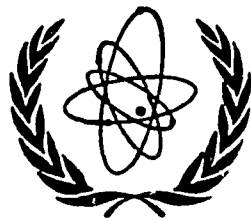


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ERROR ANALYSIS OF ISOTOPE DILUTION MASS SPECTROMETRY METHOD WITH
INTERNAL STANDARD

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SUMMARY

The computation algorithms of the normalized isotopic ratios and element concentration by isotope dilution mass spectrometry with internal standard are presented. A procedure based on the Monte-Carlo calculation is proposed for predicting the magnitude of the errors to be expected. The estimation of systematic and random errors is carried out in the case of the certification of uranium and plutonium reference materials as well as for the use of those reference materials in the analysis of irradiated nuclear fuels.

INTRODUCTION

The internal standard (internal calibration, normalization) method is based on the following

- the sample of unknown isotopic composition is mixed with a reference material (RM) (spike, internal standard) of known isotopic composition

- mass spectrometric measurements are performed on the sample alone and on its mixture with RM
- from the results of these measurements the isotopic composition of the sample and the ratio of the element (isotope) concentration in the sample and the RM are computed.

The sample and RM should not have less than 3 isotopes each, and their isotopic composition should be different.

In recent years the internal standard method has been used by SAL and NWAL for the certification of uranium and plutonium in spikes QS85, QS87 and for the analysis of irradiated nuclear fuels with the use of those spikes. These spikes are enriched in ^{233}U , ^{236}U and ^{242}Pu , ^{244}Pu . The RM NBS U-500 enriched in ^{235}U and ^{239}U and NBS 947 enriched in ^{239}Pu and ^{240}Pu were used as internal standards to certify the spikes. In the second case the sample of irradiated nuclear fuel enriched in ^{238}U and ^{239}Pu was analysed using the QS 85 spike as internal standard.

One of the important points in the application of the internal standard method is the estimation of its systematic and random errors (1,2) and the contribution of different factors, such as isotopic composition of sample and RM, ratio of RM to sample in a mixture, error of certification of RM, error of mass spectrometric measurements, to these errors. The main purpose of the present paper is to describe and discuss those aspects in the cases of the certification of a tracer of QS type and of its use in the analysis of irradiated nuclear fuels.

1. ALGORITHMS OF COMPUTATION

1.1. Certification of spikes of type QS. Calculation of isotopic ratios.

In this case the spike to be certified is treated as a sample and is denoted with an index n. The isotopic RM used as internal standard is denoted by t, a mixture of sample and RM carries index m.

Based on the equations of material balance we write for uranium (3):

$$\frac{N_n^6}{N_t^6} = \frac{R_m^{3/6} - R_t^{3/6}}{R_n^{3/6} - R_m^{3/6}} = \frac{R_m^{5/6} - R_t^{5/6}}{R_n^{5/6} - R_m^{5/6}} = \frac{R_m^{8/6} - R_t^{8/6}}{R_n^{8/6} - R_m^{8/6}} \quad (1)$$

where N_n^6, N_t^6 - are the number of moles of ^{236}U in the sample and the RM.

The relationship between the "true" and the measured ratios of ^{233}U and ^{236}U isotopes is given by the equations

$$R_n^{3/6} = R_n^{3/6*} (1 + \Delta_{3/6} F_n) \quad (2)$$

$$R_m^{3/6} = R_m^{3/6*} (1 + \Delta_{3/6} F_m) \quad (3)$$

Δ - is the difference between mass numbers of the isotope appearing in the denominator and the numerator ($\Delta_{3/6}=3$)

F_n, F_m - are the mass discrimination coefficients per mass unit for the measurement of the sample and the mixture respectively.

Substituting (2) and (3) into a second member of (1) and taking the first two members of (1), we obtain:

$$R^{3/6*} (1 + \Delta_{3/6} F_m) - R_t^{3/6} = QR_n^{3/6*} (1 + \Delta_{3/6} F_n) - QR_m^{3/6*} (1 + \Delta_{3/6} F_m) \quad (4)$$

where $Q = N_n^6 / N_t^6$

We rewrite (4) in a form

$$\Delta_{3/6} R_m^{3/6*} (F_m + QF_m) = (R_m^{3/6*} - R_n^{3/6*}) Q - \Delta_{3/6} R_n^{3/6*} QF_n = R_t^{3/6} - R_m^{3/6*} \quad (5)$$

Setting $F_m(1+Q) = X_1$ (6)

$Q = X_2$ (7)

$QF_n = X_3$ (8)

and taking (1), a system of three linear equations can be written

$$\Delta_{i/6} R_m^{i/6*} X_1 + (R_m^{i/6*} - R_n^{i/6*}) X_2 - \Delta_{i/6} R_n^{i/6*} X_3 = R_t^{i/6} - R_m^{i/6*} \quad (9)-(11)$$

where $i = 3, 5, 8$

We solve that system for X_1 , X_2 and X_3 , and use (6) - (8) to find F_n , F_m , Q . Then (1) - (3) yield the "true" (normalized) isotopic ratios in the sample - R_n , as well as the number of moles of analyzed isotope in the sample (here ^{236}U)

In the case of the certification of a plutonium spike enriched in ^{242}Pu , ^{244}Pu the calculations is exactly identical except that $j = 242$ and $i = 239, 240, 244$

1.2. Certification of spike of QS type. Calculation of element concentration.

In that case NBS960, MU1, NBS949 f, MP1 or EC210 are used for chemical assay. The QS spike is considered as a sample.

For uranium one may write:

$$\frac{C_n^3 M_n + C_t^3 M_t}{C_n^6 M_n + C_t^6 M_t} = \frac{R_n^{3/6} C_n^6 M_n + C_t^3 M_t}{C_n^6 M_n + C_t^6 M_t} = R_m^{3/6} (1 + 3F) \quad (12)$$

$$\frac{C_n^8 M_n + C_t^8 M_t}{C_n^6 M_n + C_t^6 M_t} = \frac{R_n^{8/6} C_n^6 M_n + C_t^8 M_t}{C_n^6 M_n + C_t^6 M_t} = R_m^{8/6} (1 - 2F) \quad (13)$$

were C^i - is the concentration of isotope i (mol/g), M_n, M_t - are the masses of sample and spike, respectively.

Solving (12) and (13) for F and equating the members without F , we obtain:

$$\frac{R_n^{8/6} C_n^6 M_n + C_t^8 M_t}{2R_m^{8/6} (C_n^6 M_n + C_t^6 M_t)} + \frac{R_n^{3/6} C_n^6 M_n + C_t^3 M_t}{3R_m^{3/6} (C_n^6 M_n + C_t^6 M_t)} = \frac{5}{6} \quad (14)$$

substituting $C_t^3 = R_t^{3/8} C_t^8$ and $C_t^6 = R_t^{6/8} C_t^8$ and solving (14) for C_n^6 we obtain:

$$C_n^6 = \frac{C_t^8 N_t (5R_m^{8/6} R_n^{3/6} R_t^{6/8} - 3R_m^{3/6} - 2R_m^{8/6} R_t^{3/8})}{N_n (3R_m^{3/6} R_n^{8/6} + 2R_m^{8/6} R_n^{3/6} - 5R_m^{8/6} R_t^{3/6})} \quad (15)$$

Note that R_t, R_n - true isotopic ratios, R_m - measurement isotopic ratios.

Note that $R_t^{6/8} = 0$, $R_t^{3/8} = 0$, $C^i(r \dots g) = \frac{C^i(g/g)}{M^i(g/mol)}$

$$C_t^8 = C_t^u \cdot \mu_t^8, \quad \mu_t^8 = \frac{M^8}{\sum_i M^i R_t^{i/8}} = \frac{238.05079}{\sum_i M^i R_i^{i/8}}$$

Where M^i - is the atomic mass of isotope i , C_t^u - the uranium concentration in the spike (g/g or mg/g or $\mu\text{g/g}$) finally we obtain:

$$C_n^6(\mu\text{g/g}) = C_t^u(\mu\text{g/g}) \frac{236.04556 \cdot M_t \mu_t^8 \cdot 3R_m^{3/6}}{238.05079 N_n (5R_m^{8/6} R_m^{3/6} - 3R_m^{3/6} R_n^{8/6} - 2R_m^{8/6} R_n^{3/6})} \quad (16)$$

for plutonium, assuming that $R_t^{4/9} = 0$, we obtain in a similar way:

$$C_n^2(\mu\text{g/g}) = C_t^{\text{Pu}}(\mu\text{g/g}) \frac{242.05874 N_t \mu_t^9 (5R_m^{4/2} R_m^{9/2} R_t^{2/9} - 2R_m^{4/2})}{239.05216 N_n (3R_m^{9/2} R_n^{4/2} + 2R_m^{4/2} R_n^{9/2} - 5R_m^{4/2} R_m^{9/2})} \quad (17)$$

1.3. The use of QS spikes for IDA. Determination of isotopic composition and element concentration in a solution of spent nuclear fuel.

In that case we apply the calculation method presented in parag.1.1. Note, that here the spike is used as a standard and is enriched in ^{233}U and ^{236}U or in ^{242}Pu and ^{244}Pu and certified for isotopic composition and element concentration. The sample is a solution of spent nuclear fuel.

2. ERROR ESTIMATION.

2.1. Computational Errors.

The system of equations (9) - (11) have an exact analytical solution. Computational errors can however be introduced by rounding errors if the computer programme does not carry enough digits.

Therefore we examined first the validity of the system of equations set under different conditions, i.e. different isotopic compositions for the RM, sample and mixture.

Some model calculations were performed with the following assumptions: "Known" values were defined for the isotopic composition of RM, sample and mixture and the mass discrimination coefficients for the sample and mixture, F_n , F_m . The "measured" isotopic ratios R_n^* and R_m^* were calculated from formulas (2),(3). Then the system of equations was solved for F_n and F_m , and R_n was calculated. The computational procedure was considered to be accurate as long as the calculated values of R_n agreed within 7 significant digits with the initial data. An example is given in Table 1 (In those calculations $F_n = F_m = 0.001$). It can be seen, that the computation procedure works satisfactorily in a broad range of isotopic composition. Computational errors become apparent when the composition of the RM approaches that of the sample.

2.2. Propagation of Errors in the certification of isotope ratios of a QS spike

a) Effect of the Uncertainties on the Standard.

The certificate of the isotopic RM used for the certification of the spike, specifies the magnitude of the uncertainty of the certified values. For example the uncertainty in NBS U-500 for the molar fraction of the "main" isotopes ^{235}U and ^{238}U is 0.05%. In IFCO 2702-83 that value is 0.012%. In NBS 947 the error on the ^{240}Pu abundance is 0.022%. These uncertainties constitute a component of systematic error on the final result of analysis. Due to the complexity of the calculation, it is difficult to write an analytical expression linking the error of the result of the calculation (R_n) with the uncertainty of the RM certificate.

We used instead the Monte-Carlo method as follows. We assumed that the molar fractions of isotopes in the RM are random variables having a normal distribution with the mean values as given in the certificate and with standard deviations (SD) equal to the uncertainty specified in the certificate. We assumed that the uncertainties on the molar fractions of isotopes are mutually independent. We obtained in that way "a population" of 10-30 standard reference materials. The calculation was performed as described above except that for each sample and mixture we used the whole "population" of 10-30 RM's. In that way we obtained for a given sample 10-30 different values of R_n . The standard deviation of these computed R_n was calculated and adopted as an estimate of the standard deviation of the systematic error component.

Fig.1 shows the effect of various factors on the systematic error $\theta(R_n^{3/6})$ of the normalized isotopic ratio of $^{233}\text{U}/^{236}\text{U}$ in the sample. The circles represent RM's which are used in practice - NBS U-500, IFCO 2699-83, 2702-83, 2704-83. Fig.1 can be used as an approximated nomogram for

the determination of $\theta(R_n^{3/6})$ when the sample is enriched in ^{233}U and ^{236}U with the ratio $^{233}\text{U}/^{236}\text{U} = 1$, for example for QS 87. It can be seen, that in such a case it is advisable to use RM's in which the ratio $^{235}\text{U}/^{238}\text{U}$ is in the range 0.25 - 4. In that case a change of the ratio of element masses in RM and sample in the range 0.1 to 10 has little influence on the results. It can be seen from Fig.2 that relatively abundant ^{233}U and ^{236}U in the RM do not cause a significant increase of the systematic error, which is mainly determined by the uncertainty on the certification of the major isotope ratio of the RM.

b) Effect of random errors in the mass spectrometric measurements.

In the second step we determined the dependence of the random error of the normalized ratios $Sr(R_n^{3/6})$ in the spike on the random error of the mass spectrometric measurements of the spike and mixture. The latter includes error resulting from uncontrolled changes of measurement conditions - temperature, pressure etc. This error can also result from the random appearance of non-linear effects in mass fractionation i.e. a disturbance of the relations (2),(3). The Monte-Carlo method was also used for estimating these random errors. In that case the mean of the "measured" values of isotopic ratios R_n^* and R_m^* is calculated from formulas(2),(3). A random generator is used to generate 10 - 30 individual values with a mean equal to the calculated value and a standard deviation equal to the reproducibility of the mass spectrometric measurements (RSD). The dependence of RSD on the value of the isotopic ratios (approximately estimated from experimental data) is shown in Fig.3. For isotopic ratios 0.1-1.0 three values of RSD were used - 0.05, 0.1 and 0.5%. For isotopic ratios below 0.1 the values of RSD were taken from Fig.3. The calculation was carried out for different isotopic compositions of RM and spike (the ratio

of RM to spike was always equal to 1). The following cases were considered: "pure" RM - "pure" spike, "pure" RM - "dirty" spike, "dirty" RM - "pure" spike, and "dirty" RM - "dirty" spike. The composition of the RM and the spike for the first and last cases are given in the first two rows of Table 1. In the case of a "moderately dirty" RM and spike we assumed that both contained 10% of a minor isotope. The results of the calculation are given in Fig.4. First of all it seems that in all cases the RSD of the normalized ratios is greater than the RSD of the "measured" ones. This allows to conclude, that, as a result of normalization of experimental data with the use of the discussed algorithm, the RSD of the normalized ratio is bigger than the RSD of the measured ratio, i.e. during the measurement the relations (2),(3) are not fulfilled. It can also be seen that only very significant simultaneous "contamination" of the spike and the RM (with more than 20% of minor isotopes) increases seriously the random error of the normalized ratios. Otherwise the "contamination" has little influence.

In conclusion, it is not necessary that the spike and RM have very low abundances for the minor isotopes. As a rule the systematic error is first of all determined by the certification error of the RM and the random error - by the magnitude of deviations of expressions (2),(3) from linearity, random MS measurement errors and other.

