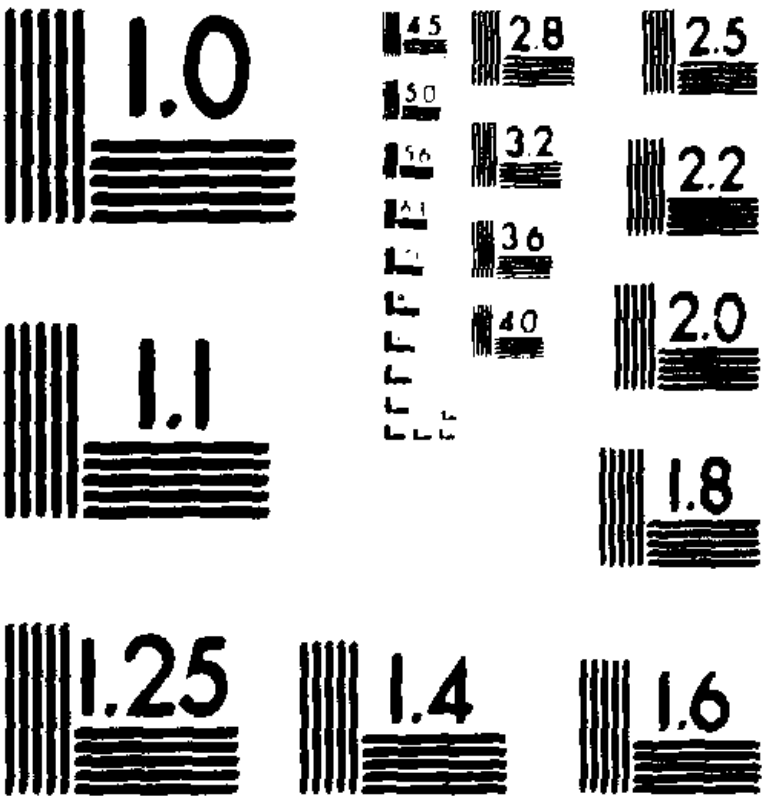

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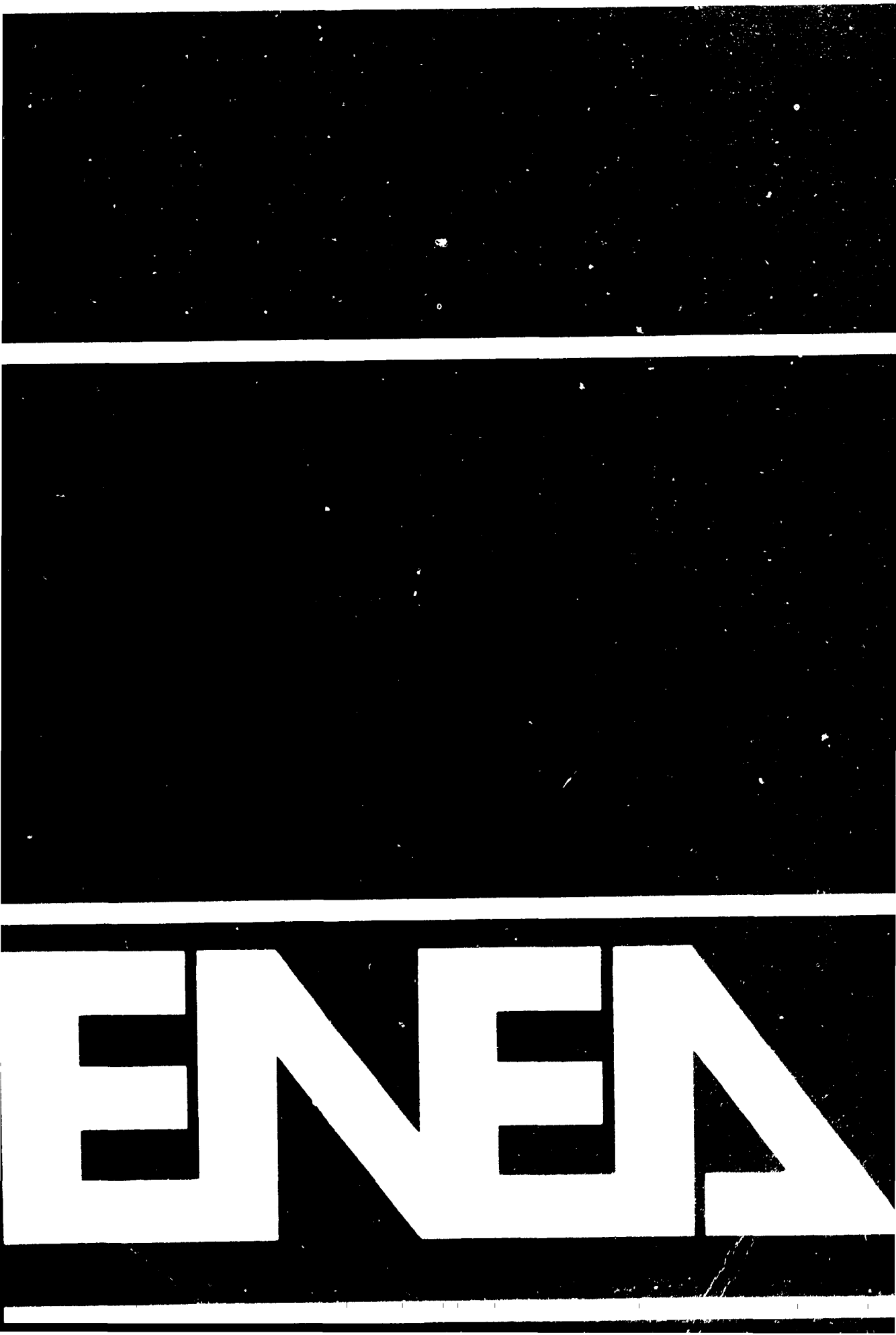
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COMITATO NAZIONALE PER LA RICERCA E PER LO SVILUPPO
DELL'ENERGIA NUCLEARE E DELLE ENERGIE ALTERNATIVE

