509566747	0.9511551155	0.9513858337	0.9513644366	0.9509242958
9	1	0.9508459076	0.9499428571	0.950137237	0.9501258293	0.9503205128	0.9503205128
	2	0.9507106832	0.9502521779	0.9502521779	0.9504587156	0.9504587156	0.9502293578
	3	0.9508497933	0.950401837	0.9490825688	0.9516166017	0.9504700757	0.9506767607
	4	0.951146789	0.9504246041	0.9506541198	0.9504359798	0.9502065168	0.9504246041
	5	0.9508949059	0.9506427916	0.9506201194	0.9508385022	0.9508045977	0.9505747126
	6	0.9506993809	0.9504587156	0.9506767607	0.9506767607	0.9506314581	0.9506314581
10	1	0.949762533	0.950287918	0.9503444999	0.9503695882	0.9503656573	0.9501056524
	2	0.9492631579	0.9487942291	0.9483140148	0.947477776	0.9492516863	0.9488301012
	3	0.9502000421	0.9507651715	0.9512697323	0.9508339491	0.9513276672	0.951084948
	4	0.9501915709	0.9501927446	0.9500587244	0.9504469987	0.9501915709	0.9500791584
	5	0.9499871762	0.9500346447	0.9495582494	0.9500565204	0.9491059501	0.9496582207
	6	0.9503458562	0.9507754031	0.9505674069	0.9504270426	0.950527399	0.9503279743

Data of "Feed" Samples (Total: 360)

Lab. Code	Period	Measurement Cycle					
		1	2	3	4	5	6
		α = (U-235: U-238)$_{\text{test material}}$ / (U-235: U-238)$_{\text{reference material}}$					
1	1	1.052750335	1.052658861	1.05280976	1.052999116	1.052683745	1.052749565
	2	1.053684747	1.053222196	1.05240323	1.053385015	1.051956669	1.052190553
	3	1.052980031	1.053003425	1.052856903	1.053088006	1.053134703	1.052909244
	4	1.05276355	1.052976081	1.052940727	1.05310425	1.053047922	1.05311955
	5	1.052817873	1.052904215	1.053034948	1.053304512	1.0531044	1.052886532
	6	1.052730639	1.05297486	1.052717	1.052775863	1.052932345	1.053119446
2	1	1.05196151	1.051486906	1.052405625	1.052273064	1.052888889	1.052124981
	2	1.051775148	1.051448847	1.051596688	1.051589061	1.051729234	1.051892371
	3	1.051573814	1.051589061	1.051293422	1.051285841	1.051448847	1.051301005
	4	1.05109704	1.050058893	1.052041869	1.052049543	1.053097345	1.052235502
	5	1.050930752	1.05171386	1.051893826	1.050655542	1.051282051	1.052662987
	6	1.05257655	1.052607981	1.053568757	1.050986842	1.051285885	1.052253331
3	1	1.052426976	1.052223416	1.052089825	1.052714144	1.052831811	1.052861258
	2	1.052764898	1.052755889	1.052976293	1.052665493	1.052370417	1.052622535
	3	1.053016807	1.052939597	1.053107515	1.052784192	1.052863341	1.052841861
	4	1.053059079	1.05294338	1.052427101	1.052345292	1.052075512	1.052359391
	5	1.052778975	1.052708055	1.05159174	1.052413585	1.052123677	1.052774849
	6	1.05287774	1.052585159	1.052821258	1.052126893	1.052282523	1.052441988
4	1	1.053	1.0529	1.0528	1.0529	1.0533	1.0529
	2	1.0533	1.0533	1.0533	1.0531	1.0537	1.0533
	3	1.053	1.0529	1.0533	1.0532	1.0531	1.053
	4	1.0532	1.0531	1.053	1.0531	1.053	1.0529
	5	1.0527	1.0528	1.0526	1.0527	1.0527	1.0529
	6	1.0529	1.0529	1.0529	1.053	1.0532	1.0532
5	1	1.052649229	1.052653643	1.052741908	1.052741908	1.052825759	1.052653643
	2	1.05271097	1.052887436	1.052808047	1.052463962	1.052728644	1.052812474
	3	1.052808611	1.052976791	1.052631579	1.052808611	1.052976791	1.052631579
	4	1.052795554	1.052786664	1.052706913	1.052706913	1.052706913	1.052791109
	5	1.052774019	1.052774019	1.052867535	1.052689445	1.052769556	1.052689445
	6	1.05289032	1.052899288	1.0527208	1.052725269	1.05255128	1.052644964
6	1	1.053125994	1.053321227	1.053274631	1.053201416	1.053198089	1.053306804
	2	1.053137085	1.053355622	1.053270193	1.05306278	1.05331457	1.053266865
	3	1.053265756	1.053303476	1.053232476	1.053253553	1.053232476	1.053206963
	4	1.05333676	1.05336117	1.052931942	1.052755693	1.053327884	1.05331457
	5	1.053106031	1.053159267	1.053232476	1.053037275	1.053252444	1.053100486
	6	1.053127103	1.053102704	1.053109358	1.053163704	1.053035058	1.053213618
7	1	1.051922508	1.052320251	1.052489209	1.05262381	1.051863116	1.052340947
	2	1.052071461	1.052182049	1.05201139	1.052063598	1.052527046	1.052862904
	3	1.052258288	1.052626012	1.05224833	1.05151206	1.051409991	1.051589205
	4	1.051061981	1.051570481	1.052147699	1.051275908	1.052021216	1.05126823
	5	1.05214122	1.050524993	1.051287814	1.052550734	1.052166565	1.052016445
	6	1.052207689	1.052376893	1.052175516	1.052546749	1.052311181	1.052158086
8	1	1.05123942	1.050739958	1.05102657	1.050980392	1.051003471	1.050860248
	2	1.051973194	1.051831993	1.051847437	1.052153181	1.052004172	1.052153181
	3	1.053536713	1.051979089	1.051829724	1.051821983	1.052277819	1.052733791
	4	1.051358615	1.0516495	1.052246604	1.051940299	1.051783316	1.052246604
	5	1.05257655	1.052584404	1.052419355	1.052254404	1.052097328	1.052254404
	6	1.05239588	1.052238806	1.05238806	1.051924799	1.052364613	1.052372426
9	1	1.052939366	1.05234796	1.052185961	1.052526186	1.052672108	1.052242256
	2	1.052218115	1.051617874	1.052088149	1.051625828	1.051471721	1.052599105
	3	1.052688338	1.052745219	1.051388889	1.052452947	1.052192712	1.052070457
	4	1.052022229	1.052176598	1.052216901	1.052062413	1.052062413	1.052062413
	5	1.052339046	1.052184653	1.052216901	1.052224969	1.052241113	1.052249188
	6	1.052257267	1.052078504	1.052257267	1.052094605	1.052249188	1.052241113
10	1	1.051531787	1.052053824	1.052240128	1.052417213	1.052612932	1.052240128
	2	1.052411496	1.053335699	1.053482388	1.053233963	1.054269072	1.053207721
	3	1.054635879	1.054979825	1.053115115	1.053827985	1.055026643	1.055991181
	4	1.052359705	1.052499743	1.052461829	1.052371558	1.052472683	1.052731828
	5	1.053128974	1.052975352	1.052622534	1.05271571	1.052389137	1.052269601
	6	1.052631579	1.052667814	1.053193505	1.052408591	1.0521588	1.052563684

Data of "Product" Samples (Total: 180)

Lab. Code	Period	Measurement Cycle					
		1	2	3	4	5	6
		α = (U-235: U-238) $_{\text{test material}}$ /(U-235: U-238) $_{\text{reference material}}$					
2	1	1.061066977	1.061476337	1.061652241	1.061773068	1.060417205	1.060211791
	2	1.062575903	1.06264783	1.062162421	1.062072492	1.062635817	1.063331627
	3	1.063284718	1.063147884	1.063572085	1.062767339	1.063117871	1.062539602
	4	1.060622576	1.061543889	1.062965579	1.063140315	1.063080135	1.063634319
	5	1.063146067	1.063091989	1.062796969	1.063206215	1.063963761	1.064520275
	6	1.06227952	1.063125481	1.062704366	1.06294693	1.062626198	1.062956789
4	1	1.0621	1.0621	1.0621	1.0622	1.0619	1.062
	2	1.0623	1.0621	1.0625	1.0625	1.0623	1.0625
	3	1.0628	1.0627	1.0627	1.0627	1.0625	1.0628
	4	1.0622	1.0625	1.062	1.0617	1.0621	1.0623
	5	1.0624	1.0623	1.0623	1.0622	1.0624	1.0628
	6	1.0625	1.0625	1.0625	1.0624	1.0627	1.0625
6	1	1.063144398	1.062939858	1.062872072	1.06274669	1.063066415	1.062860775
	2	1.062759114	1.062864164	1.062765891	1.062898055	1.062773797	1.062903704
	3	1.063057374	1.062913872	1.062698127	1.062708291	1.06292743	1.062894666
	4	1.062988443	1.062937598	1.062886758	1.062828015	1.062968105	1.06292856
	5	1.062856256	1.062741043	1.062733137	1.062602143	1.062797517	1.062751208
	6	1.062963585	1.062893536	1.062781704	1.062807683	1.062919521	1.062839311
8	1	1.062455226	1.062406407	1.062375883	1.062434896	1.062536623	1.062408438
	2	1.062613843	1.062122099	1.062233783	1.06206919	1.062418726	1.062290712
	3	1.062593581	1.062162514	1.062178862	1.061703511	1.062162689	1.062112003
	4	1.062565104	1.06211362	1.061961295	1.061890204	1.062725823	1.062723781
	5	1.062214984	1.061893283	1.062204853	1.061345496	1.061481819	1.061889251
	6	1.062640179	1.061777422	1.061597567	1.062692111	1.062311396	1.062586301
9	1	1.062343306	1.062520896	1.062648426	1.06273001	1.062665283	1.062625586
	2	1.06245825	1.062625251	1.062520896	1.062520896	1.062353728	1.062520896
	3	1.062635711	1.062813231	1.062479118	1.062479118	1.06245825	1.062479118
	4	1.062614794	1.062792251	1.062635711	1.062646174	1.062834225	1.0625
	5	1.062667112	1.06273001	1.06256273	1.062512549	1.06256273	1.062217762
	6	1.06289729	1.062426778	1.062583668	1.062573197	1.06289729	1.06256273

Annex III:

Definitions of the Statistical Terms Used

Definitions of the Statistical Terms Used

1. The Ratio of Ratios $\alpha = (\text{U-235:U-238})_{\text{test material}} / (\text{U-235:U-238})_{\text{reference material}}$

The ratios of the isotopic ratios

$$\alpha = \frac{(\text{U-235: U-238})_{\text{test material}}}{(\text{U-235: U-238})_{\text{reference material}}}$$

given in Annex II and calculated from the original data reported by the laboratories (Annex I) are the basic data for all statistical considerations.