2.3. Propagation of errors when using QS spikes for the isotopic and chemical analysis of spent nuclear fuel.

When using the QS spike to analyze a sample, the spike is now considered as the standard. In this case the major isotopes of the spike are used for normalization, i.e. ^{233}U and ^{236}U for uranium and ^{242}Pu and ^{244}Pu for plutonium. The major isotopes of interest in the sample are ^{235}U , ^{238}U and ^{239}Pu , ^{240}Pu . The computation procedure should be also error free.

We examine now the influence of the spike certification errors and of the random measurement errors of the sample and the mixture on the errors of the values calculated for element concentration and normalized isotopic ratios in the sample. The estimation procedure is similar to the one described in parag.1.1. The only difference is, that additional variables - the mass of spike and sample taken for analysis and element concentration in the spike must be taken into account. The isotopic composition of the spike and the sample used in the calculations are given in Table 2. The isotopic composition of the uranium and the plutonium in (a) is similar to the composition of QS87.

a) Estimate of System Errors.

The other variables of interest are the errors on the molar fractions of the spike, on the mass ratios of elements in spike and sample. Fig.5 gives the relative systematic errors in the uranium concentration $\theta(C_U)$ and on the normalized ratios of $^{235}\text{U}/^{238}\text{U}$ $\theta(R_n^{5/8})$ as a function of the spike certification error. It is seen (a) that when the ratio of elements in the spike and sample are less than 1, the $\theta(R_n^{5/8})$ are directly proportional to the certification errors of the spike. When the ratio increases to 5 proportionality is preserved for $\theta(C_U)$, but $\theta(R_n^{5/8})$ increases much faster. If a "contaminated" spike (b) is used, the relationship does not change for $\theta(C_U)$ as long as the ratio spike: sample 1. But when the latter ratio reaches 5, $\theta(C_U)$ is multiplied by 1.5 - 2. For $\theta(R_n)$ the error increases already faster for a ratio spike-sample equal to 1; and when $\theta(R_n)^{5/8}$ increases 5 times, $\theta(R_n)$ equals 1 - 3% rel. In practice Fig. 5a can be used to obtain an estimate of the systematic errors to be expected on the result of uranium concentration and the isotopic ratio $^{235}\text{U}/^{238}\text{U}$ when IDA is done with QS 87. A more distinct change of $\theta(C_U)$ and $\theta(R_n^{5/8})$ as a function of the ratio between spike and sample is seen in Fig. 6. The $\theta(R_n^{5/8})$ is more sensitive to an increase in the ratio spike : sample than $\theta(C_U)$. The higher the abundance

of minor isotopes in the spike, the more $\Theta(R_n^{5/8})$ depends upon the spike : sample ratio. This is because the samples have usually a low abundance in ^{235}U . If the sample contained ^{235}U and ^{238}U in equal abundance, $\Theta(R_n^{5/8})$ and $\Theta(C_u)$ would barely vary as long as the ratio spike : sample is less than 10.

The dependence of $\Theta(C_{\text{Pu}})$ upon various factors affecting the analysis of plutonium is shown in Fig.7. These relationships do not differ from that for uranium. The systematic error to be expected when using QS 87 can be estimated from Fig.7(1). It should be mentioned that ξ_t represents in this case only the effect of errors encountered in isotopic ratio measurements. It is obvious, that the total variance is the sum of ξ_t^2 plus the variance of the error of the chemical assay of the spike.

b) Estimate of Random Errors.

The estimation of the random errors was performed in the same way as for the certification of the spike. The values of RSD used for the different isotopic ratios are found in Fig.3.

The calculations yield estimates of RSD for $R_n^{5/8}$ - $\text{Sr}(R_n^{5/8})$ and uranium concentration $\text{Sr}(C_u)$ or respectively $\text{Sr}(R_n^{9/0})$ and $\text{Sr}(C_{\text{Pu}})$ for plutonium. When the spike is "pure" and the ratio spike : sample is equal to 1 the errors of the normalized ratio $^{235}\text{U}/^{238}\text{U}$ and uranium concentration are not dependent on the RSD for major isotope ratios in the sample and the spike as long as these stay in the range 0.0005 - 0.003. They depend only on the RSD's of the minor isotope ratio of $^{235}\text{U}/^{236}\text{U}$ in the mixture - $R_m^{5/6}$ (Fig.8). Examining the relationships in Fig.3 and 8 it is possible to identify the composition of the mixture, i.e. the ratio spike : sample, which will lead to minimal random errors. It is seen from Fig.9 that $\text{Sr}(R_n^{5/8})$ and $\text{Sr}(C_u)$ are minimum when the ratio spike : sample equals 0.2. With this mixture all isotopic ratios $R_m^{i/6} = 1$. According to Fig.3 the effect of measurement errors is then minimum. With the decrease of spike :sample ratio the $R_m^{6/8}$ ratio also decreases and with the increase of that ratio, the ratio $R_m^{5/6}$ decreases. In turn, the errors of those ratios increase.

This causes the values of $Sr(Rn^{5/8})$ and $Sr(Cu)$ to increase also. On the other side, even a 10% "contamination" of the spike with ^{235}U and ^{238}U has no a strong influence on the random error of normalization, as can be seen in Fig. 10.

Comparing Fig.11 and 8 it is seen that there is no minimum for plutonium and the values of $Sr(C_{Pu})$ in the extreme cases are 2 times greater than the corresponding ones for uranium. This is undoubtedly attributed to the difference in the isotopic compositions of the spike and sample in cases of uranium and plutonium. It can be expected that using a similar approach for the simulation of the errors it will be possible to find more optimal composition of the plutonium spike which might minimize the normalization errors.

CONCLUSIONS

1. The method of Monte-Carlo was applied to predict the errors to be expected in mass spectrometric analyses with internal standard. Systematic and random errors were estimated. We considered the certification of spikes as well as their use for the analysis of uranium and plutonium containing materials, such as spent nuclear fuels. A routine computer programme is desirable to apply the proposed algorithms to specific analytical situations. Several results postulated in this work are valid for the certification of the QS87 spike and its utilization.
2. Low abundance of minor isotopes is not a prerequisite for accurate normalization. For example decreasing the ratio of uranium spike : sample to 0.2 - 0.1 should allow to use tracers containing up to 5% of ^{235}U of ^{238}U without loss of accuracy.
3. The described method gives the means to define the optimal ratio of spike to sample to minimize the normalization errors.

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Table 1. Influence of the isotopic compositions of RM and sample on calculation error.

Fig.1. The Uncertainties on the Certified Values of the Reference Material; their Effect on the Relative Systematic Error $\Theta(R_n^{3/6})$ of the Normalized Isotope Ratio $^{233}\text{U}/^{236}\text{U}$ of the Spike

(1) - $\epsilon_t = 0.005\%$; (2) - $\epsilon_t = 0.01\%$, (3) - $\epsilon_t = 0.05\%$.

Fig.2. Effect of Minor ^{233}U and ^{236}U Isotopes in the Reference Material ($R_t^{5/8}=1$).

Fig.3. Variation of Random MS Measurement Errors with the Magnitude of the Isotope Ratios.

Fig.4 Effect of Random MS Measurement Errors on the Coefficient of Variation of $^{233}\text{U}/^{236}\text{U}$ Normalized Isotope Ratios of the Spike

o - "pure" RM - "pure" sample

x - "pure" RM - "dirty" sample

● - "moderately dirty" RM - "moderately dirty" sample

⊖ - "dirty" RM - "dirty" sample

△ - "dirty" RM - "pure" sample

Table 2 The Isotopic Compositions of RM
and Samples Used for Spent Fuel Sample Analysis.

Fig.5. Effect of Spike Certification Errors on the Systematic
Error of Isotopic and Chemical Assays of Spent Fuel Samples.

spike : sample

x - 1 : 1

Δ - 1 : 10

□ - 5 : 1

Fig.6 Effect of Spike : Sample Ratio on the Systematic Error of
Uranium Isotopic and Chemical Assays of Spent Fuel Samples

o - $\theta(\text{Rn}^{5/8})$

x - $\theta(\text{Cu})$

ϵ_t - 0.05% rel.

Fig.7 Effect of Various Factors on the Isotopic and Chemical
Assay of Plutonium in Spent Fuel Samples.

For (2) and (3) $\epsilon_t = 0.1\%$ rel.

o - $\theta(\text{R}_n^{9/0})$

x - $\theta(\text{C}_{\text{Pu}})$

Fig.8. The Random MS Measurement Errors of $^{235}\text{U}/^{236}\text{U}$ in the Mixture. Its Effect on the Random Errors in Isotopic and Chemical Assay of Uranium in Spent Fuel Samples.

o - $\text{Sr}(R_n^{5/8})$
x - $\text{Sr}(C_u)$

Fig.9. Effect of Spike : Sample Ratio on the Random Errors in Isotopic and Chemical Assay of Uranium samples.

o - $\text{Sr}(R_n^{5/8})$
x - $\text{Sr}(C_u)$

Fig.10. The Minor Isotops in the Spike. Their Effect on Random Errors in Isotopic and Chemical Assay of Uranium Sample.

Fig.11. Effect of Sipke : Sample Ratio on the Chemical Assay of Plutonium Sample.

Table 1.

Influence of the isotopic compositions
of RM and sample on calculation error.

	RM				Sample				Mixture				$R_n^{calc.} - R_n^{taken}$
U	233	235	236	238	233	235	236	238	233	235	236	238	233/236
	0	49	0,2	49	49	0,05	49	0,05	24,5	24,525	24,51	24,525	0
	20	30	20	30	30	20	30	20	25	25	25	25	0
	24,99	25,01	25,01	24,99	25,01	24,99	24,99	25,01	25	25	25	25	0,0008
Pu	239	240	242	244	239	240	242	244	239	240	242	244	242/244
	76	18	1,2	0	0,04	3,1	69	27	38,02	10,55	35,1	13,5	0
	76	18	1,2	0	0	70	15	10	38	44	8,1	5	0

Table 2 The Isotopic Compositions of RM and Samples Used for Spent Fuel Sample Analysis.

	RM				Sample			
U	233	235	236	238	233	235	236	238
a	49,7	0,05	49,7	0,04	0	0,9	0,3	98,8
σ	45	5	45	5				
B	40	10	40	10				
Pu	239	240	242	244	239	240	242	244
	0,04	3,1	69,5	27,1	60	25	4	0

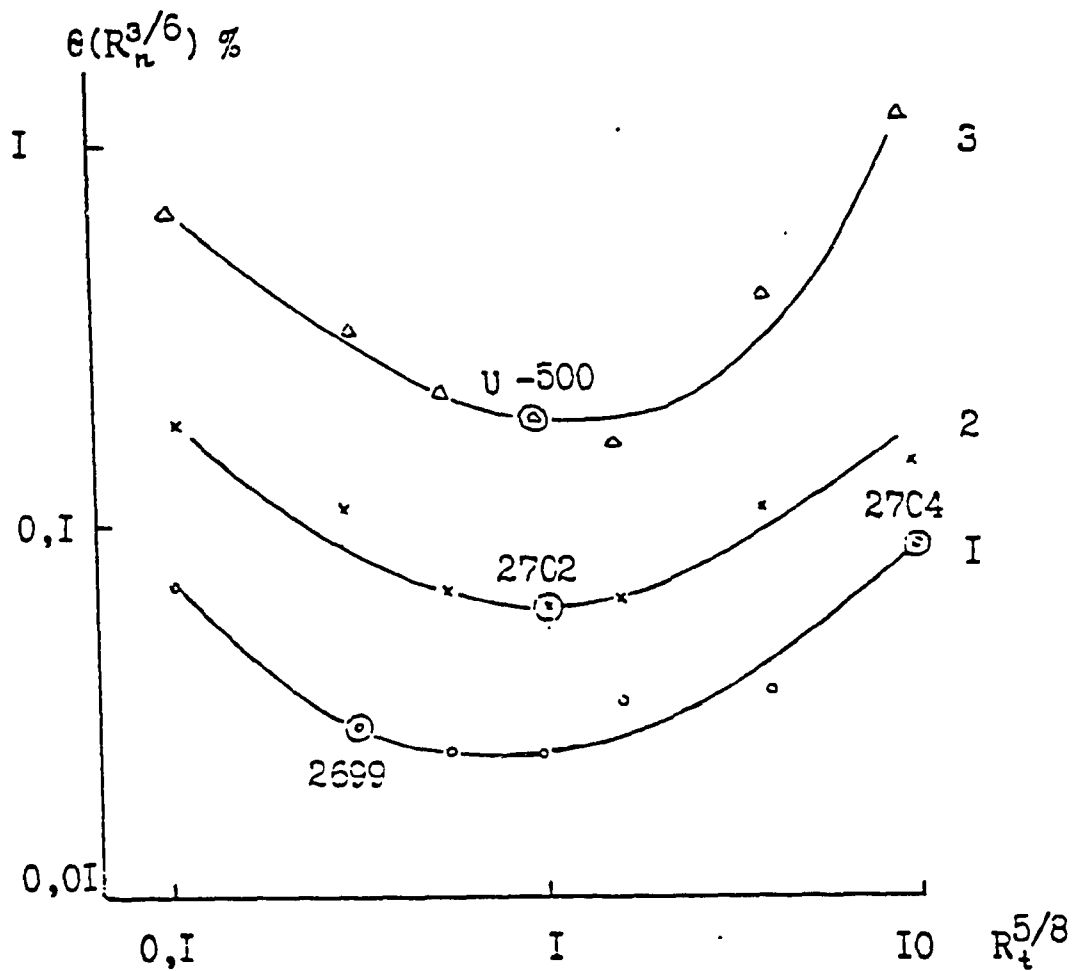


Fig.1. The Uncertainties on the Certified Values of the Reference Material; their Effect on the Relative Systematic Error $\epsilon(R_n^{3/6})$ of the Normalized Isotope Ratio $^{233}\text{U}/^{236}\text{U}$ of the Spike

(1) - $\epsilon_t = 0.005\%$; (2) - $\epsilon_t = 0.01\%$, (3) - $\epsilon_t = 0.05\%$.