ADD-A-SOURCE TECHNIQUE

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RIASSUNTO

Il conteggio dei neutroni emessi per fissione spontanea dagli isotopi del plutonio è un possibile metodo di determinazione del contenuto di plutonio in campioni di diversa forma e composizione.

Allo scopo di discriminare il conteggio dovuto ai neutroni di fissione spontanea da quello dovuto alla emissione di neutroni di tipo (α,n), sono state sviluppate tecniche di misura basate sulla correlazione temporale dei conteggi neutronici. Ulteriori procedure sono state inoltre sviluppate per correggere gli effetti dovuti al conteggio di neutroni provenienti da fissioni indotte. Tale correzione dipende da una accurata conoscenza della composizione chimica del campione.

Questo lavoro presenta una formulazione teorica di un nuovo approccio (add-a-source technique) che potrebbe permettere la correzione della misura indipendentemente dalla conoscenza della composizione chimica del campione.

SUMMARY

Counting neutrons emitted by spontaneously fissioning plutonium isotopes is a means for determining plutonium content in samples.

Correlation techniques have been developed for separating such neutrons from a background of nonfission neutrons due to (α, n) reactions. Further procedures are then used to correct the effect of neutron-induced fission. In order to perform such a correction the sample's chemical composition (including the presence of moisture) must be a well known parameter.

The present paper reports a theoretical formulation of a new approach (add-a-source technique), which could allow to correct the measurement indipendently by the knowledge of the sample's chemical composition.

INTRODUCTION

Neutron coincidence technique is currently used for the nondestructive assay of plutonium sample. Such a technique is based on the measurement of time-correlated neutrons emitted in the spontaneous fission of the even mass number plutonium isotopes. Shift Register electronic is normally used to count both the total neutron rate and the coincidence rate. This last parameter is related to the mass of ^{240}Pu-effective to give a calibration curve.

Such a calibration function is nonlinear for plutonium samples larger than a few tens of grams because of neutron-induced fission.

A correction procedure, multiplication equation, [1], based on the knowledge of the ratio of (α, n) neutrons to spontaneous-fission neutrons was developed. Such a ratio is currently calculated from the plutonium isotopic composition, ^{241}Am content, and (α,n) yields in oxides. This multiplication equation allows for the evaluation of the sample multiplication factor which is then used for determining the multiplication-corrected coincidence rate.

It must be pointed out that, in order to perform this calculation, the sample chemical composition must be a well known parameter. This requirement is due to the fact that in neutron coincidence counting there are more unknown parameters (the mass of ^{240}Pu-effective, the multiplication factor M and the aforementioned ratio) than knowns (R and T).

"^{252}Cf add-a-source" technique, proposed by H. Menlove [2], could be a promising approch to solve such a problem which is of great importance especially for medium-to-large samples that are impure or have a large moisture content. Such a technique should be able to estimate the multiplication in the sample by measuring the incremental multiplication induced by a ^{252}Cf source coupled to the sample can. This incremental multiplication is measured both on total and coincidence rate eventually obtaining a calibration curve. Such a relationship (incremental multiplication versus multiplication factor) can be calculated by means of pure PuO_2 sample with the M values obtained from the multiplication equation. The add-a-source curve can be then used to evaluate the M value for impure sample.

The purpose of this report is to determine a theoretical relationship between the incremental multiplication and M without using a calibration curve which is, in general, dependent on the geometry of the sample container.

CALIFORNIUM-252 ADD-A-SOURCE

"Add-a-source" technique is based on the measurement of the perturbation on total and coincidence rate, induced by a Californium source closely coupled to the Pu sample in assay.

The procedure foresees the passive measurement of the sample to obtain R_s and T_s, and then the repetition of the measurement with the ^{252}Cf source at the bottom of the can to obtain R_{Cf+s} and T_{Cf+s}.

The ^{252}Cf source is also measured with no plutonium in the can to obtain the external neutron flux in terms of total and coincidence rate (T_{Cf} and R_{Cf}).