Y_{ijk} represents the value of α obtained by laboratory i in the measurement cycle k of the period j.

For each type of sample ("Tails", "Feed" and "Product"), the number of laboratories is given by

$i = 1 \ldots I$ with $I \leqq 10$ corresponding to the number of laboratories considered,

the number of measurement periods per laboratory by
$j = 1 \ldots p$ with $p = 6$

and the number of measurement cycles per measurement period by
$k = 1 \ldots c$ with $c = 6$.

2. Period Means and Related Terms

The mean value of α obtained by laboratory i in the measurement period j is defined by

$$Y_{ij.} = \frac{1}{c} \sum_{k=1}^{c} Y_{ijk} \qquad (1)^{*)}$$

Therefore, the standard deviation (SD) of a single measurement cycle of this period is given by

$$\sigma(i,j) = \frac{1}{c-1} \sqrt{\sum_{k=1}^{c} (Y_{ijk} - Y_{ij.})^2} \qquad (2)$$

and the corresponding relative standard deviation (RSD) by

$$s(i,j) = \frac{\sigma(i,j)}{Y_{ij.}} \quad . \, 100 \, [\%] \qquad (3)$$

The SD and RSD of the period mean itself are expressed by

$$\sigma\,(PM) = \frac{\sigma(i,j)}{\sqrt{c}} \qquad (4)$$

and

$$s(PM) = \frac{s(i,j)}{\sqrt{c}} \; [\%] \qquad (5)$$

3. Laboratory Means and Related Terms

In the case of orthogonality, as in this experiment, the following relations are valid:

The mean value of a obtained by laboratory i can be calculated by

$$Y_{i..} = \frac{1}{p} \sum_{j=1}^{p} Y_{jk.} \qquad (6)$$

SD and RSD of this laboratory mean value are given by

$$\sigma(LM) = \sqrt{\frac{1}{p(p-1)} \sum_{j=1}^{p} (Y_{ij.} - Y_{i..})^2} \qquad (7)$$

and

$$s(LM) = \frac{\sigma\,(LM)}{Y_{i..}} \quad . \, 100 \, [\,\%\,] \qquad (8)$$

---

*) Substitution of a subscript by a point indicates averaging of the corresponding component.

4. Grand Mean and Related Terms

In the case of orthogonality, as in this experiment, the following relations are valid:

The mean value of α calculated from the measurements of all laboratories ("grand mean") is given by

$$Y_{...} = \frac{1}{I} \sum_{i=1}^{I} Y_{i..} \tag{9}$$

SD and RSD of this grand mean are defined by

$$\sigma(GM) = \sqrt{\frac{1}{I\,(I-1)} \sum_{i=1}^{I} (Y_{i..} - Y_{...})^2} \tag{10}$$

and

$$s(GM) = \frac{\sigma(GM)}{Y_{...}} \,.\, 100\ [\%] \tag{11}$$

5. Analysis of Variances

A scheme of the analysis of variances is given in Fig. III-1.

The theory is based on the assumption that the total error of a measurement is composed by addition of single error components. Therefore, in the case of three components, the equation

$$Y_{ijk} = \mu + a_i + b_{ij} + e_{ijk} \tag{12}$$

is obtained. Y_{ijk} is the measurement value of α obtained by laboratory i in the measurement cycle k of the period j, μ is the true (but unknown) value of α, a_i, b_{ij} and e_{ijk} are the single error components.

By analysis of variances, estimates $\sigma(a)^2$, $\sigma(b)^2$ and $\sigma(e)^2$ can be calculated for the variances of these three error components. Regarding the rather extended formalism of these calculations, reference is made to literature [7,8]. It is the basis for the "HP3" computer program printed out in Annex IV.

In order to facilitate understanding in the present report, the relative standard deviations S derived from the estimates of these variances are marked by the subscripts L ("Laboratory"), P ("Period") and C ("Cycle").

In the case of orthogonality, as in this experiment, the estimate for the total error of a mean value calculated from the data of l laboratories, each of them performing p measurement periods comprising c cycles each, is expressed by

$$S_{total} = \sqrt{\frac{S_L{}^2}{l} + \frac{S_P{}^2}{l \cdot p} + \frac{S_C{}^2}{l \cdot p \cdot c}} \qquad (13)$$

This corresponds to the term given in equation 11.

If the laboratory error component a_i is disregarded, the simplified version

$$Y_{ik} = \mu + b_j + e_{jk} \qquad (14)$$

of formula 12 is obtained.

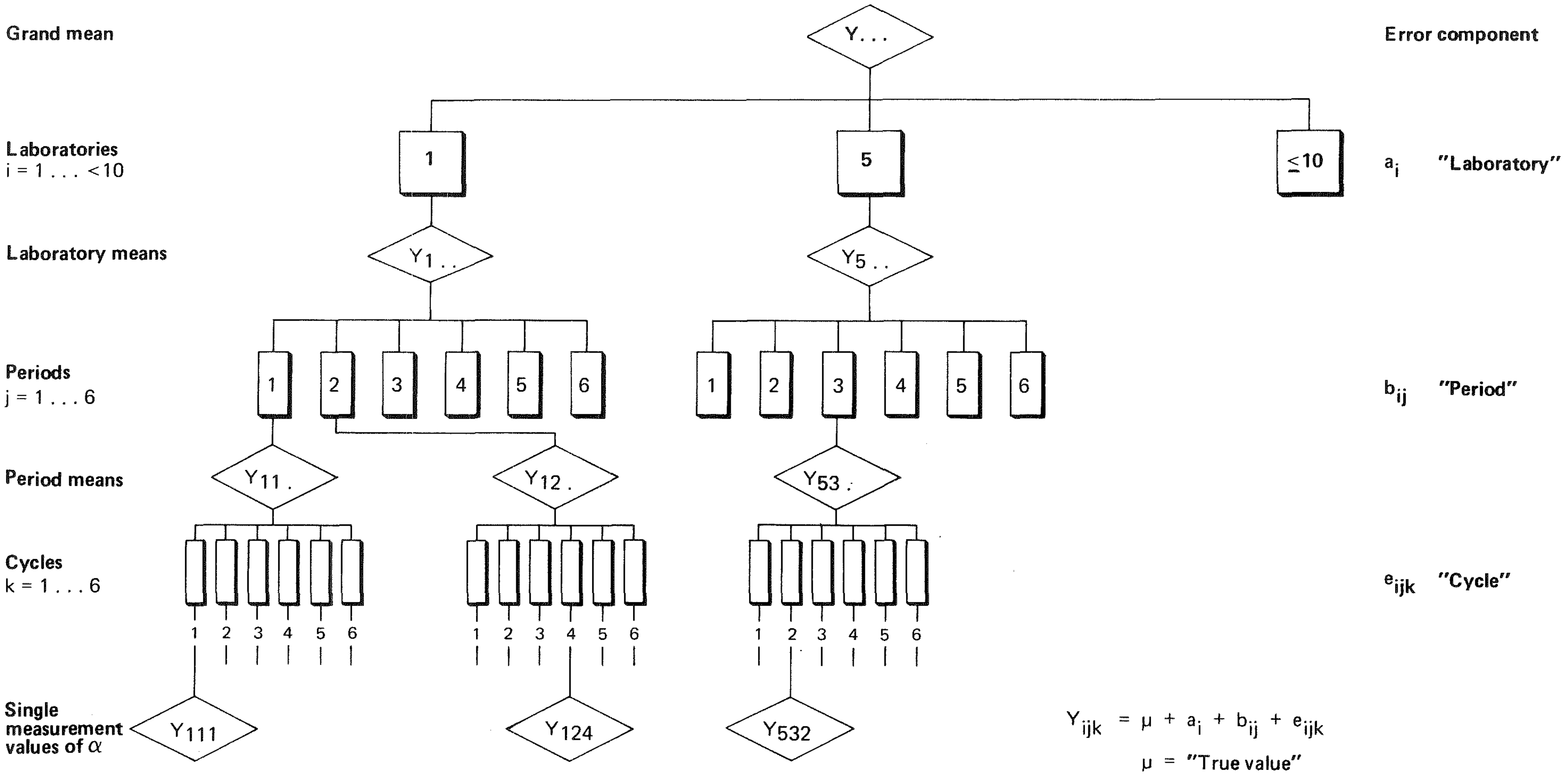
In this case, the estimates of the variances for the two error components b ("Period") and e ("Cycle") are given by

$$\sigma(b)^2 = \sigma_P{}^2 = \frac{1}{c}\left[\frac{c\sum_{j=1}^{p}(Y_{j.}-Y_{..})^2}{(p-1)} - \frac{\sum_{j=1}^{p}\sum_{k=1}^{c}(Y_{jk}-Y_{j.})^2}{p(c-1)}\right] \qquad (15)$$

and

$$\sigma(e)^2 = \sigma_C{}^2 = \frac{\sum_{j=1}^{p}\sum_{k=1}^{c}(Y_{jk}-Y_{j.})^2}{p(c-1)} \qquad (16)$$

Calculation of the SD's and RSD's of the error components was made on the basis of these formulae by use of "ZWEI" program, Annex IV.



$$Y_{ijk} = \mu + a_i + b_{ij} + e_{ijk}$$

μ = "True value"

Fig. III - 1: Scheme of the analysis of variances

Annex IV:

Computer Programs

Computer Programs

Three computer programs were elaborated for the numerical calculations using APL/360 ("**A P**rogramming **L**anguage").