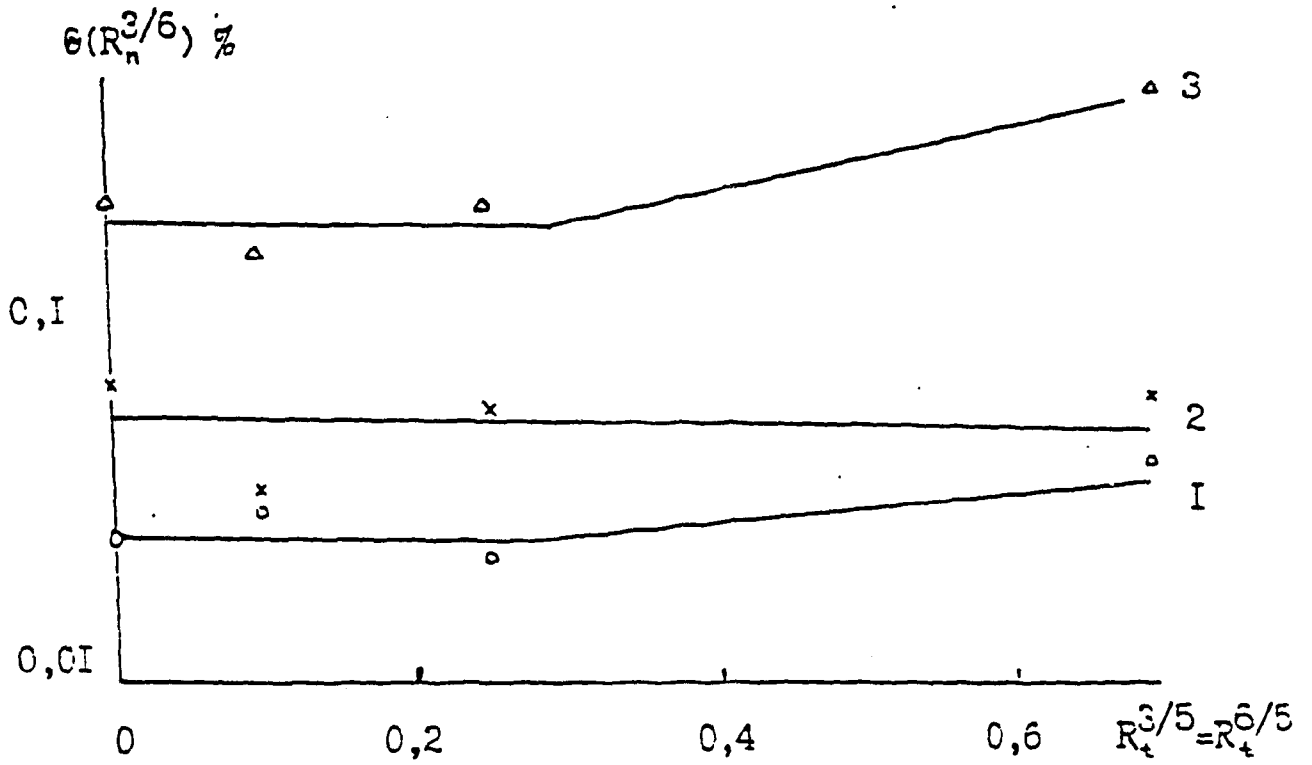


Fig.2. Effect of Minor ^{233}U and ^{236}U Isotopes in the Reference Material ($R_t^{5/8}=1$).

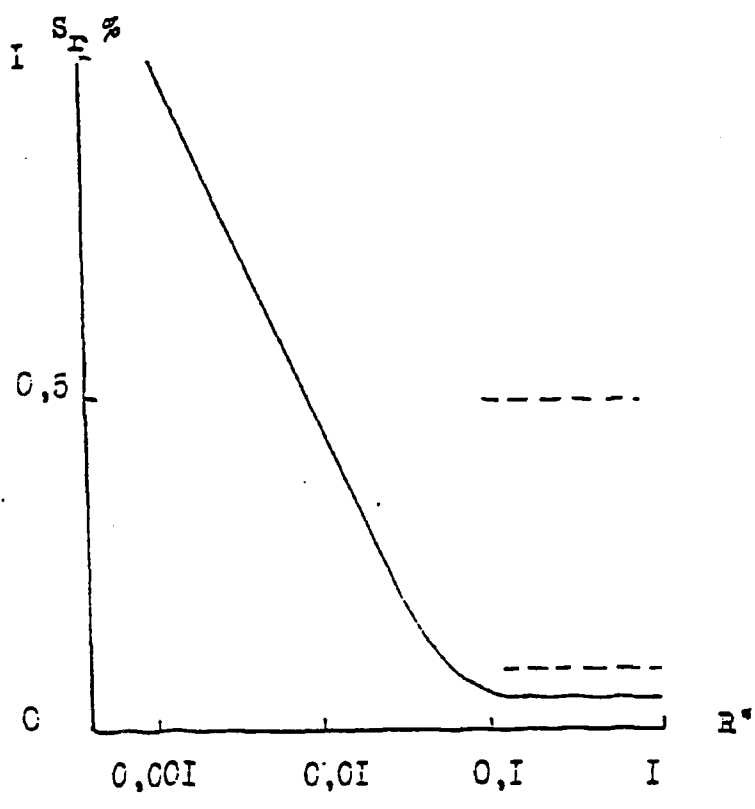


Fig.3. Variation of Random MS Measurement Errors with the Magnitude of the Isotope Ratios.

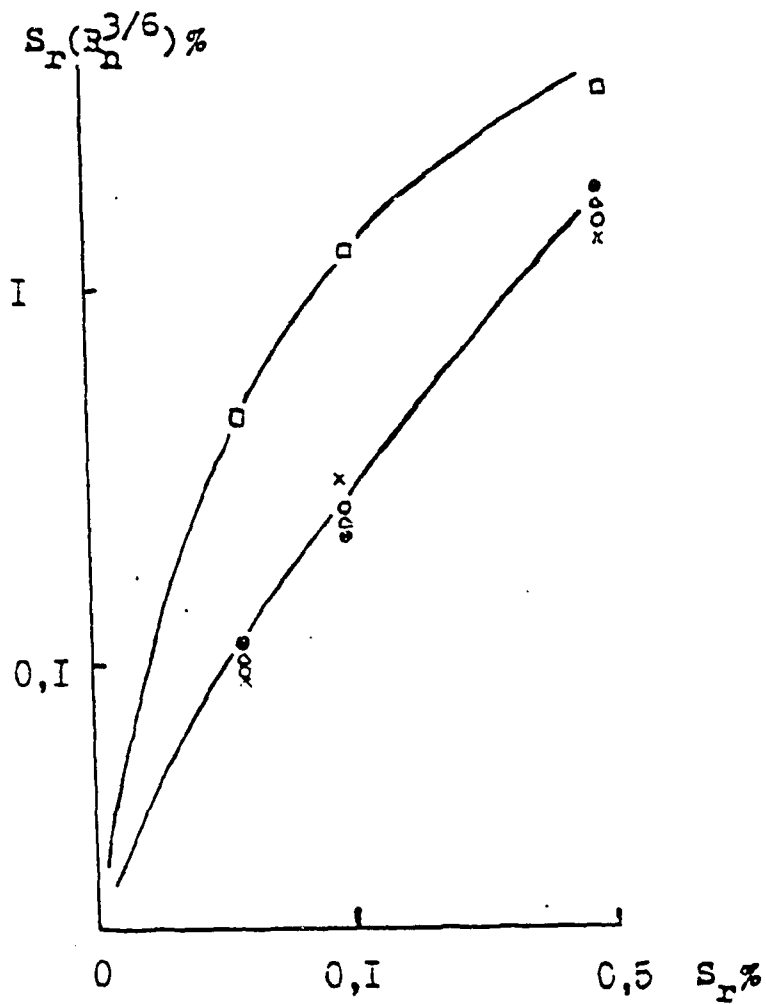


Fig.4 Effect of Random MS Measurement Errors on the Coefficient of Variation of $^{233}\text{U}/^{236}\text{U}$ Normalized Isotope Ratios of the Spike

- o - "pure" RM - "pure" sample
- x - "pure" RM - "dirty" sample
- - "moderately dirty" RM - "moderately dirty" sample
- - "dirty" RM - "dirty" sample
- △ - "dirty" RM - "pure" sample

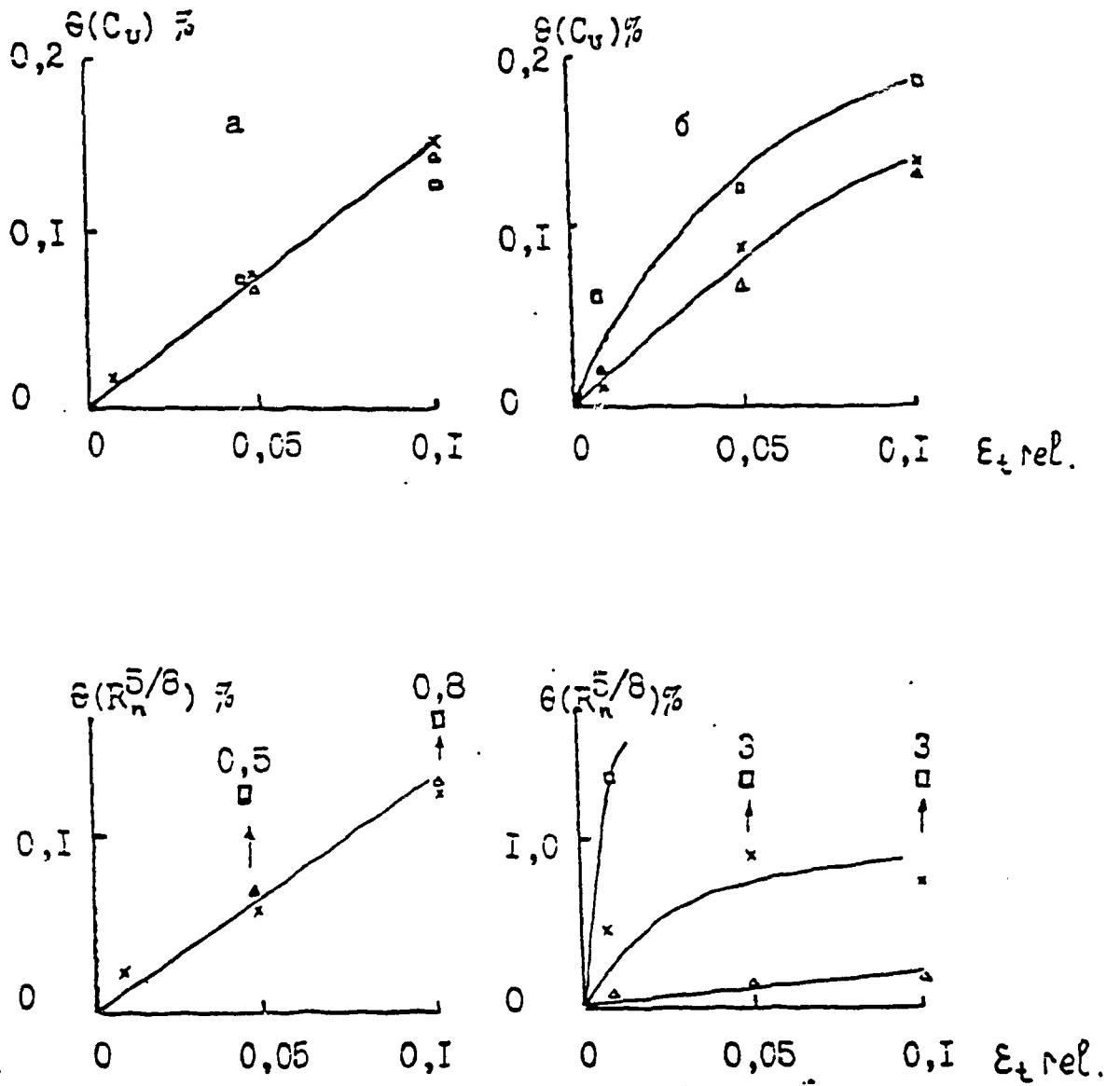


Fig.5. Effect of Spike Certification Errors on the Systematic Error of Isotopic and Chemical Assays of Spent Fuel Samples.

spike : sample

x - 1 : 1

Δ - 1 : 10

\square - 5 : 1

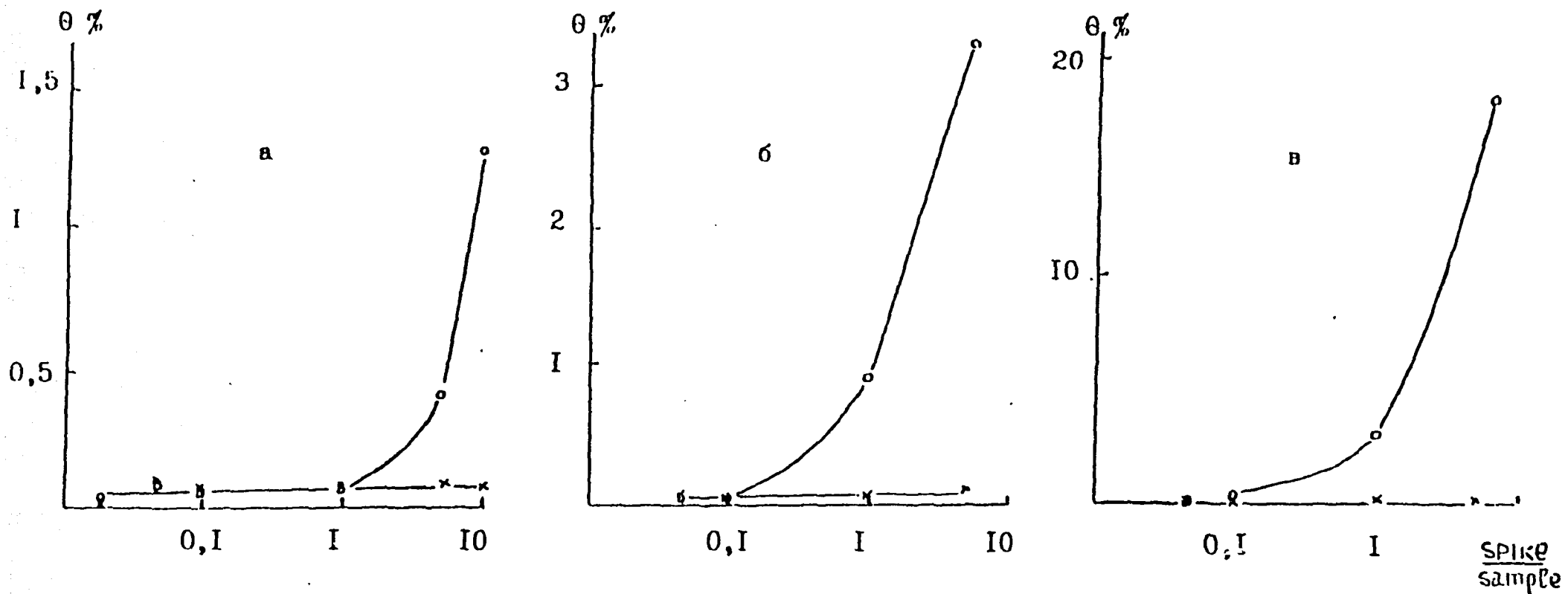


Fig.6 Effect of Spike : Sample Ratio on the Systematic Error of Uranium Isotopic and Chemical Assays of Spent Fuel Samples

o - $\theta(Rn^{5/B})$

x - $\theta(Cu)$

ϵ_t - 0.05% rel.

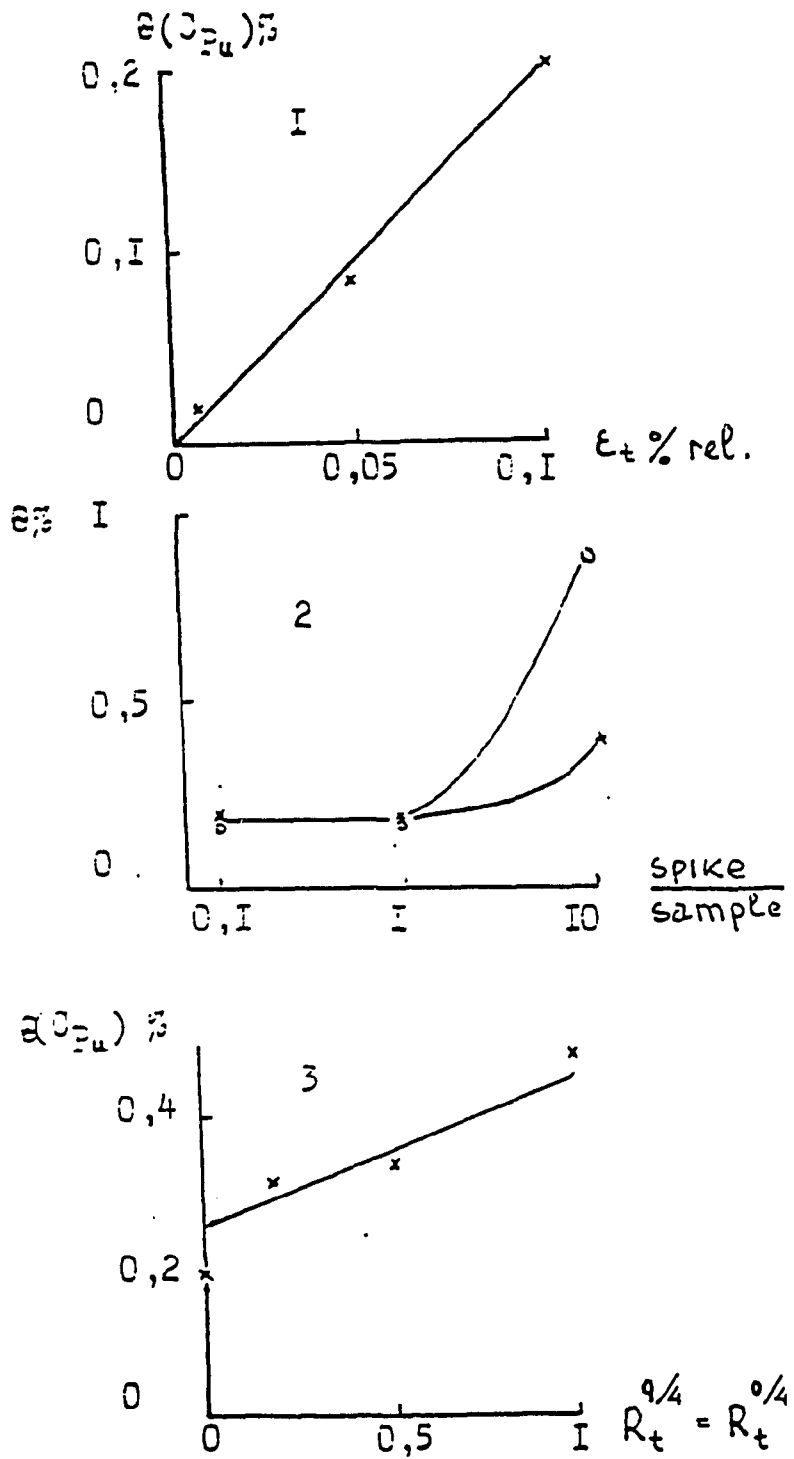


Fig.7 Effect of Various Factors on the Isotopic and Chemical Assay of Plutonium in Spent Fuel Samples.

For (2) and (3) $\varepsilon_t = 0.1\%$ rel.

o - $\varepsilon(R_n^{9/0})$

x - $\varepsilon(C_{Pu})$

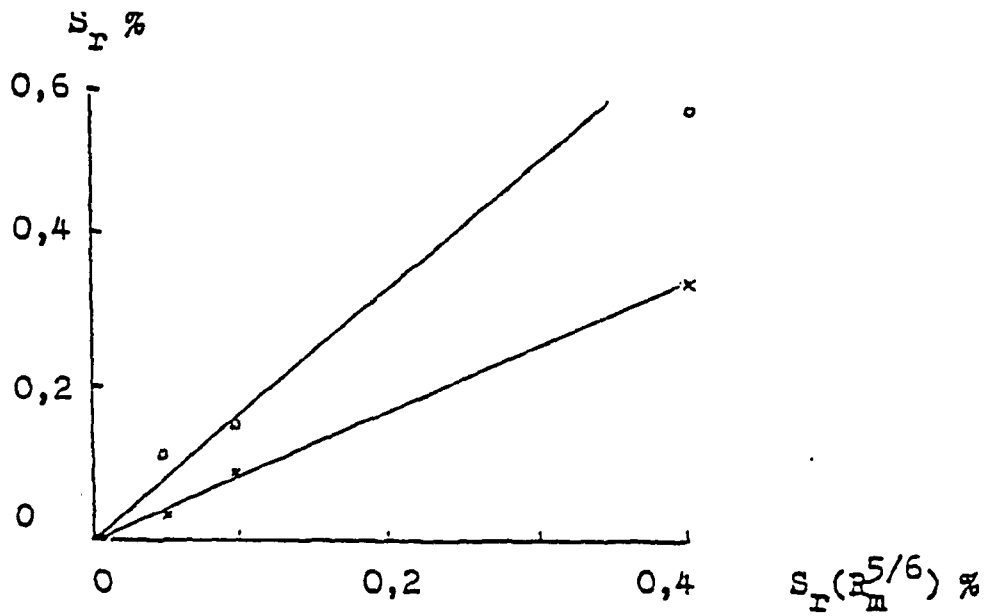


Fig.8. The Random MS Measurement Errors of $^{235}\text{U}/^{236}\text{U}$ in the Mixture. Its Effect on the Random Errors in Isotopic and Chemical Assay of Uranium in Spent Fuel Samples.

o - $Sr(R_D^{5/8})$
x - $Sr(C_U)$

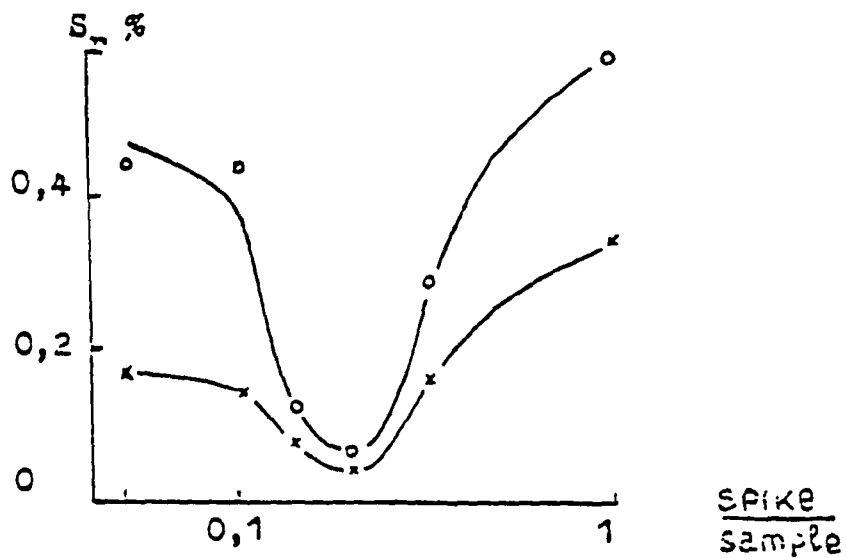


Fig.9. Effect of Spike : Sample Ratio on the Random Errors in Isotopic and Chemical Assay of Uranium samples.

o - $Sr(R_n^{5/8})$
x - $Sr(C_U)$

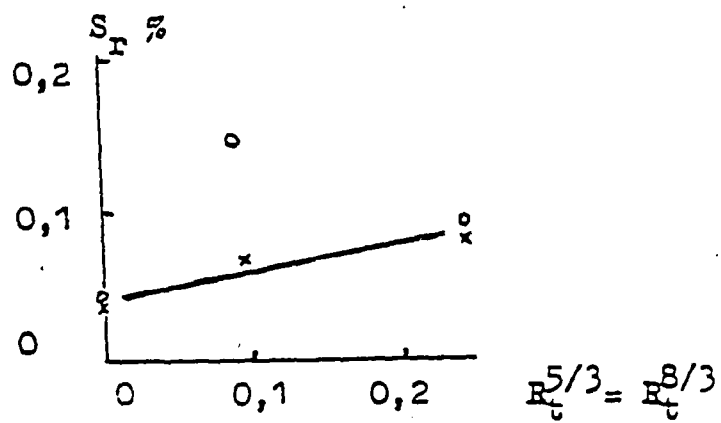


Fig.10. The Minor Isotops in the Spike. Their Effect on Random Errors in Isotopic and Chemical Assay of Uranium Sample.

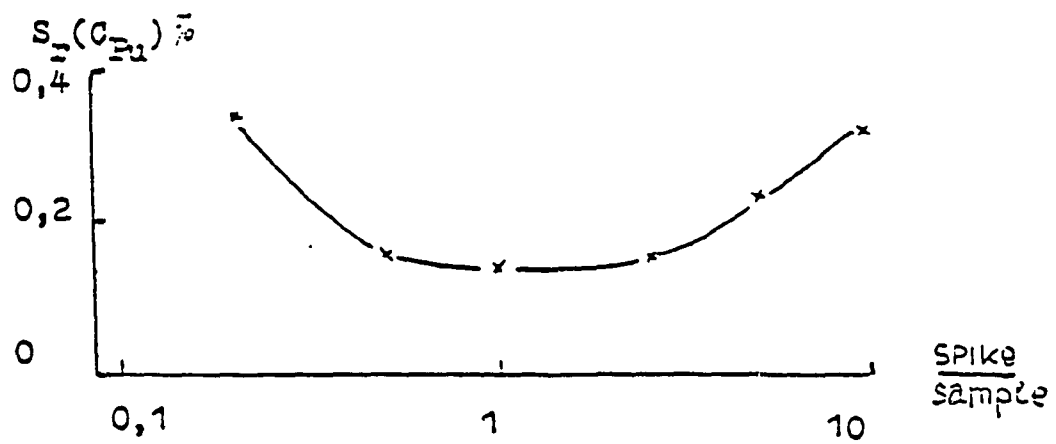


Fig.11. Effect of Spike : Sample Ratio on the Chemical Assay of Plutonium Sample.


```

10  I DSPIKE_1
20  PRINTER IS CRT
30  !CALCULATION OF ISOTOPIC COMPOSITION AND CONCENTRATION
40  !IN FUEL SAMPLES BY USE INTERNAL CALIBRATION AND
50  !DOUBLE SPIKES
60  DIM S1(20),S2(20),S3(20),S4(20),S5(20),M11(20),M21(20),M31(20)
70  DIM A(3,4),N6(4),L1(4),X(3),R(20),F1(20),F2(20)
80  DIM R1(20),R2(20),R3(20),R4(20),R5(20),C11(20)
90  DIM Y1(20),Y2(20),Y3(20),Y4(20),Y5(20)
100 !INPUT SOURCE DATA
110 INTEGER N1,N2
120 INPUT "IS SAMPLE FUEL OR DOUBLE SPIKE? 1/0",E11
130 PRINT "INPUT SAMPLE NUMBER"
140 INPUT N9
150 PRINT "INPUT SOLUTION WEIGHT SAMPLE AND SPIKE - M1,M2"
160 INPUT M1,M2
170 IF E11=0 THEN GOTO 200
180 PRINT "INPUT SPIKE TITER 236(242) MG/G OR MMG/G"
190 INPUT C
200 PRINT "URANIUM OR PLUTONIUM (1/0)"
210 INPUT E
220 PRINT "INPUT MEASUREMENT NUMBER FOR MIXTURE AND SAMPLE"
230 INPUT N1,N2
240 IF E=0 THEN GOTO D11
250 PRINT "INPUT 3 TRUE RATIOS IN SPIKE 3/6, 5/6, 8/6"
260 INPUT T1,T2,T3
270 !D - MASS DIFFERENCES
280 D1=3
290 D2=1
300 D3=-2
310 D4=2
320 D5=0
330 GOTO D12
340 D11:PRINT "INPUT 3 TRUE RATIOS IN SPIKE 4/2, 9/2, 0/2"
350 INPUT T1,T2,T3
360 D1=-2
370 D2=3
380 D3=2
390 D4=4
400 D5=1
410 D12:A1=0
420     A2=0
430     A3=0
440     A4=0
450     A5=0
460     B7=0
470     B8=0
480     B9=0
490 FOR I=1 TO N2
500 PRINT "INPUT 5 MEASUREMENT RATIOS IN SAMPLE"
510 PRINT "3/6(4/2),5/6(9/2),8/6(0/2),4/6(8/2),7/6(1/2)"
520 INPUT S1(I),S2(I),S3(I),S4(I),S5(I)
530 A1=A1+S1(I)
540 A2=A2+S2(I)
550 A3=A3+S3(I)
560 A4=A4+S4(I)
570 A5=A5+S5(I)
580 NEXT I
590 A1=A1/N2
600 A2=A2/N2
610 A3=A3/N2
620 A4=A4/N2
630 A5=A5/N2
640 FOR I=1 TO N1
650 PRINT "INPUT 3 MEASUREMENT RATIOS IN MIXTURE"
660 PRINT "3/6(4/2),5/6(9/2),8/6(0/2)"