"EINS" program allows the calculation of terms as given in formulae 1 to 11 of Annex III, "ZWEI" and "HP3" programs refer to the analysis of variances with two or three error components, respectively (see Annex III, 5).

For details of the programming language reference should be made to "APL/360 User's Manual, IBM Form GH 20-0683" and "APL/360 Primer, IBM Form GH 20-0689".

Programs and Subprograms

```
      ∇ EINS
[1]    MI←(+/XI)÷N
[2]    VAR←+/((XI-MI)*2)÷(N-1)
[3]    SGM←VAR*0.5
[4]    SGR←100×SGM÷MI
[5]    SGMM←SGM÷N*0.5
[6]    SGMMR←100×SGMM÷MI
[7]    SRM←SGR÷N*0.5
[8]    MI
[9]    SGM
[10]   SGR
[11]   N
[12]   SGMM
[13]   SRM
      ∇
```

```
      ∇ ZWEI
[1]    YM← 6 6 ρY
[2]    ZS←+/[2] YM
[3]    PMI←ZS÷6
[4]    S←6
[5]    PMM←(6,S)ρPMI
[6]    PMT← 2 1 ⍉PMM
[7]    N←ρYM
[8]    N←×/N
[9]    SY←+/ZS
[10]   C←S×36÷N
[11]   K←(N-C)÷(S-1)
[12]   D←YM-PMT
[13]   D2←D*2
[14]   ZD2←+/[2] D2
[15]   SQR←+/ZD2
[16]   VRE←SQR÷(N-S)
[17]   SIGE←VRE*0.5
[18]   YL←+/Y÷N
[19]   SE←SIGE×100÷YL
[20]   DD←(PMI-YL)*2
[21]   DDS←+/DD
[22]   SQA←S×DDS
[23]   T1←SQA÷(S-1)
[24]   T2←SQR÷(N-S)
[25]   VRA←|(T1-T2)÷K
[26]   SIGA←VRA*0.5
[27]   SA←SIGA×100÷YL
[28]   PMI
[29]   YL
[30]   SE
[31]   SA
      ∇
```

```
      ∇ HP3
[1]    DATA
[2]    ZS←+/[2] YM
[3]    ZSQ←ZS*2
[4]    LS←+/S
[5]    N←+/VN
[6]    RQ←36×AP
[7]    D←+/RQ
[8]    RN←RQ÷VN
[9]    E←+/RN
[10]   FN←VN×VN
[11]   F←+/FN
[12]   YMM←YM*2
[13]   YMMV←,YMM
[14]   G←+/YMMV
[15]   HH←ZSQ÷6
[16]   H←+/HH
[17]   GM←LS÷N
[18]   II←(S*2)÷VN
[19]   I←+/II
[20]   FA←R-1
[21]   K11←(H-F÷N)÷FA
[22]   K12←(E-D÷N)÷FA
[23]   ZPG←+/AP
[24]   FB←ZPG-R
[25]   FR←N-ZPG
[26]   K22←(H-E)÷FB
[27]   LSQ←LS*2
[28]   SQA←I-LSQ÷N
[29]   SQB←H-I
[30]   SQR←G-H
[31]   MQA←SQA÷FA
[32]   MQB←SQB÷FB
[33]   MQR←SQR÷FR
[34]   VRE←MQR
[35]   SIGE←VRE*0.5
[36]   SE←SIGE×100÷GM
[37]   VRB←(MQB-MQR)÷K22
[38]   SIGB←VRB*0.5
[39]   SB←SIGB×100÷GM
[40]   A←K12÷K22
[41]   B←A×MQB
[42]   C←1-A
[43]   C1←C×MQR
[44]   VRA←(MQA-B-C1)÷K11
[45]   SIGA←VRA*0.5
[46]   SA←SIGA×100÷GM
[47]   ERG3
      ∇
```

Subprogram "Data"

(Example feed)

```
      ∇ DATA
[1]     LAB1
[2]     LAB2
[3]     LAB3
[4]     LAB4
[5]     LAB5
[6]     LAB6
[7]     LAB7
[8]     LAB8
[9]     LAB9
[10]    LAB10
[11]    YM←Y1M,[1] Y2M,[1] Y3M,[1] Y4M,[1] Y5M,[1] Y6M,[1] Y7M,[1] Y8M,[1] Y9M,[1] Y0M
[12]    S←S1,S2,S3,S4,S5,S6,S7,S8,S9,S0
[13]    VN←N1,N2,N3,N4,N5,N6,N7,N8,N9,N0
[14]    R←⍴VN
[15]    AP← 6 6 6 6 6 6 6 6 6 6
      ∇
```

Subprogram "ERG 3"

```
      ∇ ERG3
[1]     ⎕←'Y1M'
[2]     Y1M
[3]     ⎕←'Y2M'
[4]     Y2M
[5]     ⎕←'Y3M'
[6]     Y3M
[7]     ⎕←'Y4M'
[8]     Y4M
[9]     ⎕←'Y5M'
[10]    Y5M
[11]    ⎕←'Y6M'
[12]    Y6M
[13]    ⎕←'Y7M'
[14]    Y7M
[15]    ⎕←'Y8M'
[16]    Y8M
[17]    ⎕←'Y9M'
[18]    Y9M
[19]    ⎕←'Y0M'
[20]    Y0M
[21]    ⎕←TEXT6←'STANDARDABWEICHUNG DER REPRODUZIERBARKEIT=',⍕SIGE
[22]    ⎕←TEXT7←'RELATIVE STANDARDABWEICHUNG DER REPRODUZIERBARKEIT=',(⍕SE),'PROZENT'
[23]    ⎕←TEXT9←'STANDARDABWEICHUNG DER PERIODEN=',⍕SIGB
[24]    ⎕←TEXT10←'RELATIVE STANDARDABWEICHUNG DER PERIODEN=',(⍕SB),'PROZENT'
[25]    ⎕←TEXT11←'SCHAETZWERT DER VARIANZ DER LABORS=',⍕VRA
[26]    ⎕←TEXT12←'STANDARDABWEICHUNG DER INTERLABABWEICHUNG=',⍕SIGA
[27]    ⎕←TEXT13←'RELATIVE STANDARDABWEICHUNG DER INTERLABABWEICHUNG=',(⍕SA),' PROZENT'
      ∇
```

Annex V:

"The Characterization of Test Materials for the GfK/Dornier Interlab Test on UF_6"

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(October 1976)

Note:
For particular reasons, CBNM considered the "Test Materials" of the Interlab Test to be the "Reference Materials" and vice versa. Therefore, the data of a presented in this Annex correspond to $1/a$ in the other parts of this report.

1. Introduction

Plans for the establishment of UF_6 Isotopic Reference Materials (IRM) had been conceived at the CBNM Mass Spectrometry for some time in order to have available a set of UF_6 Safeguard IRM's which would serve as common references for all measurements of UF_6 samples taken for verification purposes by EEC Safeguard Authorities when inspecting EEC enrichment installations. The philosophy hereby is that these IRM's should also be made available directly to plant operators to refer their own measurements to. This will contribute to minimize discrepancies between declared and verified contents of fissionable isotope. These IRM's will also be continuously intercompared with any existing RM which might exist or come into existence in various countries. In fact thousands of measurements have already been performed continuously since 1973 at an ultracentrifuge enrichment plant in the EEC against UF_6 batches characterized at CBNM for isotopic composition.

Additional support for the IRM's came in July 1974 when UF_6 measuring laboratories connected with Ultracentrifuge and "Trenndüse" development or exploitation, decided to determine their ability to accurately measure isotopic differences between two materials (i.e. "ratio differences" or "ratios of ratios").

For this purpose an "Interlab Test" (IT) was estimated to be necessary. Complete isotopic characterization of 3 pairs hence six Isotopic Test Materials (ITM) for the IT was expected from CBNM:

- 1 pair of ITM's around 3 % ^{235}U abundance:
  3.1 and 3.3 % ("product")
- 1 pair of ITM's around 0.7 % ^{235}U abundance:
  0.68 and 0.72 % ("feed" i.e. natural Uranium)
- 1 pair of ITM's around 0.4 % ^{235}U abundance:
  0.46 and 0.44 % ("tails")

Discussions at the meeting of participating laboratories (July 1974) quickly resulted in a concept where one material per pair would be established as an IRM with fully characterized isotopic composition, the other remaining an Isotopic Test Material (ITM). On the one hand the three IRM's - one per pair - would in the future be held available from CBNM. On the other hand the Interlab Test would yield a picture of how good laboratories could measure the ratio of IRM and ITM in each pair. Consequently laboratories could in the future make their daily measurements of unknown samples against these IRM's with an "a priori" knowledge of their ability to measure the ratio. It was expected that this would also minimize discrepancies between plant "declaration" values and inspection's "verification" values on the occasion of safeguard inspections.

Finally the expressed need specified "that the materials would be welcomed ready for direct use as daily working standards and should be directly connectable to the measurement installations at the plants i.e. be available in ampoules with a valve and standardized connection".

This report brings the result of the characterization of

a) the three IRM's:
one at 3.3 % ^{235}U abundance
one at 0.72 % ^{235}U abundance
one at 0.44 % ^{235}U abundance

b) the three ITM's:
one at 3.1 % ^{235}U abundance
one at 0.68 % ^{235}U abundance
one at 0.46 % ^{235}U abundance

c) the interval IRM - ITM expressed for the three pairs as the ratio a of the ^{235}U/^{238}U ratio in the ITM to the ^{235}U/^{238}U ratio in the IRM i.e.