```

```

670 INPUT M11(I),M21(I),M31(I)
680 B7=B7+M11(I)
690 B8=B8+M21(I)
700 B9=B9+M31(I)
710 NEXT I
720 B7=B7/N1
730 B8=B8/N1
740 B9=B9/N1
750 PRINT "SOURCE DATA"
760 IF E=0 THEN GOTO D13
770 PRINT "ELEMENT - URANIUM"
780 GOTO D14
790 D13:PRINT "ELEMENT - PLUTONIUM"
800 D14:PRINT "SAMPLE NUMBER";N9
810 PRINT "SAMPLE WEIGHT";M1
820 PRINT "SPIKE WEIGHT";M2
830 PRINT "SPIKE TITER";C
840 PRINT "SOURCE ISOTOPIC COMPOSITION"
850 PRINT "SAMPLE"
860 PRINT "      3/6(4/2)      5/6(9/2)      8/6(0/2)      4/6(8/2)      7/5(1/2)"
870 FOR I=1 TO N2
880 PRINT USING I11;S1(I),S2(I),S3(I),S4(I),S5(I)
890 I11:IMAGE 5(3X,3D.6D)
900 NEXT I
910 PRINT "MEAN"
920 PRINT USING I12;A1,A2,A3,A4,A5
930 I12:IMAGE 5(3X,3D.6D)
940 PRINT "MIXTURE"
950 PRINT "      3/6(4/2)      5/6(9/2)      8/6(0/2)"
960 FOR I=1 TO N1
970 PRINT USING I13;M11(I),M21(I),M31(I)
980 I13:IMAGE 3(3X,3D.6D)
990 NEXT I
1000 PRINT "MEAN"
1010 PRINT USING I14;B7,B8,B9
1020 I14:IMAGE 3(3X,3D.6D)
1025 IF E25=1 THEN GOTO 1910
1030!SOLUTION OF 3 LINEAR EQUATION SYSTEM
1040!BY MEANS GAUSS-JORDAN
1050!THE PROGRAM WAS TAKEN FROM THE BOOK BY K.J.JONSON
1060!MARCEL DEKKER, INC. 1980
1070!RANGE OF SYSTEM N3=3
1080!MATRIX OF COEFFICIENTS AND RIGHT PARTS A(3,4)
1090 B1=0
1100 B2=0
1110 B3=0
1120 B4=0
1130 B5=0
1140 FOR I=1 TO N1
1150 A(1,1)=D1*M11(I)
1160 A(1,2)=M11(I)-A1
1170 A(1,3)=-D1*A1
1180 A(1,4)=T1-M11(I)
1190 A(2,1)=D2*M21(I)
1200 A(2,2)=M21(I)-A2
1210 A(2,3)=-D2*A2
1220 A(2,4)=T2-M21(I)
1230 A(3,1)=D3*M31(I)
1240 A(3,2)=M31(I)-A3
1250 A(3,3)=-D3*A3
1260 A(3,4)=T3-M31(I)
1270 !BEGINING OF PROGRAM GAUJOR
1280 N3=3
1290 N4=N3+1
1300 I1=0
1310 FOR T=1 TO N3

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1320 N6(T)=0
1330 NEXT T
1340 IGROUND CYCLE; EXCLUDING SUCCESSIVE COLUMNS
1350 FOR T=1 TO N3
1360 N5=T+1
1370 I DETERMINATION OF MAX ELEMENT FOR 1-st COLUMN
1380 C1=0
1390 FOR K=1 TO N3
1400 E1=ABS(A(K,T))
1410 IF E1<C1 THEN GOTO D15
1420 !WAS GIVEN LINE USED AS LIDING LINE?
1430 IF N6(K)=1 THEN GOTO D15
1440 L1(T)=K
1450 C1=E1
1460 D15:NEXT K
1470 !IS MATRIX DEGENERATE?
1480 IF C1<10^(-25) THEN GOTO D17
1490 L=L1(T)
1500 N6(L)=1
1510 !PERFORM EXCLUDING: L-LEADING LINE, A(L,T)-LEADING ELEMENT
1520 FOR J=1 TO N3
1530 IF J=L THEN GOTO D16
1540 F=-A(J,T)/A(L,T)
1550 FOR K=N5 TO N4
1560 A(J,K)=A(J,K)+F*A(L,K)
1570 NEXT K
1580 D16:NEXT J
1590 NEXT T !CALCULATION OF SOLUTION OF VECTOR X
1600 FOR T=1 TO N3
1610 L=L1(T)
1620 X(T)=A(L,N4)/A(L,T)
1630 NEXT T
1640 GOTO D18
1650 D17:PRINT "MATRIX IS DEGENERATE"
1660 STOP
1670 !THE END OF GAUJOR PROGRAM
1680 D18: !THE ROOTS OF SYSTEM
1690 !R(I) - THE RATIO OF MOL QUALITY 236(242) FOR SAMPLE AND SPIKE
1700 !F1(I),F2(I) - THE DISCRIMINATION FACTORS FOR MIXTURE AND SAMPLE
1710 R(I)=X(2)
1720 F1(I)=X(1)/(1+X(2))
1730 F2(I)=X(3)/X(2)
1740 !CALCULATION OF NORMALIZED RATIOS IN SAMPLE
1750 R1(I)=A1*(1+D1*F2(I))
1760 R2(I)=A2*(1+D2*F2(I))
1770 R3(I)=A3*(1+D3*F2(I))
1780 R4(I)=A4*(1+D4*F2(I))
1790 R5(I)=A5*(1+D5*F2(I))
1800 B1=B1+R1(I)
1810 B2=B2+R2(I)
1820 B3=B3+R3(I)
1830 B4=B4+R4(I)
1840 B5=B5+R5(I)
1850 NEXT I
1860 B1=B1/N1
1870 B2=B2/N1
1880 B3=B3/N1
1890 B4=B4/N1
1900 B5=B5/N1
1910 IF E11=1 THEN GOTO D251
1920 Z1=0
1930 Z2=0
1940 Z3=0
1950 Z4=0
1960 Z5=0
1970 FOR I=1 TO N1

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1980 Z1=Z1+(B1-R1(I))^2
1990 Z2=Z2+(B2-R2(I))^2
2000 Z3=Z3+(B3-R3(I))^2
2010 Z4=Z4+(B4-R4(I))^2
2020 Z5=Z5+(B5-R5(I))^2
2030 NEXT I
2040 Z1=SQR(Z1/(N1-1))
2050 Z2=SQR(Z2/(N1-1))
2060 Z3=SQR(Z3/(N1-1))
2070 Z4=SQR(Z4/(N1-1))
2080 Z5=SQR(Z5/(N1-1))
2090 PRINT "CALCULATION RESULTS"
2100 IF E=0 THEN GOTO D521
2110 PRINT "NORMALIZED RATIOS FOR DOUBLE SPIKE (URANIUM)"
2120 PRINT "      3/6          4/6          5/6          8/6"
2130 FOR I=1 TO N1
2140 PRINT USING I21;R1(I),R4(I),R2(I),R3(I)
2150 I21:IMAGE 4(3X,3D.6D)
2160 NEXT I
2170 PRINT "MEAN"
2180 PRINT USING I22;B1,B4,B2,B3
2190 I22:IMAGE 4(3X,3D.6D)
2200 PRINT "SD"
2210 PRINT USING I23;Z1,Z4,Z2,Z3
2220 I23:IMAGE 4(3X,3D.6D)
2230 PRINT "RSD"
2240 PRINT USING I24;Z1/B1,Z4/B4,Z2/B2,Z3/B3
2250 I24:IMAGE 4(3X,3D.6D)
2260 GOTO 3351
2270 D521:PRINT "NORMALIZED RATIOS FOR DOUBLE SPIKE (PLUTONIUM)"
2280 PRINT "      8/2          9/2          0/2          1/2          4/2"
2290 FOR I=1 TO N1
2300 PRINT USING I25;R4(I),R2(I),R3(I),R5(I),R1(I)
2310 I25:IMAGE 5(3X,3D.6D)
2320 NEXT I
2330 PRINT "MEAN"
2340 PRINT USING I26;B4,B2,B3,B5,B1
2350 I26:IMAGE 5(3X,3D.6D)
2360 PRINT "SD"
2370 PRINT USING I27;Z4,Z2,Z3,Z5,Z1
2380 I27:IMAGE 5(3X,3D.6D)
2390 PRINT "RSD"
2400 PRINT USING I28;Z4/B4,Z2/B2,Z3/B3,Z5/B5,Z1/B1
2410 I28:IMAGE 5(3X,3D.6D)
2420 GOTO 3351
2430 D251: !CALCULATION OF ELEMENT ASSAY
2440 IF E=0 THEN GOTO D19
2450 H1=236.046
2460 H=233.04*B1+234.041*B4+235.044*B2+H1+238.051*B3
2470 GOTO D191
2480 D19:H1=242.059
2490 H=238.05*B4+239.052*B2+240.054*B3+241.057*B5+H1+244.064*B1
2500 D191:P1=0
2510 P2=0
2520 FOR I=1 TO N1
2530 C11(I)=R(I)*C*H*M2/M1/H1
2540 P1=P1+C11(I)
2550 NEXT I
2560 P1=P1/N1
2570 FOR I=1 TO N1
2580 P2=P2+(P1-C11(I))^2
2590 NEXT I
2600 IF N1=1 THEN GOTO D20
2610 P2=SQR(P2/(N1-1))
2620 D20:IF E=0 THEN GOTO D21
2630 FOR I=1 TO N1

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2640 Y1(I)=R1(I)/R3(I)
2650 Y2(I)=R4(I)/R3(I)
2660 Y3(I)=R2(I)/R3(I)
2670 Y4(I)=1/R3(I)
2680 Y5(I)=0
2690 NEXT I
2700 Y11=B1/B3
2710 Y21=B4/B3
2720 Y31=B2/B3
2730 Y41=1/B3
2740 Y51=0
2750 GOTO D22
2760 D21:FOR I=1 TO N1
2770 Y1(I)=R4(I)/R2(I)
2780 Y2(I)=R3(I)/R2(I)
2790 Y3(I)=R5(I)/R2(I)
2800 Y4(I)=1/R2(I)
2810 Y5(I)=R1(I)/R2(I)
2820 NEXT I
2830 Y11=B4/B2
2840 Y21=B3/B2
2850 Y31=B5/B2
2860 Y41=1/B2
2870 Y51=B1/B2
2880 D22:IF N1=1 THEN GOTO D23
2890 Z1=0
2900 Z2=0
2910 Z3=0
2920 Z4=0
2930 Z5=0
2940 FOR I=1 TO N1
2950 Z1=Z1+(Y11-Y1(I))^2
2960 Z2=Z2+(Y21-Y2(I))^2
2970 Z3=Z3+(Y31-Y3(I))^2
2980 Z4=Z4+(Y41-Y4(I))^2
2990 Z5=Z5+(Y51-Y5(I))^2
3000 NEXT I
3010 Z1=SQR(Z1/(N1-1))
3020 Z2=SQR(Z2/(N1-1))
3030 Z3=SQR(Z3/(N1-1))
3040 Z4=SQR(Z4/(N1-1))
3050 IF E=1 THEN GOTO D23
3060 Z5=SQR(Z5/(N1-1))
3070 GOTO D23
3080 Z5=0
3090 D23:PRINT "CALCULATION RESULTS"
3100 IF E=0 THEN GOTO D24
3110 PRINT "URANIUM ISOTOPIC COMPOSITION AND CONCENTRATION FOR SAMPLE"
3120 PRINT "      3/8          4/8          5/8          6/8          Cu"
3130 FOR I=1 TO N1 STEP 1
3140 PRINT USING I31;Y1(I),Y2(I),Y3(I),Y4(I),C11(I)
3150 I31:IMAGE 5(3X,3D.6D)
3160 NEXT I
3170 PRINT "MEAN"
3180 PRINT USING I32;Y11,Y21,Y31,Y41,P1
3190 I32:IMAGE 5(3X,3D.6D)
3200 PRINT "SD"
3210 PRINT USING I33;Z1,Z2,Z3,Z4,P2
3220 I33:IMAGE 5(3X,3D.6D)
3230 GOTO 3351
3240 D24:PRINT "PLUTONIUM ISOTOPIC COMPOSITION AND CONCENTRATION FOR SAMPLE"
3250 PRINT "      8/9          0/9          1/9          2/9          4/9          CPU"
"
3260 FOR I=1 TO N1
3270 PRINT USING I34;Y1(I),Y2(I),Y3(I),Y4(I),Y5(I),C11(I)
3280 I34:IMAGE 6(3X,3D.6D)