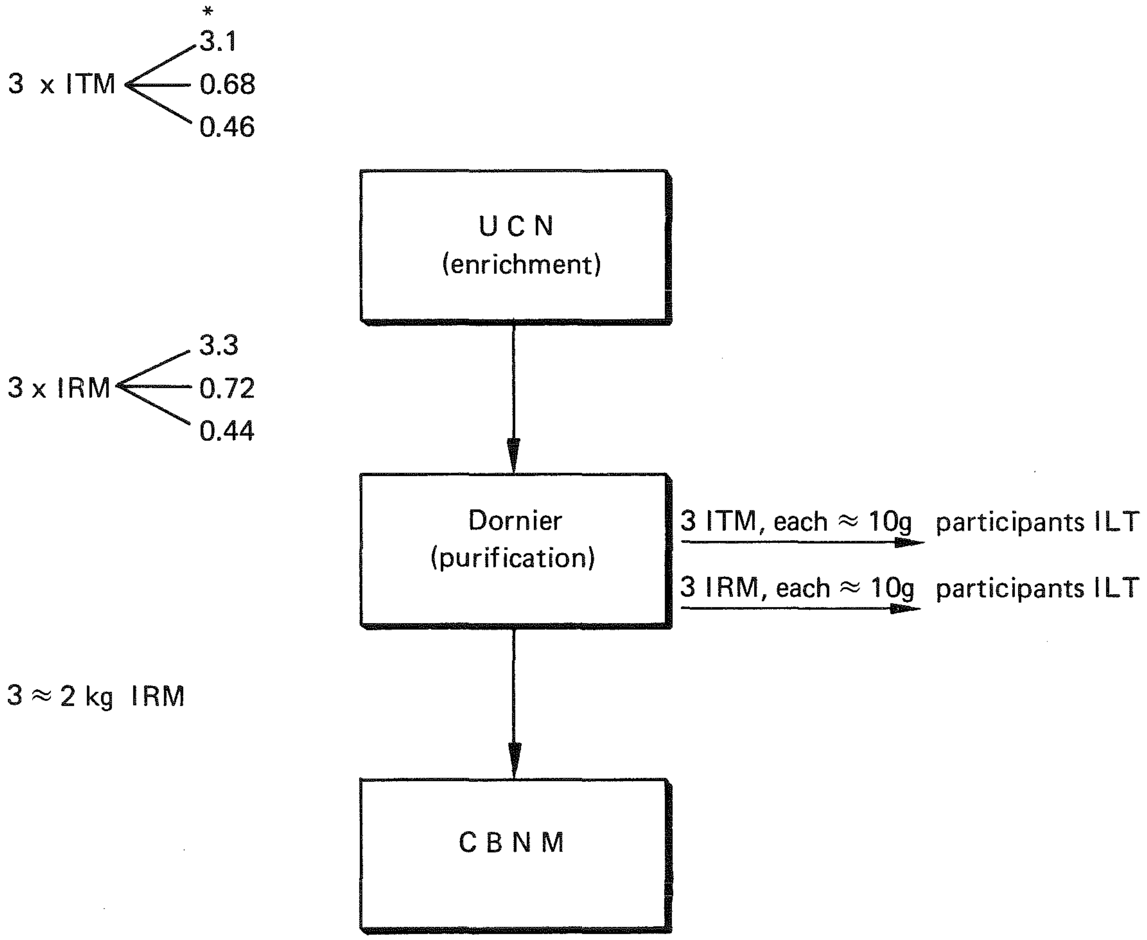
"Product" pair with $a = \frac{0.031}{0.033}$

"Feed" pair with $a = \frac{0.0068}{0.0072}$

"Tails" pair with $a = \frac{0.0046}{0.0044}$

2. Preparation of the Materials for Isotopic measurements

About 2 kg of each of the three IRM's were purchased from the Ultracentrifuge enrichment plant, ECN, Almelo (see Fig. V-1).



* ^{235}U enrichment

Fig. V - 1: Flow scheme of ITM's and IRM's

They were withdrawn from the Ultracentrifuges so as to provide materials truly representative for this step in the nuclear fuel cycle. They were purified by distillation and homogenized at DornierFriedrichshafen, and transported to CBNM. All transports were organized and performed by CBNM services in specially made Monel ampoules equipped with valve and NW 10 connection.

Small ampoules containing about 10 g of each of the three ITM's were also received from Dornier who distributed similar ITM samples to all participants in the Interlab Test. Each participant was also provided with a small sample of the IRM's directly by Dornier.

At CBNM all IRM's and ITM's were measured by UF_6 Mass Spectrometry and fractions of the batches hydrolyzed and converted to solid (see Fig. V-2), a small part of which was dissolved in HNO_3 and a standardized Uranyl nitrate solution made up for Thermionic Mass Spectrometry (ThMS). The larger part of the solid was fluorinated back to UF_6 by CoF_3. This UF_6 was then checked by UF_6 Mass Spectrometry against the starting UF_6 for isotopic identity. This check was intended to make sure that no isotopic contamination in the conversion to U_3O_8 and Uranyl nitrate solutions would invalidate the comparison of the ThMS results with UF_6 MS stage "A" measurements (see again Fig. V-2).

The hydrolysis consisted of following steps:

a) distill ~ 3 g UF_6 from room temperature into a connected ampoule at -196° C (30 seconds)
b) heat the sample to -80°C and pump off impurities
c) cool again to -196°C and add 10 ml H_2O
d) after reaction is over, evaporate solution to dryness
e) dry residue in oven at 350°C for 4 hours
f) dissolve residue in excess HNO_3, evaporate to dryness and dry residue again at 350°C in oven overnight.
The sample for ThMS is taken and separately calcined at 900°C for 1 hour
g) the fraction to be fluorinated is mixed with CoF_3 (a factor 2.3 excess by weight), put into a glass ampoule and degassed under vacuum at 200°C
h) the ampoule is heated at 425°C for 70 minutes and the UF_6 formed is condensed at -196°C
i) all gases are pumped off with the UF_6 condensed at -196°C
j) the UF_6 is heated to -80°C and gases pumped off until a pressure of 1×10^{-4} Torr or less is reached
k) the UF_6 is again condensed at -196°C and transported in a closed (sealed) ampoule to the mass spectrometer for measurement.

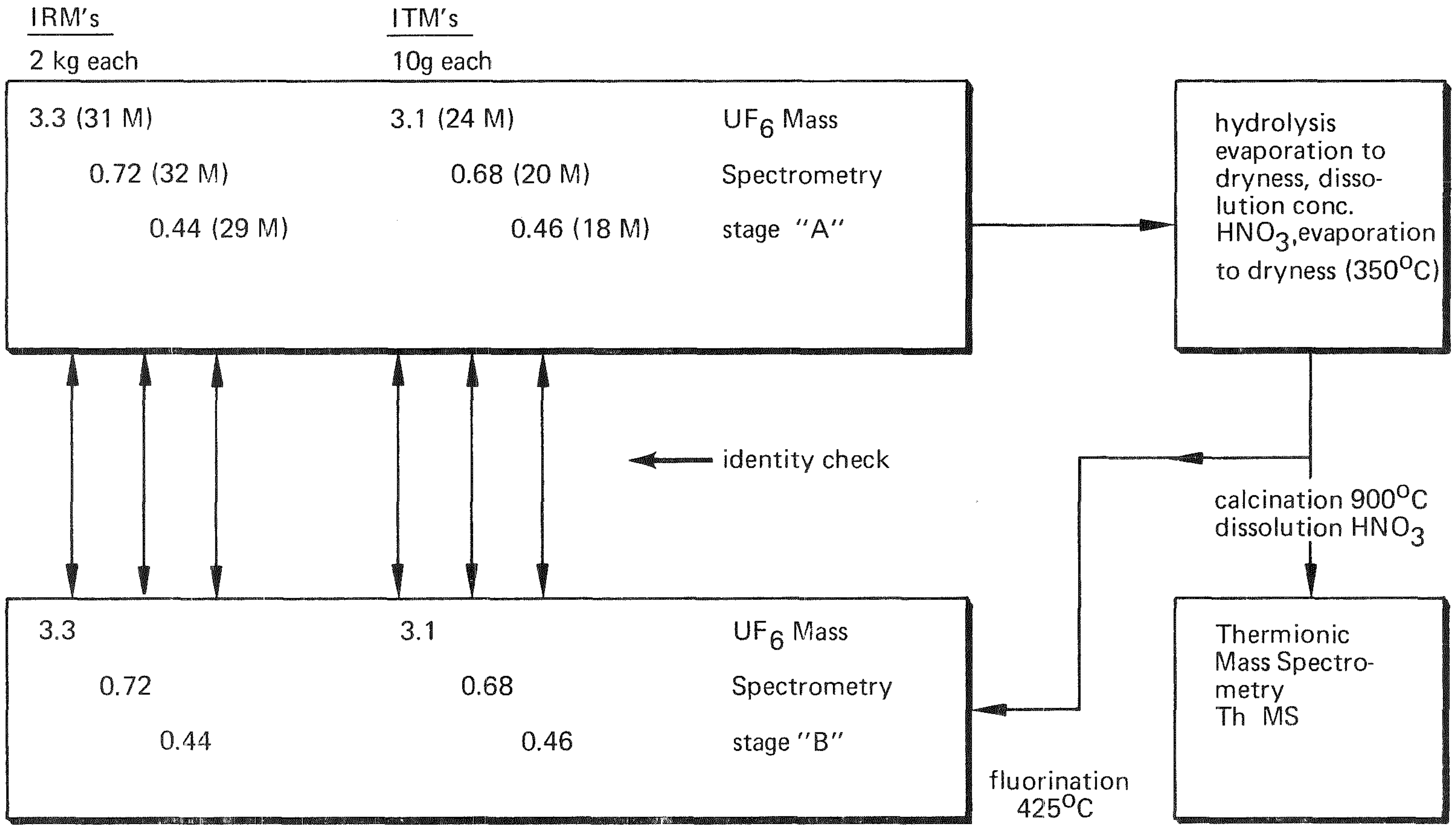


Fig. V - 2: Flow scheme for preparations and measurements of IRM's and ITM's.
Figures indicate approximate ^{235}U enrichments in % with ampoule identification numbers between brackets.

3. Isotopic measurements

3.1 UF_6 mass spectrometric measurements were performed with a Varian MAT 511 UF_6 Mass Spectrometer equipped with a permanent magnet, a double collector for direct $^{235}U/^{238}U$ ratio measurements and a GA 18/30 Computer which takes care of data taking and reduction, as well as of the operation of the pneumatic inlet and outlet gas valves of the sources. Two liquid nitrogen traps extending into the ion source keep the source pressure below 1×10^{-6} Torr. In addition, one of them traps the UF_6 molecular beam immediately after it has gone through the ionizing region of the source. Source and traps were taken apart and cleaned when Relative Standard Deviation of α - measurements * increased to 5×10^{-4} (about every 2 – 3 months). For a measurement two ampoules were connected to the inlet system (NW 10). Prior to each measurement the ampoules were cooled to -40°C and evacuated for possible impurities, heated again to room temperature at which point gas inlet into the mass spectrometer was started. Following sequence for the i-th analysis for each pair was used:

		period (in seconds)		
A.	Ampoule n^o 1 (ITM)			
	gas inlet into ion source:	30''		
	measurement of 40 235/238 ratios R:	60''	→	mean R_{i1}
	pump off:	45''		
	zero measurements:	30''		
B.	Ampoule n^o 2 (IRM)			
	gas inlet into ion source:	30''		
	measurement of 40 235/238 ratios R:	60''	→	mean R_{i2}
	pump off:	45''		
	zero measurements:	30''		
C.	Ampoule n^o 1 (ITM)			
	id.		→	mean R_{i3}
D.	Ampoule n^o 2 (IRM)			
	id.		→	mean R_{i4}
	etc.			etc. to R_{i9}

---

* $$\alpha = \frac{(^{235}U\,/\,^{238}U)\ \text{ITM}}{(^{235}U\,/\,^{238}U)\ \text{IRM}}$$

a-values were then determined as

$$\frac{R_{i1} + R_{i3}}{2\,R_{i2}} = a_{i1} \qquad \frac{R_{i3} + R_{i5}}{2\,R_{i4}} = a_{i2}$$

$$\frac{R_{i5} + R_{i7}}{2\,R_{i6}} = a_{i3} \qquad \frac{R_{i7} + R_{i9}}{2\,R_{i8}} = a_{i4}$$

The entire operation is called a "UF_6 analysis" yielding one value for

$$a_i = \frac{1}{4} \sum_{k=1}^{4} a_{ik}$$

For all measurements in this report, 6 "UF_6 analysis" were performed, yielding six a-values a_i (i = 1 . . . 6), a mean value $\bar{a}$ and a standard deviation

$$s_{\bar{a}} = \sqrt{\frac{\sum_{i=1}^{6} (\bar{a} - a_i)^2}{6 - 1}}$$

3.2 Thermionic measurements were performed on 1 mg U/ml solutions on a 12"-90° Teledyne Mass Spectrometer, equipped with a Cary Vibrating Reed Electrometer, Model 401 MR using a 10^{11} ohm input resistor. All measurements were performed using a Faraday cage exclusively. The VRE output was fed through a Philbrick VF converter into a HP 9830 desk computer which also operated the VRE attenuator ranges and a Varian FR-41 magnetic field controller. This enabled jumping from ^{235}U to ^{238}U peak tops according to an operator chosen schedule. Minor isotopes were measured by conventional magnetic field scanning and recording on a L.N.-recorder equipped with a facility for expanded-scale measurements.

4. Characterization of the Pair Intervals

4.1 Direct ratio of ratios measurement for each pair by UF_6 Mass Spectrometry

Using the measurement procedure described in 3.1 following results were obtained:

"Product" Pair	"Feed" Pair	"Tails" Pair
$\frac{24\text{ M}}{31\text{ M}} = 0.940867$	$\frac{20\text{ M}}{32\text{ M}} = 0.949509$	$\frac{18\text{ M}}{29\text{ M}} = 1.051739$
± 0.000061	± 0.000060	± 0.000052
(n = 6)	(n = 6)	(n = 6)

These α-values are listed in Table V-2 and Fig. V-3 as first α-value in each pair interval.

4.2 Measurement of each pair member against two known Isotopic Reference Materials by UF_6 Mass Spectrometry

Each IRM and TM of a pair was measured against two Standard Reference Materials with different ^{235}U enrichments:

- one as close to the pair as possible
- one other up to 50 % off (i.e. $\alpha \sim 1.5$).

(Where required these Reference Materials had to be converted from U_3O_8 to UF_6). The results are summarized hereafter in Table V-1.

From these measurements calculated values α for each pair interval are derived. They are listed as second and third value in each pair interval in Table V-2 and Fig. V-3.

4.3 Measurement of Pair Interval by Thermionic Mass Spectrometry

The $^{235}U/^{238}U$ ratios measured are given in Section 5.1.

Fig. V-4 shows how "Product pair", "Feed pair" and "Tails pair" measurements were converted to "absolute" values by correction with a carefully determined factor defined as $R_{theor.}/R_{observed}$. It is the correction factor for isotope fractionation effects. It was determined to be 0.9956 from repeated measurements of NBS-005, NBS-020 and NBS-030.

Table V - 1:

Pair	a	s_a (n = 6)
"Product"		
^{24M}/NBS 030	1.013 769	0.000 044
^{31M}/NBS 030	1.077 520	0.000 100
^{24M}/31 M	0.940 835	0.000 106
^{24M}/NBS 020	1.532 539	0.000 170
^{31M}/NBS 020	1.628 814	0.000 062
^{24M}/31 M	0.940 893	0.000 110
"Feed"		
^{20M}/NBS 950 a	0.949 092	0.000 052
^{32M}/NBS 950 a	0.999 660	0.000 051
^{20M}/32 M	0.949 415	0.000 071
^{20M}/NBS 005	1.401 257	0.000 121
^{32M}/NBS 005	1.476 106	0.000 154
^{20M}/32 M	0.949 293	0.000 129
"Tails"		
^{18M}/NBS 005	0.940 915	0.000 064
^{29M}/NBS 005	0.894 688	0.000 028
^{18M}/29 M	1.051 668	0.000 078
^{18M}/NBS 950 a	0.637 328	0.000 058
^{29M}/NBS 950 a	0.605 962	0.000 082
^{18M}/29 M	1.051 762	0.000 172

Table V - 2: a - values of pair intervals

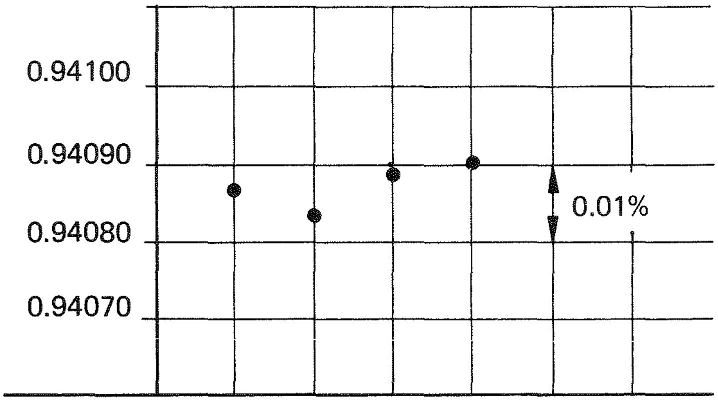
Pair Interval	a	s_a (n = 6)	s_a %	Standard Reference Material if used
"Product" 24M/31M	0.940 867	0.000 061	0.006	direct UF_6 MS
	0.940 835	0.000 106	0.01	NBS 030 UF_6 MS
	0.940 893	0.000 110	0.01	NBS 020 UF_6 MS
	0.940 90		0.15	Thermionic MS
"Feed" 20M/32M	0.949 509	0.000 060	0.006	direct UF_6 MS
	0.949 415	0.000 071	0.007	NBS 950a UF_6 MS
	0.949 293	0.000 129	0.014	NBS 005 UF_6 MS
	0.949 41		0.15	Thermionic MS
"Tails" 18M/29M	1.051 739	0.000 052	0.005	direct UF_6 MS
	1.051 668	0.000 078	0.007	NBS 005 UF_6 MS
	1.051 762	0.000 172	0.016	NBS 950a UF_6 MS
	1.051 68		0.15	Thermionic MS

"Product" Pair

0.940867
0.94085
0.940835

0.940893 *

0.94090 **

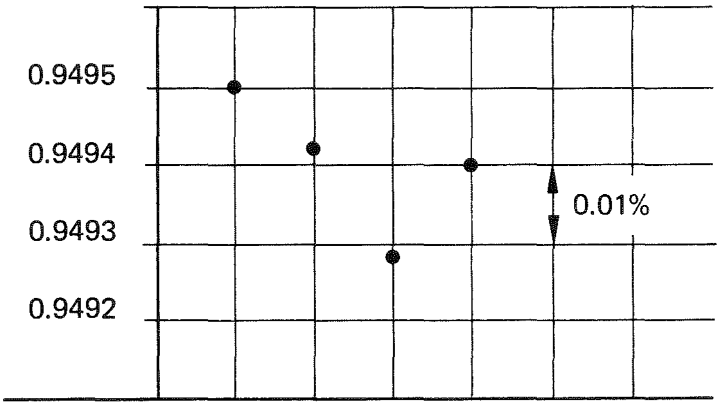


"Feed" Pair

0.949509
0.94946
0.949415

0.949293 *

0.94941 **

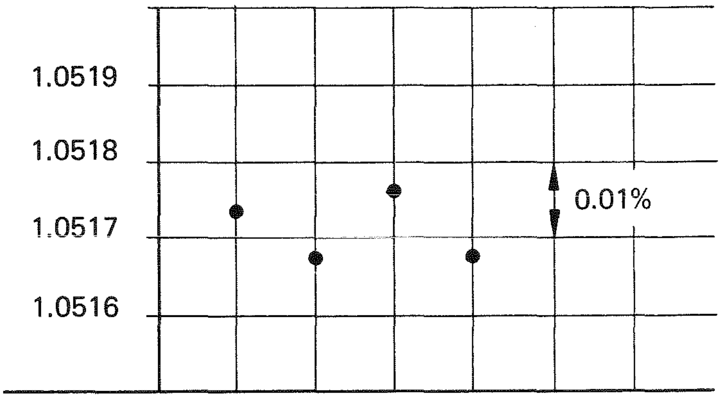


"Tails" Pair

1.051739
1.05170
1.051668

1.051762 *

1.05168 **



* Considering the fact that this is a ratio of α-values around 1.5 (or 0.6, which is = 1/1.5), the agreement with a ratio of α-values in the region 0.9/1.07 is gratifying. Nevertheless the values carries less weight because of the large α-values and is considered as having confirmatory value only.