```

```
3290 NEXT I
3300 PRINT "MEAN"
3310 PRINT USING I35;Y11,Y21,Y31,Y41,Y51,P1
3320 I35:IMAGE 6(3X,3D.6D)
3330 PRINT "SD"
3340 PRINT USING I36;Z1,Z2,Z3,Z4,Z5,P2
3350 I36:IMAGE 6(3X,3D.6D)
3351 INPUT "IS THERE NEED PRINT ON PRINTER ? 1/0",E25
3352 IF E25=1 THEN GOTO 3354
3353 GOTO 3360
3354 PRINTER IS PRT
3355 GOTO 750
3360 END
```

```

10 IDS_SYS_ER1
20 PRINTER IS CRT
30 !MODEL OF SYSTEMATIC ERROR FOR ISOTOPIC COMPOSITION
40 !AND URANIUM AND PLUTONIUM ASSAY IN FUEL SAMPLES
50 !BY USING INTERNAL CALIBRATION AND DOUBLE SPIKE'
60 DIM S11(100),S21(100),S31(100),M11(100),M21(100),M31(100),A(3,4),N6(4)
70 DIM L1(4),X(3),R(100),R1(100),R2(100),P3(100),J(6),Y1(100),F11(100)
80 DIM F21(100),Y2(100),Y3(100),C11(100),R6(6),S9(6),R8(6),R7(100,6)
90 DIM W1(5),W2(100,5),W3(5),W4(5),T11(100),T21(100),T31(100)
100 DIM U1(100),U2(100),U3(100),U4(100),M41(100),M51(100),M61(100)
110 DIM M71(100),M81(100),M91(100),R4(100),R5(100),Y4(100),Y5(100)
120 !INPUT SOURCE DATA FOR MODEL
130 PRINT "IS SAMPLE FUEL OR DOUBLE SPIKE ? 1/0"
140 INPUT E11
150 INPUT "ENTER MEASUREMENT NUMBER FOR MIXTURE N<100",N
160 N1=N
170 N2=N
180 INPUT "URANIUM OR PLUTONIUM 1/0",E
190 IF E=0 THEN GOTO D31
200 PRINT "MODEL OF ERROR FOR URANIUM"
210 D1=3
220 D2=1
230 D3=-2
240 D4=2
250 D5=0
260 GOTO D32
270 D31: PRINT "MODEL OF ERROR FOR PLUTONIUM"
280 D1=-2
290 D2=3
300 D3=2
310 D4=4
320 D5=0
330 GOTO D331
340 D32:PRINT "ENTER AT. % ISOTOPE IN SPIKE 3, 5, 6, 8"
350 GOTO D33
360 D331:PRINT "ENTER AT. % ISOTOPE IN SPIKE 4, 9, 2, 0"
370 D33:INPUT W1(1),W1(2),W1(3),W1(4)
380 T1=W1(1)/W1(3)
390 T2=W1(2)/W1(3)
400 T3=W1(4)/W1(3)
410 PRINT "ENTER 4 ERROR VALUES FOR AT.% FOR SPIKE FROM CERTIF."
420 INPUT S9(1),S9(2),S9(3),S9(4)
430 IF E=0 THEN GOTO D341
440 PRINT "ENTER AT. % ISOTOPE IN SAMPLE 3, 5, 6, 8, 4, 7=0"
450 GOTO D34
460 D341:PRINT "ENTER AT.% ISOTOPE IN SAMPLE 4, 9, 2, 0, 8, 1"
470 D34:INPUT W5,W6,W7,W8,W9,W10
480 IF E11=0 THEN GOTO 510
490 INPUT "ENTER SPIKE TITER 236(242) MG/G OR MMG/G",C
500 !CONSIDER C element (SAMPLE) = C element (SPIKE) (MOL/G)
510 INPUT "ENTER SOLUTION WEIGHTS SAMPLE AND SPIKE M1,M2",M1,M2
520 INPUT "ENTER DISCRIMINATION FACTORS FOR SAMPLE F2, MIX F1",F2,F1
530 !DATA DRAWING FOR SPIKE AT.%
540 FOR J=1 TO 4
550 W3(J)=0
560 NEXT J
570 FOR J=1 TO 4
580 N7=11
590 N8=15
600 FOR I=1 TO N
610 S8=0
620 FOR N9=N7 TO N8
630 S=RND
640 S8=S8+S
650 NEXT N9
660 Z=1.54919*S8-3.87928

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670 Z=(Z^2+97)*Z*.01
680 Z=Z*S9(J)+W1(J)
690 W2(I,J)=Z
700 W3(J)=W3(J)+W2(I,J)
710 N7=N8+1
720 N8=N7+4
730 NEXT I
740 NEXT J
750 FOR J=1 TO 4
760 W3(J)=W3(J)/N
770 W4(J)=0
780 NEXT J
790 FOR J=1 TO 4
800 FOR I=1 TO N
810 W4(J)=W4(J)+(W3(J)-W2(I,J))^2
820 NEXT I
830 NEXT J
840 FOR J=1 TO 4
850 W4(J)=SQR(W4(J)/(N-1))
860 NEXT J
870 PRINT "RESULTS OF DRAWING"
880 PRINT "SPIKE AT.% 233(244) 235(239) 236(242) 238(240)"
890 GOTO D35
900 FOR I=1 TO N
910 PRINT W2(I,1),W2(I,2),W2(I,3),W2(I,4)
920 NEXT I
930 PRINT "MEAN"
940 D35: PRINT USING I5;W3(1),W3(2),W3(3),W3(4)
950 I5: IMAGE 4(3X,3D.6D)
960 PRINT "SD"
970 PRINT USING I6;W4(1),W4(2),W4(3),W4(4)
980 I6: IMAGE 4(3X,3D.6D)
990 PRINT "RSD"
1000 IF W3(1)=0 THEN GOTO 1040
1010 PRINT USING I7;W4(1)/W3(1),W4(2)/W3(2),W4(3)/W3(3),W4(4)/W3(4)
1020 I7: IMAGE 4(3X,3D.6D)
1030 GOTO 1060
1040 PRINT USING I8;0,W4(2)/W3(2),W4(3)/W3(3),W4(4)/W3(4)
1050 I8: IMAGE 4(3X,3D.6D)
1060!CALCULATION TARGET RATIOS FOR SPIKE, SAMPLE, MIXTURE
1070!FOR SPIKE
1080 FOR I=1 TO N
1090 T1(I)=W2(I,1)/W2(I,3)
1100 T2(I)=W2(I,2)/W2(I,3)
1110 T3(I)=W2(I,4)/W2(I,3)
1120 NEXT I
1130!FOR SAMPLE
1140 S1=W5/W7
1150 S2=W6/W7
1160 S3=W8/W7
1170 S4=W9/W7
1180 S5=W10/W7
1190 A1=S1*(1+D1*F2)
1200 A2=S2*(1+D2*F2)
1210 A3=S3*(1+D3*F2)
1220 A4=S4*(1+D4*F2)
1230 A5=S5*(1+D5*F2)
1240 !FOR MIXTURE
1250 M4=0
1260 M5=0
1270 M6=0
1280 FOR I=1 TO N
1290 U1(I)=W5*M1+W2(I,1)*M2
1300 U2(I)=W6*M1+W2(I,2)*M2
1310 U3(I)=W7*M1+W2(I,3)*M2
1320 U4(I)=W8*M1+W2(I,4)*M2

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1330 M11(I)=U1(I)/U3(I)
1340 M11(I)=M11(I)*(1+D1*F1)
1350 M4=M4+M11(I)
1360 M21(I)=U2(I)/U3(I)
1370 M21(I)=M21(I)*(1+D2*F1)
1380 M5=M5+M21(I)
1390 M31(I)=U4(I)/U3(I)
1400 M31(I)=M31(I)*(1+D3*F1)
1410 M6=M6+M31(I)
1420 NEXT I
1430 M4=M4/N
1440 M5=M5/N
1450 M6=M6/N
1460 M7=0
1470 M8=0
1480 M9=0
1490 FOR I=1 TO N
1500 M7=M7+(M11(I)-M4)^2
1510 M8=M8+(M21(I)-M5)^2
1520 M9=M9+(M31(I)-M6)^2
1530 NEXT I
1540 M7=SQR(M7/(N-1))
1550 M8=SQR(M8/(N-1))
1560 M9=SQR(M9/(N-1))
1570 PRINT "SOURCE DATA (RATIOS)"
1580 PRINT "      3/6(4/2)      5/6(9/2)      8/6(0/2)"
1590 PRINT "SPIKE MEAN"
1600 PRINT USING I9;W3(1)/W3(3),W3(2)/W3(3),W3(4)/W3(3)
1610 I9: IMAGE 3(3X,3D.6D)
1620 PRINT "SAMPLE"
1630 PRINT USING I10;A1,A2,A3
1640 I10: IMAGE 3(3X,3D.6D)
1650 PRINT "MIXTURE"
1660 PRINT USING I11;M4,M5,M6
1670 I11: IMAGE 3(3X,3D.6D)
1680!SOLUTION OF 3 LINEAR EQUATION SYSTEM
1690!BY MEANS GAUSS-JORDAN
1700!THE PROGRAM WAS TAKEN FROM THE BOOK BY K.J.JONSON
1710!MARCEL DEKKER, INC, 1980
1720!RANGE OF SYSTEM N3=3
1730!MATRIX OF COEFFICIENTS AND RIGHT PARTS A(3,4)
1740 B1=0
1750 B2=0
1760 B3=0
1770 B4=0
1780 B5=0
1790 FOR I=1 TO N1
1800 A(1,1)=D1*M11(I)
1810 A(1,2)=M11(I)-A1
1820 A(1,3)=-D1*A1
1830 A(1,4)=T1-M11(I)
1840 A(2,1)=D2*M21(I)
1850 A(2,2)=M21(I)-A2
1860 A(2,3)=-D2*A2
1870 A(2,4)=T2-M21(I)
1880 A(3,1)=D3*M31(I)
1890 A(3,2)=M31(I)-A3
1900 A(3,3)=-D3*A3
1910 A(3,4)=T3-M31(I)
1920 !BEGINING OF PROGRAM GAUJOK
1930 N3=3
1940 N4=N3+1
1950 I1=0
1960 FOR T=1 TO N3
1970 N6(T)=0
1980 NEXT T

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1990 !GROUND CYCLE; EXCLUDING SUCCESSIVE COLUMNS
2000 FOR T=1 TO N3
2010 N5=T+1
2020 !DETERMINATION OF MAX ELEMENT FOR 1-st COLUMN
2030 C1=0
2040 FOR K=1 TO N3
2050 E1=ABS(A(K,T))
2060 IF E1<C1 THEN GOTO D15
2070 !WAS GIVEN LINE USED AS LEADING LINE?
2080 IF N6(K)=1 THEN GOTO D15
2090 L1(T)=K
2100 C1=E1
2110 D15:NEXT K
2120 !IS MATRIX DEGENERATE?
2130 IF C1<10^(-25) THEN GOTO D17
2140 L=L1(T)
2150 N6(L)=1
2160 !PERFORM EXCLUDING: L-LEADING LINE, A(L,T)-LEADING ELEMENT
2170 FOR J=1 TO N3
2180 IF J=L THEN GOTO D16
2190 F=-A(J,T)/A(L,T)
2200 FOR K=N5 TO N4
2210 A(J,K)=A(J,K)+F*A(L,K)
2220 NEXT K
2230 D16:NEXT J
2240 NEXT T!CALCULATION OF SOLUTION OF VECTOR X
2250 FOR T=1 TO N3
2260 L=L1(T)
2270 X(T)=A(L,N4)/A(L,T)
2280 NEXT T
2290 GOTO D18
2300 D17:PRINT "MATRIX IS DEGENERATE"
2310 STOP
2320 !THE END OF GAUJOR PROGRAM
2330 D18: !THE ROOTS OF SYSTEM
2340 !THE RATIO OF MOL QUALITY Z36(242) FOR SAMPLE AND SPIKE
2350 !F11(I),F21(I) - THE DISCRIMINATION FACTORS FOR MIXTURE AND SAMPLE
2360 R(I)=X(2)
2370 F11(I)=X(1)/(1+X(2))
2380 F21(I)=X(3)/X(2)
2390 !CALCULATION OF NORMALIZED RATIOS IN SAMPLE
2400 R1(I)=A1*(1+D1*F21(I))
2410 R2(I)=A2*(1+D2*F21(I))
2420 R3(I)=A3*(1+D3*F21(I))
2430 R4(I)=A4*(1+D4*F21(I))
2440 R5(I)=A5*(1+D5*F21(I))
2450 B1=B1+R1(I)
2460 B2=B2+R2(I)
2470 B3=B3+R3(I)
2480 B4=B4+R4(I)
2490 B5=B5+R5(I)
2500 NEXT I
2510 B1=B1/N1
2520 B2=B2/N1
2530 B3=B3/N1
2540 B4=B4/N1
2550 B5=B5/N1
2560 IF E11=1 THEN GOTO D25
2570 Z1=0
2580 Z2=0
2590 Z3=0
2600 Z4=0
2610 Z5=0
2620 FOR I=1 TO N1
2630 Z1=Z1+(B1-R1(I))^2
2640 Z2=Z2+(B2-R2(I))^2

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2650 Z3=Z3+(B3-R3(I))^2
2660 Z4=Z4+(B4-R4(I))^2
2670 Z5=Z5+(B5-R5(I))^2
2680 NEXT I
2690 Z1=SQR(Z1/(N1-1))
2700 Z2=SQR(Z2/(N1-1))
2710 Z3=SQR(Z3/(N1-1))
2720 Z4=SQR(Z4/(N1-1))
2730 Z5=SQR(Z5/(N1-1))
2740 PRINT "CALCULATION RESULTS"
2750 IF E=0 THEN GOTO D521
2760 PRINT "NORMALIZED RATIOS FOR DOUBLE SPIKE (URANIUM)"
2770 PRINT "          3/6          4/6          5/6          8/6"
2780 GOTO 2820
2790 FOR I=1 TO N1
2800 PRINT R1(I),R4(I),R2(I),R3(I)
2810 NEXT I
2820 PRINT "MEAN"
2830 PRINT USING I13;B1,B4,B2,B3
2840 I13: IMAGE 4(3X,3D.7D)
2850 PRINT "SD"
2860 PRINT USING I14;Z1,Z4,Z2,Z3
2870 I14: IMAGE 4(3X,3D.7D)
2880 PRINT "RSD"
2890 PRINT USING I15;Z1/B1,Z4/B4,Z2/B2,Z3/B3
2900 I15: IMAGE 4(3X,3D.7D)
2910 GOTO 4110
2920 D521:PRINT "NORMALIZED RATIOS FOR DOUBLE SPIKE (PLUTONIUM)"
2930 PRINT "          8/2          9/2          0/2          1/2          4/2"
2940 GOTO 2980
2950 FOR I=1 TO N1
2960 PRINT R4(I),R2(I),R3(I),R5(I),R1(I)
2970 NEXT I
2980 PRINT "MEAN"
2990 PRINT USING I16;B4,B2,B3,B5,B1
3000 I16: IMAGE 5(3X,3D.7D)
3010 PRINT "SD"
3020 PRINT USING I17;Z4,Z2,Z3,Z5,Z1
3030 I17: IMAGE 5(3X,3D.7D)
3040 PRINT "RSD"
3050 PRINT USING I18;Z4/B4,Z2/B2,Z3/B3,Z5/B5,Z1/B1
3060 I18: IMAGE 5(3X,3D.7D)
3070 GOTO 4110
3080 D25: !CALCULATION OF ELEMENT ASSAY
3090 IF E=0 THEN GOTO D19
3100 H1=236.046
3110 H=233.04*B1+234.041*B4+235.044*B2+H1+238.051*B3
3120 GOTO D191
3130 D19:H1=242.059
3140 H=238.05*B4+239.052*B2+240.054*B3+241.057*B5+H1+244.064*B1
3150 D191:P1=0
3160 P2=0