** As Thermionic Mass Spectrometry is intrinsically less precise than UF_6 Mass Spectrometry, these values have confirmatory character only. However the agreement is gratifying.

Fig. V - 3: Summary of Pair interval characterization

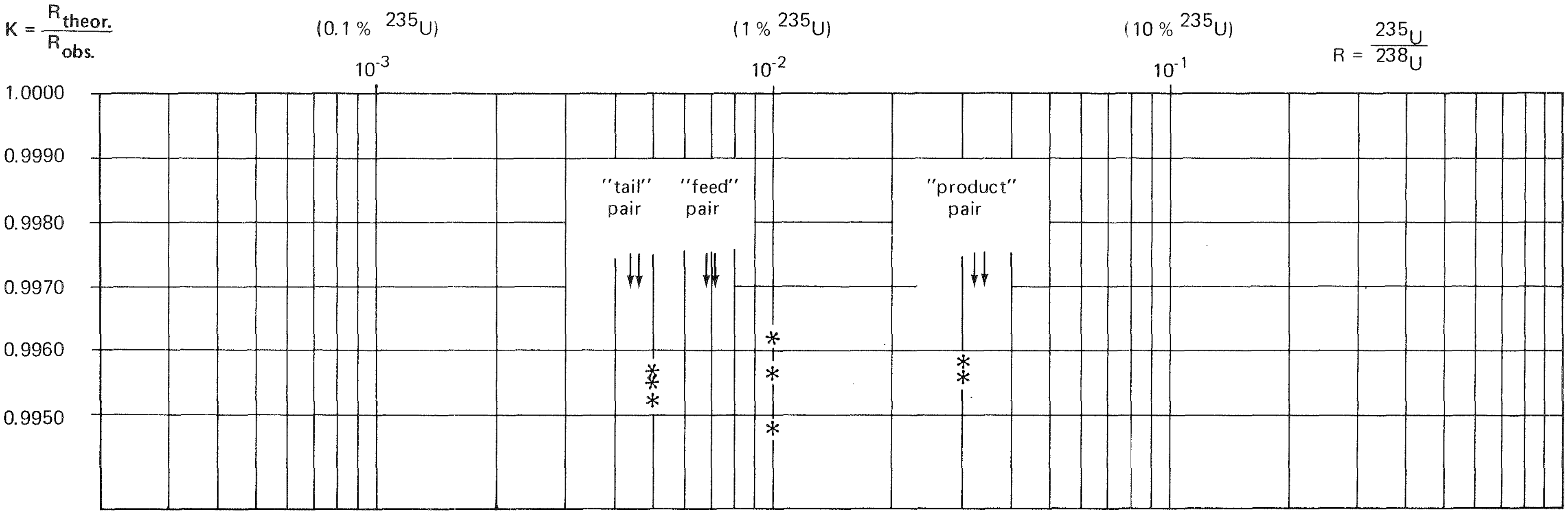


Fig. V - 4: Calibration of the Thermionic measurements

5. Characterization of complete Isotopic Composition by Thermionic Mass Spectrometry

Using measurement procedures given in 3.2, ratios of all isotopes were measured. Results are summarized in 5.1 and derived isotopic compositions in 5.2.

5.1 Measured isotope ratios

	$^{234}U/^{235}U$	$^{236}U/^{235}U$	observed $^{235}U/^{238}U$	mean
"Product"				
24 M	0.00997	$<$ 0.0001 (det. lim.)	0.031818 0.031847	0.031833
31 M	0.01001	"	0.033844 0.033820	0.033832
"Feed"				
20 M	0.00724	"	0.006889 0.006885	0.006887
32 M	0.00732	"	0.007257 0.007251	0.007254
"Tails"				
18 M	0.00577	"	0.004629 0.004630	0.004629
29 M	0.00562	"	0.004403 0.004401	0.004402

5.2 Isotopic Compositions (Atom %)

"Product"	**24 M**	**31 M**	**Accuracy**
^{234}U	0.0307	0.0327	± 0.0007
^{235}U	3.0841	3.2714	± 0.0049
^{236}U	< 0.0001	< 0.0001	Detection Limit
^{238}U	96.8852	96.6959	± 0.0056

"Feed"	**20 M**	**32 M**	**Accuracy**
^{234}U	0.0050	0.0053	± 0.0003
^{235}U	0.6840	0.7201	± 0.0011
^{236}U	< 0.0001	< 0.0001	Detection Limit
^{238}U	99.3110	99.2746	± 0.0014

"Tails"	**18 M**	**29 M**	**Accuracy**
^{234}U	0.0027	0.0025	± 0.0002
^{235}U	0.4608	0.4383	± 0.0007
^{236}U	< 0.0001	< 0.0001	Detection Limit
^{238}U	99.5365	99.5592	± 0.0009

6. Some Remarks on the "Memory Effect"

In the measurement process described in Section 3.1, the Test Material and the Reference Material are alternately admitted to the ion source.

It is possible that the Mass Spectrometer "remembers" the previously introduced gas via sample condensation in adsorbed layers in the inlet system or ion source. Values for this "memory" given in the literature are dependant on the instrument used and may go up to several %.
It is to be noted that the measured difference in the $^{235}U/^{238}U$ ratios caused by memory is always diminished. If we define the so called memory factor by

$$M = \frac{(a - 1)\text{ real}}{(a - 1)\text{ observed}}$$

this means that the condition $M \geqslant 1$ is always fulfilled.
We tested the "memory" of our instrumentation over an extended range of ^{235}U enrichments and obtained the values summarized in Table V-3. These measurements using NBS standards do not yield memory factors (i.e. outside the precision level given in the last column).

7. Availability of the UF_6 Reference Materials

All three Reference Materials are available in 20 g amounts contained in a Monel ampoule equipped with a valve and an NW 10 connection.

Table V - 3: Memory effekt $$M = \frac{(\alpha - 1)\ \mathrm{NBS}}{(\alpha - 1)\ \mathrm{Obs.}}$$

NBS	$(\alpha - 1)$ NBS	$(\alpha - 1)$ Obs.	M	s
$\frac{005}{950\,\mathrm{a}}$	–0.321 798	–0.322 647	0.997 369	± 0.004
$\frac{010}{950\,\mathrm{a}}$	0.398 042	0.398 266	0.999 438	± 0.007
$\frac{015}{950\,\mathrm{a}}$	1.146 009	1.147 513	0.998 689	± 0.004
$\frac{020}{950\,\mathrm{a}}$	1.869 158	1.871 498	0.998 750	± 0.003
$\frac{030}{950\,\mathrm{a}}$	3.333 379	3.336 984	0.998 920	± 0.003
$\frac{050}{950\,\mathrm{a}}$	6.276 989	6.290 943	0.997 782	± 0.002
$\frac{010}{005}$	1.061 395	1.063 953 1.061 806	0.997 596 0.999 613	± 0.005 ± 0.006
$\frac{015}{005}$	2.164 261	2.168 856 2.171 068	0.997 881 0.996 865	± 0.004 ± 0.005
$\frac{020}{005}$	3.230 535	3.235 727 3.236 692	0.998 395 0.998 098	± 0.003 ± 0.003
$\frac{030}{005}$	5.389 510	5.403 527 5.404 455	0.997 406 0.997 235	± 0.004 ± 0.003
$\frac{050}{005}$	9.729 823	9.777 209 9.765 486	0.995 153 0.996 348	± 0.002 ± 0.002

Annex VI:

"Analytical Report and Description of Standard Measurement Procedures"

British Nuclear Fuels Ltd.
Mass Spectrometry Group
Capenhurst Works
Chester, England

BNFL
BRITISH NUCLEAR FUELS LIMITED
Capenhurst Works Chester Cheshire CH1 6ER
Telephone: 051-339 4101 Telegrams: NUFUEL Ellesmere Port Telex: 62202
Extension:

ANALYTICAL REPORT

Dr Walter Durr
Abt NTK
C/O Dornier System GmbH,
D-7990 Friedrichshafen,
Postfach 648
West Germany.

Material Order No.7666
Customers Order No.602/A3/339445/BERICH IASR

Sample No.	%atom U-235	%atom U-234	%atom U-236
13	0.4379 ± 0.0007	0.003 ± 0.0003	<0.001
28	0.4611 ± 0.0007	0.003 ± 0.0003	<0.001
54	3.273 ± 0.005	0.032 ± 0.003	<0.001
51	3.086 ± 0.005	0.030 ± 0.003	<0.001
11	0.719 ± 0.001	0.0045 ± 0.0005	<0.001
44	0.684 ± 0.001	0.0045 ± 0.0005	<0.001

The errors quoted are to 95% confidence limits and include the errors on our reference standards.

R. Tushingham.
pp W K Matthews
Technical Manager
Chemical & Metallurgical Services
28th October 1975

Standard Double Collection Procedure for U-235 Isotopic Analysis of Uranium Hexafluoride

1. INSTRUMENT

The mass spectrometer used for the U-235 isotopic analysis of uranium hexafluoride is a single focussing 90° sector magnet type, having an analyser tube radius of 12 inches (305 mm). It is fitted with a Nier type electron bombardment ion source modified for use with UF_6, and a double ion-collector system. The collector system enables any single ion beam in the mass range 15-400 A.M.U. to be collected by magnetic scanning or the UF_5 ions due to U-235 to be collected on one collector simultaneously to the UF_5 ions due to U-234, U-236 and U-238 being collected on the second collector. This latter method is used in the Double Collection Procedure.

The sample introduction system is so arranged to permit the alternate admission to the ion source of a sample and a reference standard which are normally contained in glass septums and are attached to the appropriate points by compression couplings. By suitable valve operations a pumped volume is always maintained between sample and standard gas flows. The flow of gas to the ion source is controlled by a very fine control needle valve.