3170 FOR I=1 TO N1
3180 C11(I)=R(I)*C*H*M2/M1/H1
3190 P1=P1+C11(I)
3200 NEXT I
3210 P1=P1/N1
3220 FOR I=1 TO N1
3230 P2=P2+(P1-C11(I))^2
3240 NEXT I
3250 IF N1=1 THEN GOTO D20
3260 P2=SQR(P2/(N1-1))
3270 D20:IF E=0 THEN GOTO D21
3280 FOR I=1 TO N1
3290 Y1(I)=R1(I)/R3(I)
3300 Y2(I)=R4(I)/R3(I)

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3310 Y3(I)=R2(I)/R3(I)
3320 Y4(I)=1/R3(I)
3330 Y5(I)=0
3340 NEXT I
3350 Y11=B1/B3
3360 Y21=B4/B3
3370 Y31=B2/B3
3380 Y41=1/B3
3390 Y51=0
3400 GOTO D22
3410 D21:FOR I=1 TO N1
3420 Y1(I)=R4(I)/R2(I)
3430 Y2(I)=R3(I)/R2(I)
3440 Y3(I)=R5(I)/R2(I)
3450 Y4(I)=1/R2(I)
3460 Y5(I)=R1(I)/R2(I)
3470 NEXT I
3480 Y11=B4/B2
3490 Y21=B3/B2
3500 Y31=B5/B2
3510 Y41=1/B2
3520 Y51=B1/B2
3530 D22:IF N1=1 THEN GOTO D23
3540 Z1=0
3550 Z2=0
3560 Z3=0
3570 Z4=0
3580 Z5=0
3590 FOR I=1 TO N1
3600 Z1=Z1+(Y11-Y1(I))^2
3610 Z2=Z2+(Y21-Y2(I))^2
3620 Z3=Z3+(Y31-Y3(I))^2
3630 Z4=Z4+(Y41-Y4(I))^2
3640 Z5=Z5+(Y51-Y5(I))^2
3650 NEXT I
3660 Z1=SQR(Z1/(N1-1))
3670 Z2=SQR(Z2/(N1-1))
3680 Z3=SQR(Z3/(N1-1))
3690 Z4=SQR(Z4/(N1-1))
3700 IF E=1 THEN GOTO D23
3710 Z5=SQR(Z5/(N1-1))
3720 GOTO D23
3730 Z5=0
3740 D23:PRINT "CALCULATION RESULTS"
3750 IF E=0 THEN GOTO D24
3760 PRINT "URANIUM ISOTOPIC COMPOSITION AND CONCENTRATION FOR SAMPLE"
3770 PRINT "      3/8          4/8          5/8          6/8          Cu"
3780 GOTO 3820
3790 FOR I=1 TO N1 STEP 1
3800 PRINT Y1(I),Y2(I),Y3(I),Y4(I),C11(I)
3810 NEXT I
3820 PRINT "MEAN"
3830 PRINT USING I20;Y11,Y21,Y31,Y41,P1
3840 I20: IMAGE 5(3X,3D.6D)
3850 PRINT "SD"
3860 PRINT USING I21;Z1,Z2,Z3,Z4,P2
3870 I21: IMAGE 5(3X,3D.6D)
3880 IF Y11=0 THEN GOTO 3910
3890 Z11=Z1/Y11
3900 GOTO 3920
3910 Z11=0
3920 PRINT "RSD"
3930 PRINT USING I22;Z11,Z2/Y21,Z3/Y31,Z4/Y41,P2/P1
3940 I22: IMAGE 5(3X,3D.6D)
3950 GOTO 4110
3960 D24:PRINT "PLUTONIUM ISOTOPIC COMPOSITION AND CONCENTRATION FOR SAMPLE"

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3970 PRINT "      8/9      0/9      1/9      2/9      4/9
      CPU"
3980 GOTO 4020
3990 FOR I=1 TO N1
4000 PRINT Y1(I),Y2(I),Y3(I),Y4(I),Y5(I),C11(I)
4010 NEXT I
4020 PRINT "MEAN"
4030 PRINT USING I23;Y11,Y21,Y31,Y41,Y51,P1
4040 I23:IMAGE 6(3X,3D.6D)
4050 PRINT "SD"
4060 PRINT USING I24;Z1,Z2,Z3,Z4,Z5,P2
4070 I24:IMAGE 6(3X,3D.6D)
4071 IF Y51=0 THEN GOTO 4074
4072 Z51=Z5/Y51
4073 GOTO 4080
4074 Z51=0
4080 PRINT "RSD"
4090 PRINT USING I25;Z1/Y11,Z2/Y21,Z3/Y31,Z4/Y41,Z51,P2/P1
4100 I25:IMAGE 6(3X,3D.6D)
4110 INPUT "IS THERE NEED PRINT ON PRINTER ? 1/0",E25
4120 IF E25=1 THEN GOTO 4140
4130 GOTO 4160
4140 PRINTER IS PRT
4150 GOTO 870
4160 END
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10 IDS_RND_ER1
20 PRINTER IS CRT
30 !MODEL OF RANDOM ERROR FOR ISOTOPIC COMPOSITION
40 !AND URANIUM AND PLUTONIUM ASSAY IN FUEL SAMPLES
50 !BY USING INTERNAL CALIBRATION AND DOUBLE SPIKES
60 DIM S1(100),S2(100),S3(100),M11(100),M21(100),M31(100),A(3,4),R7(100,6)
70 DIM N6(4),L1(4),X(3),R(100),R1(100),R2(100),R3(100),R4(100),R9(6),Y1(100)
80 DIM F11(100),F21(100),Y2(100),Y3(100),C11(100),R6(6),S9(6),R8(6),R5(100)
90 DIM Y4(100),Y5(100)
100 !INPUT SOURCE DATA FOR MODEL
110 INPUT "IS SAMPLE SPENT FUEL OR DOUBLE SPIKE 1/0 ?",E11
120 PRINT "INPUT MEASUREMENTS NUMBER FOR MIXTURE   N<100"
130 INPUT N
140 N1=N
150 N2=N
160 PRINT "URANIUM OR PLUTONIUM  1/0"
170 INPUT E
180 IF E=0 THEN GOTO D41
190 PRINT "MODEL OF ERROR FOR URANIUM"
200 GOTO D42
210 D41:PRINT "MODEL OF ERROR  FOR PLUTONIUM"
220 GOTO D43
230 D42:PRINT "INPUT AT.% ISOTOPES IN SPIKE 3, 5, 6,  8"
240 GOTO D44
250 D43:PRINT "INPUT AT.% ISOTOPES IN SPIKE 4, 9, 2,  0"
260 D44:   INPUT W1,W2,W3,W4
270 IF E=0 THEN GOTO D45
280 PRINT "INPUT AT.% ISOTOPES IN SAMPLE 3, 5, 6,  8"
290 GOTO D46
300 D45:PRINT "INPUT AT.% ISOTOPES IN SAMPLE 4, 9, 2,  0"
310 D46:INPUT W5,W6,W7,W8
320 IF E11=0 THEN GOTO 350
330 PRINT "INPUT SPIKE TITER 236(242) MG/G OR MMG/G"
340 INPUT C
350 !CONSIDER C11(SAMPLE)=C11(SPIKE) (MOL/G)
360   INPUT "SOLUTION WEIGHTS SAMPLE AND SPIKE M1, M2",M1,M2
370 !CALCULATION OF AT.% ISOTOPE IN MIXTURE U1, U2, U3, U4
380 U1=W1*M2+W5*M1
390 U2=W2*M2+W6*M1
400 U3=W3*M2+W7*M1
410 U4=W4*M2+W8*M1
420 !CALCULATION OF TRUE RATIOS FOR SPIKE, SAMPLE AND MIXTURE
430 T1=W1/W3
440 T2=W2/W3
450 T3=W4/W3
460 R6(1)=W5/W7
470 R6(2)=W6/W7
480 R6(3)=W8/W7
490 R6(4)=U1/U3
500 R6(5)=U2/U3
510 R6(6)=U4/U3
520 PRINT "INPUT THE DISCRIMINATION FACTOR VALUES"
530 PRINT "FOR SAMPLE F2 AND MIXTURE F1"
540 INPUT F1,F2
550 !D - MASS DIFFERENCES
560 IF E=0 THEN GOTO D47
570 D1=3
580 D2=1
590 D3=-2
600 GOTO D48
610 D47:D1=-2
620 D2=3
630 D3=2
640 D48:!CALCULATION OF MEASURED RATIOS FOR SAMPLE AND MIXTURE
650 R6(1)=R6(1)*(1+D1*F2)
660 R6(2)=R6(2)*(1+D2*F2)

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670 R6(3)=R6(3)*(1+D3*F2)
680 R6(4)=R6(4)*(1+D1*F1)
690 R6(5)=R6(5)*(1+D2*F1)
700 R6(6)=R6(6)*(1+D3*F1)
710 PRINT "TARGET MEAN MEASUREMENT RATIOS"
720 PRINT "      3/6(4/2)      5/6(9/2)      8/6(0/2)"
730 PRINT "SAMPLE"
740 PRINT USING I15;R6(1),R6(2),R6(3)
750 I15:IMAGE 3(3X,3D.6D)
760 PRINT " MIXTURE"
770 PRINT USING I16;R6(4),R6(5),R6(6)
780 I16: IMAGE 3(3X,3D.6D)
790 PRINT "INPUT RANDOM ERROR - RSD % OF MEASUREMENT"
800 PRINT "3 VALUES FOR SAMPLE - FOR 3/6, 5/6, 8/6 AND THE SAME FOR MIXTURE"
810 FOR I=1 TO 6
820 INPUT S9(I)
830 NEXT I
840 PRINT "TARGET MEAN MEASUREMENT RATIOS AND THEIR RSD %"
850 PRINT "      3/6(4/2)      5/6(9/2)      8/6(0/2)"
860 PRINT "SPIKE"
870 PRINT USING I17;T1,T2,T3
880 I17: IMAGE 3(3X,3D.6D)
890 PRINT "SAMPLE"
900 PRINT USING I18;R6(1),R6(2),R6(3)
910 I18: IMAGE 3(3X,3D.6D)
920 PRINT "RSD %"
930 PRINT USING I19;S9(1),S9(2),S9(3)
940 I19: IMAGE 3(3X,3D.6D)
950 PRINT "MIXTURE"
960 PRINT USING I20;R6(4),R6(5),R6(6)
970 I20:IMAGE 3(3X,3D.6D)
980 PRINT "RSD %"
990 PRINT USING I21;S9(4),S9(5),S9(6)
1000 I21:IMAGE 3(3X,3D.6D)
1010!DATA DRAWING FOR MEASUREMENT RATIOS
1020 FOR I=1 TO 6
1030 R8(I)=0
1040 NEXT I
1050 FOR J=1 TO 6
1060 N7=11
1070 N8=15
1080 FOR I=1 TO N
1090 S8=0
1100 FOR N9=N7 TO N8
1110 S=RND
1120 S8=S8+S
1130 NEXT N9
1140 Z=1.54919*S8-3.87928
1150 Z=(Z^2+97)*Z*.01
1160 Z=Z*(S9(J)*R6(J)/100)+R6(J)
1170 R7(I,J)=Z
1180 R8(J)=R8(J)+R7(I,J)
1190 N7=N8+1
1200 N8=N7+4
1210 NEXT I
1220 NEXT J
1230 FOR I=1 TO N
1240 S1(I)=R7(I,1)
1250 S2(I)=R7(I,2)
1260 S3(I)=R7(I,3)
1270 M11(I)=R7(I,4)
1280 M21(I)=R7(I,5)
1290 M31(I)=R7(I,6)
1300 NEXT I
1310 FOR J=1 TO 6
1320 R8(J)=R8(J)/N

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1330 R9(J)=0
1340 NEXT J
1350 FOR J=1 TO 6
1360 FOR I=1 TO N
1370 R9(J)=R9(J)+(R8(J)-R7(I,J))^2
1380 NEXT I
1390 NEXT J
1400 FOR J=1 TO 6
1410 R9(J)=SQR(R9(J)/(N-1))
1420 NEXT J
1430 PRINT "SOURCE DATA FOR CALCULATION"
1440 IF E=0 THEN GOTO D52
1450 PRINT "ELEMENT - URANIUM"
1460 GOTO D53
1470 D52:PRINT "ELEMENT - PLUTONIUM"
1480 D53:PRINT "SAMPLE WEIGHT";M1
1490 PRINT "SPIKE WEIGHT";M2
1500 PRINT "SPIKE TITER";C
1510 PRINT "ISOTOPIC COMPOSITION"
1520 PRINT "      3/6(4/2)          5/6(9/2)          8/6(0/2)"
1530 PRINT "SAMPLE"
1540 GOTO D49
1550 FOR I=1 TO N
1560 PRINT S1(I),S2(I),S3(I)
1570 NEXT I
1580 D49:PRINT "MEAN"
1590     PRINT USING I22;R8(1),R8(2),R8(3)
1600 I22: IMAGE 3(3X,3D.6D)
1610 PRINT "SD"
1620 PRINT USING I23;R9(1),R9(2),R9(3)
1630 I23: IMAGE 3(3X,3D.6D)
1640 PRINT "RSD"
1650 IF R8(1)=0 THEN GOTO 1680
1660 PRINT USING I24;R9(1)/R8(1),R9(2)/R8(2),R9(3)/R8(3)
1670 I24: IMAGE 3(3X,3D.6D)
1680     PRINT USING I25;0,R9(2)/R8(2),R9(3)/R8(3)
1690 I25: IMAGE 3(3X,3D.6D)
1700     PRINT "MIXTURE"
1710 GOTO D51
1720 FOR I=1 TO N
1730 PRINT M11(I),M21(I),M31(I)
1740 NEXT I
1750 PRINT "MEAN"
1760 D51:PRINT USING I26;R8(4),R8(5),R8(6)
1770 I26: IMAGE 3(3X,3D.6D)
1780 PRINT "SD"
1790     PRINT USING I27;R9(4),R9(5),R9(6)
1800 I27: IMAGE 3(3X,3D.6D)
1810 PRINT "RSD"
1820 PRINT USING I28;R9(4)/R8(4),R9(5)/R8(5),R9(6)/R8(6)
1830 I28: IMAGE 3(3X,3D.6D)
1840 IF E45=1 THEN GOTO 3910
1850 D54:A1=R8(1)
1860 A2=R8(2)
1870 A3=R8(3)
1880 D55:B7=R8(4)
1890 B8=R8(5)
1900 B9=R8(6)
1910!SOLUTION OF 3 LINEAR EQUATION SYSTEM
1920!BY MEANS GAUSS-JORDAN
1930!THE PROGRAM WAS TAKEN FROM THE BOOK BY K.J.JONSON
1940!MARCEL DEKKER, INC, 1980
1950!RANGE OF SYSTEM N3=3
1960!MATRIX OF COEFFICIENTS AND RIGHT PARTS A(3,4)
1970 B1=0
1980 B2=0

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1990 B3=0
2000 B4=0
2010 B5=0
2020 FOR I=1 TO N1
2030 A(1,1)=D1*M11(I)
2040 A(1,2)=M11(I)-A1
2050 A(1,3)=-D1*A1
2060 A(1,4)=T1-M11(I)
2070 A(2,1)=D2*M21(I)
2080 A(2,2)=M21(I)-A2
2090 A(2,3)=-D2*A2
2100 A(2,4)=T2-M21(I)
2110 A(3,1)=D3*M31(I)
2120 A(3,2)=M31(I)-A3
2130 A(3,3)=-D3*A3
2140 A(3,4)=T3-M31(I)
2150 !BEGINING OF PROGRAM GAUJOR
2160 N3=3
2170 N4=N3+1
2180 I1=0
2190 FOR T=1 TO N3
2200 N6(T)=0
2210 NEXT T
2220 !GROUND CYCLE; EXCLUDING SUCCESSIVE COLUMNS
2230 FOR T=1 TO N3
2240 N5=T+1
2250 !DETERMINATION OF MAX ELEMENT FOR 1-st COLUMN
2260 C1=0
2270 FOR K=1 TO N3
2280 E1=ABS(A(K,T))
2290 IF E1<C1 THEN GOTO D15
2300 !WAS GIVEN LINE USED AS LIDING LINE?
2310 IF N6(K)=1 THEN GOTO D15
2320 L1(T)=K
2330 C1=E1
2340 D15:NEXT K
2350 !IS MATRIX DEGENERATE?
2360 IF C1<10^(-25) THEN GOTO D17
2370 L=L1(T)
2380 N6(L)=1
2390 !PERFORM EXCLUDING: L-LEADING LINE, A(L,T)-LEADING ELEMENT
2400 FOR J=1 TO N3
2410 IF J=L THEN GOTO D16
2420 F=-A(J,T)/(A(L,T))
2430 FOR K=N5 TO N4
2440 A(J,K)=A(J,K)+F*A(L,K)
2450 NEXT K
2460 D16:NEXT J
2470 NEXT T !CALCULATION OF SOLUTION OF VECTOR X
2480 FOR T=1 TO N3
2490 L=L1(T)
2500 X(T)=A(L,N4)/A(L,T)
2510 NEXT T
2520 GOTO D18
2530 D17:PRINT "MATRIX IS DEGENERATE"
2540 STOP
2550 !THE END OF GAUJOR PROGRAM
2560 D18:!THE ROOTS OF SYSTEM
2570 !THE RATIO OF MOL QUALITY 236(242) FOR SAMPLE AND SPIKE
2580 F11(I),F21(I) - THE DISCRIMINATION FACTORS FOR MIXTURE AND SAMPLE
2590 R(I)=X(2)
2600 F11(I)=X(1)/(1+X(2))
2610 F21(I)=X(3)/X(2)
2620 !CALCULATION OF NORMALIZED RATIOS IN SAMPLE
2630 R1(I)=A1*(1+D1*F21(I))
2640 R2(I)=A2*(1+D2*F21(I))

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2650 R3(I)=A3*(1+D3*F21(I))
2660 R4(I)=A4*(1+D4*F21(I))
2670 R5(I)=A5*(1+D5*F21(I))
2680 B1=B1+R1(I)
2690 B2=B2+R2(I)
2700 B3=B3+R3(I)
2710 B4=B4+R4(I)
2720 B5=B5+R5(I)
2730 NEXT I
2740 B1=B1/N1
2750 B2=B2/N1
2760 B3=B3/N1
2770 B4=B4/N1
2780 B5=B5/N1
2790 IF E11=1 THEN GOTO D251
2800 Z1=0
2810 Z2=0
2820 Z3=0
2830 Z4=0
2840 Z5=0
2850 FOR I=1 TO N1
2860 Z1=Z1+(B1-R1(I))^2
2870 Z2=Z2+(B2-R2(I))^2
2880 Z3=Z3+(B3-R3(I))^2
2890 Z4=Z4+(B4-R4(I))^2
2900 Z5=Z5+(B5-R5(I))^2
2910 NEXT I
2920 Z1=SQR(Z1/(N1-1))
2930 Z2=SQR(Z2/(N1-1))
2940 Z3=SQR(Z3/(N1-1))
2950 Z4=SQR(Z4/(N1-1))
2960 Z5=SQR(Z5/(N1-1))
2970 PRINT "CALCULATION RESULTS"
2980 IF E=0 THEN GOTO D521
2990 PRINT "NORMALIZED RATIOS FOR DOUBLE SPIKE (URANIUM)"
3000 PRINT " 3/6      4/6      5/6      8/6"
3010 GOTO 3050
3020 FOR I=1 TO N1
3030 PRINT R1(I),R4(I),R2(I),R3(I)
3040 NEXT I
3050 PRINT "MEAN";B1,B4,B2,B3
3060 PRINT "SD";Z1,Z4,Z2,Z3
3070 IF B4=0 THEN GOTO 3100
3080 PRINT "RSD";Z1/B1,Z4/B4,Z2/B2,Z3/B3
3090 GOTO 4390
3100 PRINT "RSD";Z1/B1,0,Z2/B2,Z3/B3
3110 GOTO 4390
3120 D521:PRINT "NORMALIZED RATIOS FOR DOUBLE SPIKE (PLUTONIUM)"
3130 PRINT " 8/2      9/2      0/2      1/2      4/2"
3140 GOTO 3180
3150 FOR I=1 TO N1
3160 PRINT R4(I),R2(I),R3(I),R5(I),R1(I)
3170 NEXT I
3180 PRINT "MEAN";B4,B2,B3,B5,B1
3190 PRINT "SD";Z4,Z2,Z3,Z5,Z1
3200 IF Z4=0 AND Z5=0 THEN GOTO 3230
3210 PRINT "RSD";Z4/B4,Z2/B2,Z3/B3,Z5/B5,Z1/B1
3220 GOTO 4390
3230 PRINT "RSD";0,Z2/B2,Z3/B3,0,Z1/B1
3240 GOTO 4390
3250 D251:CALCULATION OF ELEMENT ASSAY
3260 IF E=0 THEN GOTO D19
3270 H1=236.046
3280 H=233.04*B1+234.041*B4+235.044*B2+H1+238.051*B3
3290 GOTO D191
3300 D19:H1=242.059

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3310      H=238.05*B4+239.052*B2+240.054*B3+241.057*B5+H1+244.064*B1
3320 D191:P1=0
3330 P2=0
3340 FOR I=1 TO N1
3350 C11(I)=R(I)*C*H*M2/M1/H1
3360 P1=P1+C11(I)
3370 NEXT I
3380 P1=P1/N1
3390 FOR I=1 TO N1
3400 P2=P2+(P1-C11(I))^2
3410 NEXT I
3420 IF N1=1 THEN GOTO D20
3430 P2=SQR(P2/(N1-1))
3440 D20:IF E=0 THEN GOTO D21
3450 FOR I=1 TO N1
3460 Y1(I)=R1(I)/R3(I)
3470 Y2(I)=R4(I)/R3(I)
3480 Y3(I)=R2(I)/R3(I)
3490 Y4(I)=1/R3(I)
3500 Y5(I)=0
3510 NEXT I
3520 Y11=B1/B3
3530 Y21=B4/B3
3540 Y31=B2/B3
3550 Y41=1/B3.
3560 Y51=0
3570 GOTO D22
3580 D21:FOR I=1 TO N1
3590 Y1(I)=R4(I)/R2(I)
3600 Y2(I)=R3(I)/R2(I)
3610 Y3(I)=R5(I)/R2(I)
3620 Y4(I)=1/R2(I)
3630 Y5(I)=R1(I)/R2(I)
3640 NEXT I
3650 Y11=B4/B2
3660 Y21=B3/B2
3670 Y31=B5/B2
3680 Y41=1/B2
3690 Y51=B1/B2
3700 D22:IF N1=1 THEN GOTO D23
3710 Z1=0
3720 Z2=0
3730 Z3=0
3740 Z4=0
3750 Z5=0
3760 FOR I=1 TO N1
3770 Z1=Z1+(Y11-Y1(I))^2
3780 Z2=Z2+(Y21-Y2(I))^2
3790 Z3=Z3+(Y31-Y3(I))^2
3800 Z4=Z4+(Y41-Y4(I))^2
3810 Z5=Z5+(Y51-Y5(I))^2
3820 NEXT I
3830 Z1=SQR(Z1/(N1-1))
3840 Z2=SQR(Z2/(N1-1))
3850 Z3=SQR(Z3/(N1-1))
3860 Z4=SQR(Z4/(N1-1))
3870 IF E=1 THEN GOTO D23
3880 Z5=SQR(Z5/(N1-1))
3890 GOTO D23
3900 Z5=0
3910 D23:PRINT "CALCULATION RESULTS"
3920 IF E=0 THEN GOTO D24
3930 PRINT "URANIUM ISOTOPIC COMPOSITION AND CONCENTRATION FOR SAMPLE"
3940 PRINT "      3/8          4/8          5/8          6/8          Cu"
3950 GOTO 4000
3960 FOR I=1 TO N1 STEP 1

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3970 PRINT USING IS;Y1(I),Y2(I),Y3(I),Y4(I),C11(I)
3980 IS:IMAGE 5(3X,3D.6D)
3990 NEXT I
4000 PRINT "MEAN"
4010 PRINT USING I6;Y11,Y21,Y31,Y41,P1
4020 I6:IMAGE 5(3X,3D.6D)
4030 PRINT "SD"
4040 PRINT USING I7;Z1,Z2,Z3,Z4,P2
4050 I7:IMAGE 5(3X,3D.6D)
4060 PRINT "RSD"
4070 IF Y21=0 THEN GOTO 4110
4080 PRINT USING I8;0,Z2/Y21,Z3/Y31,Z4/Y41,P2/P1
4090 I8:IMAGE 5(3X,3D.6D)
4100 GOTO 4390
4110 PRINT USING I9;0,0,Z3/Y31,Z4/Y41,P2/P1
4120 I9:IMAGE 5(3X,3D.6D)
4130 GOTO 4340
4140 D24:PRINT "PLUTONIUM ISOTOPIC COMPOSITION AND CONCENTRATION FOR SAMPLE"
4150 PRINT "      8/9          0/9          1/9          2/9          4/9          CPU"
4160 GOTO 4210
4170 FOR I=1 TO N1
4180 PRINT USING I10;Y1(I),Y2(I),Y3(I),Y4(I),Y5(I),C11(I)
4190 I10:IMAGE 6(2X,3D.6D)
4200 NEXT I
4210 PRINT "MEAN"
4220 PRINT USING I11;Y11,Y21,Y31,Y41,Y51,P1
4230 I11:IMAGE 6(2X,3D.6D)
4240 PRINT "SD"
4250 PRINT USING I12;Z1,Z2,Z3,Z4,Z5,P2
4260 I12:IMAGE 6(2X,3D.6D)
4270 PRINT "RSD"
4280 IF Y11=0 AND Y31=0 THEN GOTO 4320
4290 PRINT USING I13;Z1/Y11,Z2/Y21,Z3/Y31,Z4/Y41,Z5/Y51,P2/P1
4300 I13:IMAGE 6(2X,3D.6D)
4310 GOTO 4390
4320 PRINT USING I14;0,Z2/Y21,0,Z4/Y41,0,P2/P1
4330 I14:IMAGE 6(2X,3D.6D)
4340 INPUT "IS THERE NEED PRINT ON PRINTER ? 1/0",E45
4350 IF E45=1 THEN GOTO 4370
4360 GOTO 4390
4370 PRINTER IS PRT
4380 GOTO 1430
4390 END

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10 !CEL_IC_ER1
15 PRINTER IS CRT
16 DIM S9(4),R8(4),R(4),R7(100,4),Ct(100),Rn36(100),Rn46(100)
17 DIM Rn56(100),Rn86(100),R10(4),Su(100),X3(100),X5(100)
18 DIM X8(100),N3(100),N6(100),N8(100),Rm36(100),Rm86(100),Cn6(100)
20 !MODEL OF SYSTEMATIC ERROR FOR ELEMENT ASSAY BY
30 !CERTIFICATION OF QS87 BY USING INTERNAL CALIBRATION
40 !INPUT SOURCE DATA
50 INPUT "ENTER AT.% OF ISOTOPE FOR SPIKE (NBS 960) - 4,5,8",W4,W5,W8
60 INPUT "ENTER TRUE ISOTOPIC RATIOS FOR QS 3/6,4/6,5/6,8/6",R(1),R(2),R(3),R(4)
70 PRINT "ENTER THE ERRORS FOR RATIOS"
80 FOR I=1 TO 4
90 INPUT S9(I)
100 NEXT I
110 INPUT "ENTER THE SPIKE AND QS WEIGHTS MT, MN",Mt,Mn
120 INPUT "ENTER THE DISCRIMINATION FACTOR F",F
130 INPUT "ENTER THE ELEMENT CONCENTRATION FOR SPIKE, mgU/g",Cu
140 INPUT "ENTER THE ERROR FOR Cu, mgU/g",Et
150 INPUT "ENTER THE MIXTURE MEASUREMENTS NUMBER",N
170 FOR I=1 TO 4
180 R8(I)=0
190 NEXT I
200 FOR J=1 TO 4
210 N7=11
220 N10=15
230 FOR I=1 TO N
240 S8=0
250 FOR N9=N7 TO N10
260 S=RND
270 S8=S8+S
280 NEXT N9
290 Z=1.54919*S8-3.87928
300 Z=(Z^2+97)*Z*.01
310 Z1=Z*S9(J)+R(J)
320 Z2=Z*Et+Cu
330 R7(I,J)=Z1
340 Ct(I)=Z2
350 R8(J)=R8(J)+R7(I,J)
370 N7=N10+1
380 N10=N7+4
390 NEXT I
400 NEXT J
405 R9=0
410 FOR I=1 TO N
411 R9=R9+Ct(I)
420 Rn36(I)=R7(I,1)
430 Rn46(I)=R7(I,2)
440 Rn56(I)=R7(I,3)
450 Rn86(I)=R7(I,4)
460 NEXT I
470 R11=0
480 FOR I=1 TO 4
490 R8(I)=R8(I)/N
500 R10(I)=0
510 NEXT I
520 R9=R9/N
521 FOR I=1 TO N
522 R11=R11+(R9-Ct(I))^2
523 NEXT I
530 FOR J=1 TO 4
540 FOR I=1 TO N
550 R10(J)=R10(J)+(R8(J)-R7(I,J))^2
570 NEXT I
580 NEXT J
590 R11=SQR(R11/(N-1))
600 FOR I=1 TO 4

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610 R10(I)=SQR(R10(I)/(N-1))
620 NEXT I
630 PRINT "DRAWING MEANS"
640 PRINT "      Rn3/6      Rn4/6      Rn5/6      Rn8/6      Cu"
650 PRINT USING A1;R8(1),R8(2),R8(3),R8(4),R9
660 A1:IMAGE 5(2X,3D.6D)
670 PRINT "SD"
680 PRINT USING A2;R10(1),R10(2),R10(3),R10(4),R11
690 A2:IMAGE 5(2X,3D.6D)
700 FOR I=1 TO N
710 Su(I)=Rn36(I)+Rn46(I)+Rn56(I)+Rn86(I)+1
720 X3(I)=Rn36(I)/Su(I)
730 X5(I)=Rn56(I)/Su(I)
740 X6(I)=1/Su(I)
750 X8(I)=Rn86(I)/Su(I)
760 N3(I)=X3(I)*Mn*1
770 N6(I)=X6(I)*Mn*1
780 N8(I)=X8(I)*Mn*1+W8*Cu*.993*Mt/100
790 Rm36(I)=N3(I)/(N6(I)*(1+3*F))
800 Rm86(I)=N8(I)/(N6(I)*(1-2*F))
810 NEXT I
820 K=236.04556*Mt/(238.05079*Mn)
825 S5=0
830 FOR I=1 TO N
840 Cn6(I)=Ct(I)*K*3*Rm36(I)
850 Cn6(I)=Cn6(I)/(5*Rm86(I)*Rm36(I)-3*Rm36(I)*R(4)-2*Rm86(I)*R(1))
860 S5=S5+Cn6(I)
870 NEXT I
880 Cu6=S5/N
890 Sd=0
900 FOR I=1 TO N
910 Sd=Sd+(Cu6-Cn6(I))^2
920 NEXT I
930 Sd=SQR(Sd/(N-1))
940 PRINT "MEAN CONCENTRATION"
945 PRINT USING A3;Cu6
946 A3:IMAGE 1(2X,3D.6D)
950 PRINT "SD"
956 PRINT USING A4;Sd
957 A4:IMAGE 1(2X,3D.6D)
960 PRINT "RSD"
965 PRINT USING A5;Sd/Cu6
966 A5:IMAGE 1(2X,3D.6D)
970 INPUT "IS THERE NEED PRINT ON PRINTER ? 1/0",E25
980 IF E25=1 THEN GOTO 1000
990 GOTO 1020
1000 PRINTER IS PRT
1010 GOTO 630
1020 END

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