2. MEASUREMENT PROCEDURE

The Double Collection procedure is used whereby the ratio R of the sum of the U-234, U-236 and U-238 isotopes to the U-235 isotope is measured. Basically, the method is a comparison technique in which the sample A and a reference standard B, having a U-235 concentration closely approximating to that of the sample, are alternately admitted to the ion source. The comparison of sample A with standard B is carried out in the sequence ABBA to a strict time schedule. This comprises one cycle of measurement and is used to determine the degree of enrichment n of the sample with respect to the standard. Each cycle provides two measurements of each of the ratios R_A and R_B, where

$$R = \frac{N234 + N236 + N238}{N235}$$

Suffixes A and B refer to the sample and standard respectively and N is the concentration of the uranium isotope of mass number indicated.

The measured degree of enrichment n_m is then calculated from

$$n_m = \frac{\bar{R}_B}{\bar{R}_A} - 1$$

where $\bar{R}_A$ and $\bar{R}_B$ are the means of the two values of R_A and R_B respectively determined in the cycle of measurement.

This measured degree of enrichment is converted to a true degree of enrichment n by multiplying by an instrument calibration factor. This factor is obtained by performing a series of similar measurements on two known standards. These standards are chosen to have U-235 concentrations which differ by approximately 10 % of the U-235 concentration and which bracket the U-235 concentration of the sample.

The U-235 concentration C of the sample is then calculated from the U-235 concentration C_s of the reference standard from the equation

$$\frac{C_1 - C_s}{C_s\,(1 - C_1)} = n$$

Ten cycles of measurement of the sample against the reference standard are made to produce a mean value $\bar{C}$ for the U-235 concentration of the sample.

The standard error ϵ of the mean is then calculated from

$$\epsilon = \sqrt{\frac{\sum_{x=1}^{10} (C_x - \bar{C})^2}{(10)\,(9)}}$$

The U-235 concentration of the sample and its precision is quoted as

$$\bar{C} \pm 2\epsilon$$

3. STANDARDS AND ACCURACY

The mass spectrometry techniques employed by BNFL require the use of accurate uranium isotopic reference standards. A programme, started approximately 15 years ago and continually expanded in succeeding years, has resulted in a range of some 80 isotopic standards to cover U-235 concentrations from 0.2 % to 4 %. Each standard is accurate to ± 0.1 % of the stated U-235 content.

The precision of measurement 2ϵ of the U-235 concentration of the sample is normally insignificant compared with the accuracy of the reference standard and so the U-235 concentration of the sample is reported with an accuracy equal to that of the reference standard, i.e. ±0.1 % of the reported U-235 concentration.

Standard Procedure for the U-234 and U-236 Isotopic Analysis of Uranium Hexafluoride

1. INSTRUMENT

The mass spectrometer used for the U-234 and U-236 isotopic analysis of uranium is a single focussing 90° sector magnet type, having an analyser tube radius of 12 inches (305 mm). It is fitted with a double filament thermal ionisation source and the ion current is measured by an electron multiplier. The ion beams are scanned across the collector by varying the field of the electromagnet.

2. MEASUREMENT PROCEDURE

The sample is prepared by hydrolysing the UF_6 using distilled water. A solution with a U concentration of at least 1 mg/ml is used. A small quantity of this solution is dried onto the side filament of the ion source. Ion beams are obtained by operating the centre filament at maximum temperature and the side filament at sufficient temperature to provide the required size of ion beams.

The ion beams are scanned across the collector in the sequence U-234, U-235, U-236, U-238, U-238, U-236, U-235, U-234, and their beam heights recorded. This comprises one cycle of measurement. The mean beam height for each isotope is obtained from the pair of measurements for each isotope. These mean beam heights are then used to calculate the ratios

$$\frac{\text{U-234}}{\text{U-235}}, \quad \frac{\text{U-236}}{\text{U-235}}, \quad \text{and} \quad \frac{\text{U-238}}{\text{U-235}}$$

The above cycle of measurement is repeated four or five times to give five or six values for the ratios which are then meaned. The mean values of the ratios

$$\frac{\text{U-234}}{\text{U-235}}, \quad \frac{\text{U-236}}{\text{U-235}}, \quad \text{and} \quad \frac{\text{U-238}}{\text{U-235}}$$

are used to calculate the U-234 and U-236 atomic percentage concentrations in the sample.

3. ACCURACY

Normally the U-234 and U-236 concentrations in a sample are only required to enable an atomic U-235 concentration to be converted to a mass U-235 concentration, and so great accuracy in the measurement of the very small U-234 and U-236 concentrations is not necessary. A precision is therefore not quoted for these measurements and the accuracy quoted is intended to indicate the likely range in which the true result lies. As a result of long experience of comparing the measured U-234 and U-236 contents of various standards with their true values, an accuracy of ± 10 % of the U-234 and U-236 content is quoted.

Annex VII:

"Absolute Evaluation of U-235 Abundance in Two Samples from the UF_6 Interlab Test"

O. Coenegracht, H. Zeckendorf

URENCO Nederland Operations B.V.
Almelo, Netherlands

1. Description of the Measurement Process

In the "tails" and "feed" test materials of the interlabtest the U-235 abundance was determined at the laboratory of UOBV by measurement of the samples against a standard sample.

The value of the standard sample (supplied by CBNM, Geel) was stated as follows (atomic ratio):

U-235/U-238 = 0.0033183 ± 0.00000 50

According to the specification the supplier gives for the mass spectrometer

- the test material and the standard sample have to be measured within 20 minutes;
- the ratio of isotopic ratios $a = \frac{(\text{U-235:U-238})_{\text{test material}}}{(\text{U-235:U-238})_{\text{standard sample}}}$

  has to lie within the interval $0.94 < a < 1.06$.

The first condition was observed. The second condition was largely exceeded because the "tails" sample and the "feed" sample were measured against the standard sample above. The ratios a in this test were for the

"tails" sample: $a = 1.33$
"feed" sample: $a = 2.19$.

For operational reasons the samples should be measured against the standard sample coupled to the mass spectrometer in normal operation and the standard samples should not be exchanged. The measurements were made in usual routine operation, i.e., that the mass spectrometer had not been optimized for these measurements. Deviating from the measurement procedures followed in general in the experiment, the measurement cycles of a period were distributed over an average time interval of one month.

2. Evaluation

2.1 Table VII-1 is a list of all measured values. Each one is an isotopic ratio (atomic ratio) of the test material ($R_{\text{test material, true}}$) and was obtained in the following way:

$$R_{\text{test material, true}} = R_{\text{test material, measured}} \times \frac{R_{\text{stand. sample, true}}}{R_{\text{stand. sample, measured}}}$$

The data were evaluated by analogy with the other measurements made in the interlaboratory test so that the relative standard deviations indicated are roughly comparable with those of the interlaboratory test.

2.2 In Tables VII-2 and VII-3 the mean values calculated for each period of measurement from the data of Table VII-1 have been compiled together with the standard deviation of the individual measurement cycle and its relative value. To define and calculate the variables used here and in the following paragraphs reference is made to Annex III. Tables VII-2 and VII-3 should be compared with Tables 4-1 and 4-2.

2.3 Besides the laboratory mean values repeated in column 2, columns 3 and 4 of Tables VII-4 and VII-5 contain the standard deviations of the individual values measured during a cycle and their relative values, respectively, determined on the basis of all measurements. Columns 5 and 6 show the respective variables for the period of measurement.

Tables VII-4 and VII-5 should be compared with Tables 4-4 and 4-5.

2.4 In Table VII-6 the error bars of the laboratory means and their relative values, respectively, have been indicated in columns 3 and 4. The relative values in column 4 should be compared with the relative values in Fig. 4-4.

2.5 The determinations of the total isotopic compositions of sample materials by BNFL and CBNM (Table 3-1) were used to calculate the ratios $(\text{U-235/U-238})_{\text{test material}}$. They are compared in Table VII-7 with the mean values obtained by our laboratory.

Table VII - 1:

Data of "Tails" Samples

Lab. Code	Period	Measurement Cycle					
		1	2	3	4	5	6
	1	0.0044032	0.0044009	0.0044025	0.0044016	0.004404	0.0044018
	2	0.0043973	0.0044057	0.0044062	0.0044032	0.0044004	0.0044027
	3	0.0044022	0.0044007	0.0044022	0.0044057	0.0044028	0.0044042
	4	0.0044013	0.0044025	0.0044017	0.0044021	0.0044036	0.0044038
	5	0.0044012	0.0044009	0.0044064	0.0044056	0.0044034	0.0044051
	6	0.0044014	0.0044045	0.0044034	0.0044025	0.0044017	0.0044024

Data of "Feed" Samples

Lab. Code	Period	Measurement Cycle					
		1	2	3	4	5	6
	1	0.007256	0.0072553	0.0072513	0.007252	0.0072581	0.0072521
	2	0.0072434	0.0072498	0.0072603	0.0072581	0.0072504	0.0072577
	3	0.0072564	0.007257	0.0072556	0.0072579	0.0072515	0.0072568
	4	0.0072548	0.0072498	0.007253	0.0072523	0.007249	0.0072492
	5	0.0072523	0.0072449	0.0072575	0.0072634	0.0072565	0.0072624
	6	0.0072557	0.0072544	0.0072545	0.0072566	0.007255	0.0072559

Table VII - 2: Period Means for "Tails" Measurements and Related Data *)

1 Lab. Code	2 Period Mean $Y_{ij.}$	3 SD of Cycle $\pm \sigma$ (i,j)	4 RSD of Cycle $\pm s$ (i,j) [^{o}/oo]	5 Lab. Mean $Y_{i..}$
	0.0044023	0.0000011	0.26	
	0.0044026	0.0000033	0.76	
	0.0044030	0.0000018	0.40	0.0044028
	0.0044025	0.0000010	0.23	
	0.0044038	0.0000023	0.53	
	0.0044027	0.0000011	0.26	

Table VII - 3: Period Means for "Feed" Measurements and Related Data *)

1 Lab. Code	2 Period Mean $Y_{ij.}$	3 SD of Cycle $\pm \sigma$ (i,j)	4 RSD of Cycle $\pm s$ (i,j) [^{o}/oo]	5 Lab. Mean $Y_{i..}$
	0.0072541	0.0000027	0.38	
	0.0072533	0.0000065	0.89	
	0.0072559	0.0000023	0.31	0.0072544
	0.0072514	0.0000024	0.33	
	0.0072562	0.0000069	0.94	
	0.0072554	0.0000009	0.12	

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*) The "EINS" program was used to calculate these data (Annex IV)

Table VII - 4: Laboratory Mean for "Tails" Measurements and Related Data *)

1 Lab. Code	2 Lab. Mean $Y_{i..}$	3 SD of Cycle $\pm \sigma_C$	4 RSD of Cycle $\pm S_C$ [°/oo]	5 SD of Period $\pm \sigma_P$	6 RSD of Period $\pm S_P$ [°/oo]
	0.0044028	0.0000020	0.45	not significant	

Table VII - 5: Laboratory Mean for "Feed" Measurements and Related Data *)

1 Lab. Code	2 Lab. Mean $Y_{i..}$	3 SD of Cycle $\pm \sigma_C$	4 RSD of Cycle $\pm S_C$ [°/oo]	5 SD of Period $\pm \sigma_P$	6 RSD of Period $\pm S_P$ [°/oo]
	0.0072544	0.0000042	0.58	0.000006	0.08

*) The "ZWEI" program was used to calculate these data (Annex IV)

Table VII - 6: Laboratory Means and Related Data *)

1 Sample	2 Lab. Mean $Y_{i..}$	3 SD of Lab. Mean ± σ (LM)	4 RSD of Lab. Mean ± S (LM) [^{o}/oo]
"Tails"	0.0044028	0.0000002	0.05
"Feed"	0.0072544	0.0000007	0.10

*) The data were calculated using formulae 7 and 8 of Annex III,3

Table VII - 7: Comparison of Laboratory Means with the Values Obtained from the Determinations of Total Isotopic Composition

1 Sample	2 UOBV Lab. Mean	3 BNFL	4 CBNM
"Tails"	0.004403	0.004398	0.004402
"Feed"	0.007254	0.007244	0.007254

Annex VIII:

"Memory Effect in Uranium Isotope Determination"

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Memory Effect in Uranium Isotope Determination

In the UF_6 isotope determination by mass spectrometry a so-called memory effect may occur which leads in practice to a reduction of the observed difference in the isotopic composition of two UF_6 gas samples.[1]. This memory effect is defined as follows

$$M = \frac{(a-1)_{\text{true}}}{(a-1)_{\text{measured}}}$$

with $$a = \frac{(\text{U235/U238})\ \text{sample 1}}{(\text{U235/U238})\ \text{sample 2}}$$

The effect is probably due to the depositing of low-valency uranium fluoride and uranium oxide fluoride compounds and of UF_6 itself on the metallic surfaces of the gas pipes and the ion source of the mass spectrometer due to the high corrosivity of uranium hexafluoride. These deposits might later-on give rise to isotope exchange reactions with the following measured samples. This memory effect depends on the types of mass spectrometers used. It has been proved that it is higher with older types of instruments not yet provided with a molecular beam inlet system and in which no cryogenic traps have been installed in the vicinity to the ion source space. In these older types of instruments the memory factor may lie between 1.04 and 1.12, whereas for mass spectrometers with a molecular beam inlet system it reaches about 1.01 – 1.03[1].

To determine the memory factor, two UF_6 samples of precisely known isotopic compositions are needed. Previous measurements performed at our Institute on two mixed samples made of natural uranium and NBS standards 010 and 005 of precisely known compositions [10] have shown that a memory factor of about 1.02 ± 0.01 must be expected for the UF_6 mass spectrometers of the types MAT 511, Atlas CH4-UF and MAT UF5 which we used in the UF_6 interlaboratory test [2]. This is in good agreement with the results measured by the authors Smith and Langdon[1].

In case that UF_6 gas samples of precisely known isotopic compositions are not available, the memory factors of different mass spectrometers can be determined relative to each other by comparison measurements using two available UF_6 samples of different isotopic compositions (e.g. the interlab samples).

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[1] Cf. also [9].

[2] The relatively high uncertainty in the determination of the memory factor is presently due to the uncertainty of the standards used.

To show up clearly the memory effect and to demonstrate that it cannot be eliminated by scavenging or extended pumping between the sample introductions, the feed samples of the interlaboratory tests were determined in addition by a CEC mass spectrometer [1]. Fig. VIII-1 shows the deviations of the CEC measured values of the ratio a = $(\text{U-235: U-238})_{\text{test material}}/(\text{U-235:U-238})_{\text{reference material}}$ from the interlab mean. It can be noted that the deviations of the CEC measurements from the interlab mean are subjected to strong variations in time and, obviously, get smaller with increasing time of operation of the respective ion source. It is further shown by the measurements that intermediate scavengings with the gas to be measured and extension of the pumping times between the sample introductions do not practically influence the value measured.

The values given for the three spectrometers by the Institute of Nuclear Engineering (IKVT) of Gesellschaft für Kernforschung within the interlaboratory test were not corrected to take into account the memory effect. If the memory effect is taken into account, which exists in the opinion of IKVT, the ratio a of the feed samples are found to be higher for the 3 spectrometers of IKVT than stated in the report. Since the memory effect always reduces the value of the ratio a [2] IKVT holds that the "true value" of the ratio a of interlab samples ("feed") is higher than the mean value obtained in the interlab test. The mean value corrected for the memory effect of the three spectrometers used in the test by IKVT has been given in addition in the figure. For comparison, also the values have been entered which were determined from the absolute determinations of the CBNM and BNFL laboratories participating in the test [3].

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[1] Type CEC 21-220 A, date of delivery 1958. The instrument operates with a viscous gas inlet system and has no cryogenic traps close to the ion source so that the non-ionized gas can not be rapidly pumped off.

[2] This applies to values of $a > 1.0$ (cf. also Chapter 4).

[3] The error bars indicated of ± 0.5 ⁰/oo and ± 0.85 ⁰/oo , respectively, are estimated values of these laboratories communicated during the final discussion of the interlab test and thereafter.

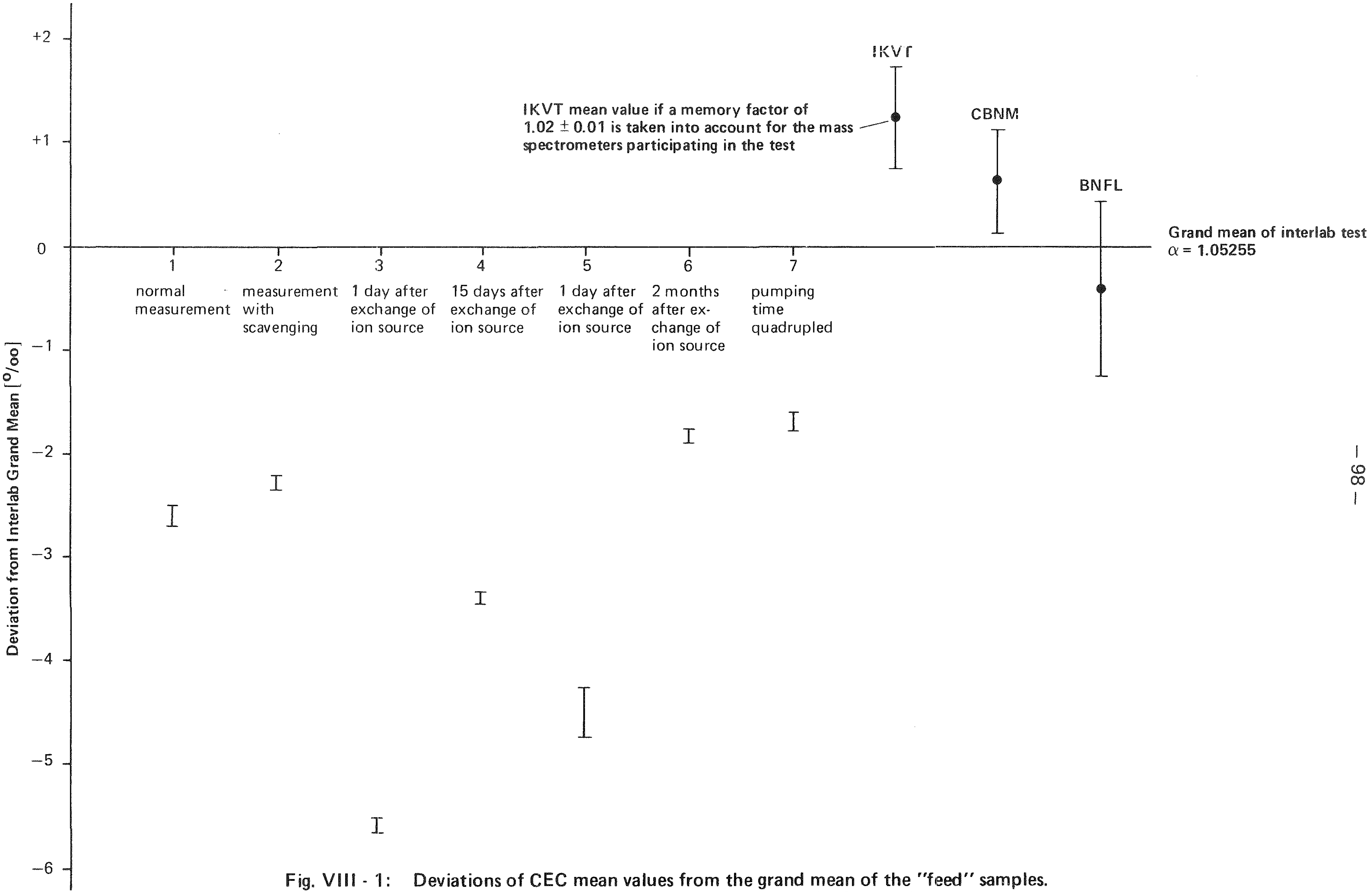

Fig. VIII - 1: Deviations of CEC mean values from the grand mean of the "feed" samples. The CEC mass spectrometer did not participate in the interlab test.

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