As a first approximation we can say that the induced perturbation, which is expressed by the following two formula

$$(T_{Cf+s}-T_s - T_{Cf})/T_{Cf} = Y_T$$

$$(R_{Cf+s}-R_s - R_{Cf})/R_{Cf} = Y_R$$

is a function of the sample multiplication factor and the probability for Cf neutrons of interacting with the sample itself.

We want to express such dependence with a system of two equations (with the perturbations on total and coincidence rate as dependent variables), whose solution allows for the sample multiplication factor determination independently from the knowledge of the sample chemical composition.

DEFINITION OF THE PROBLEM

We define a system consisting of a Californium source and of a multiplication sample. Since our aim is to obtain a formulas for the perturbation in both total and coincidence rate due to Californium source, we assume that the multiplication sample does not emit any neutron for spontaneous fissions.

We need now to define some parameter useful for the solution of the problem. Let $q(\nu)$ be the probability that ν neutrons are emitted for induced fissions and $q_{Cf}(\nu)$ the same probability for Cf spontaneous fissions.

Let p be the probability that a neutron induces fission in the multiplication sample. Then (1-p) will be the probability that a neutron

does not cause fission but leaves the system. In an energy independent model, the detector efficiency defines the fraction of these neutrons which is counted.

We introduce a global parameter k which denotes the probability that a neutron, coming from a Cf spontaneous fission event, leaves the system without entering the sample, then (1-k) is the probability that a neutron enters the multiplication sample. We assume that the only possible interaction for a Cf neutron entering the sample is the induced fission.

Let now $R(\mu)$ be the probability that μ neutrons leave the system as a consequence of one source event. We want to express such a probability distribution and its factorial moments as a function of the neutron emission probability for Cf fissions, the probability of interaction with the multiplication sample, the probability of induced fissions in the sample and of the neutron emission probability of such fissions.

In order to accomplish such requirement we make use of the concept of Probability Generating Functions (PGF).

Any random process resulting in various integral numbers, each with its own probability, may be described by a PGF.

In general a PGF is defined as

$$f(u) = \Sigma q(n)u^n$$

where q(n) is the probability for the realization of the integer random variable n.

Among the properties of the PGF, the following will be used in our paper:

(P1): The PGF of a random process which is the combination of two or more statistically independent processes is obtained by multiplying together the constituent PGF's.

(P2) The derivates at u=1 of a PGF yield the factorial moments of p(n)

(P3) Let $g_i(u)$ be the PGF formed with the conditional probabilities p(n/i), which describes the probabilities that the variable takes the value n under the condition i. If the probability of occurrence of this condition is a P_i, then the PGF for the quantity n without conditions is:

$$f(u) = \Sigma P_i g_i(u)$$

THEORETICAL FORMULATION

First of all we need to define the PGF for the number of neutrons from a Cf source event ($f_{Cf}(u)$) and that one for the number of neutrons emitted by an induced fission.

$$f_{Cf}(u) = \Sigma q_{Cf}(i)u^i$$

$$f(u) = \Sigma q(i)u^i$$

We can now define, as a consequence of one Cf source neutron, the PGF for the number of neutrons which leave the system in the case of absence of multiplication sample:

$$L(u) = u$$

We can also define, assuming one neutron in the multiplication sample, the PGF for the number of neutrons of this first and of all successive generations that leave the multiplication sample:

$$h(u) = (1-p)u + p \cdot f[h(u)]$$

(defined by Bohnel [3])

We have thus, two different ways of evolution of the same initial event (one Cf source neutron) namely:

- neutrons leaving immediately the system
- neutrons leaving the system after interaction with the multiplication sample.

Since these two possible evolutions are not independent to each other, the PGF for the number of all the neutrons that leave the system (always starting from one Cf source neutron) can be expressed by the sum of the relative PGFs weighted by the probability that the evolution itself comes about:

$$N(u) = ku + (1-k)\{(1-p)u + pf[h(u)]\}$$

We now introduce a new PGF:

$$H(u) = \Sigma R(i)u^i$$

with the probabilities R(i) previously defined. This is the PGF for the number of neutrons leaving the system as the result of a Cf source event. It can be expressed making use of a PGF's property as:

$$H(u) = f_{Cf}[N(u)]$$

From this equation and from a PGF's property we can evaluate the probability distribution $R(\mu)$ and its factorial moments describing the multiplicity of the Cf source neutron "burst" in presence of a multiplication sample.

FACTORIAL MOMENTS EVALUATION

For a PGFs' property, the derivates at u=1 of a PGF yield the factorial moments of the probability distribution p(n):

$$df(u)/du \Big|_{u=1} = \bar{n}$$

$$d^2f(u)/du^2 \Big|_{u=1} = \overline{n(n-1)}$$

In our case we have:

$$\bar{\nu} = dH/du \Big|_{u=1} = \left(df_{Cf}/dN \cdot dN/du \right)_{u=1} \qquad (1)$$

$$\overline{\nu(\nu-1)} = d^2H/du^2 \Big|_{u=1} \qquad (2)$$

We know from [3] that

$$dh(u)/du \Big|_{u=1} = (1-p)/(1-p\bar{\nu}_I) \qquad (3)$$

and $$d^2h(u)/du^2 \Big|_{u=1} = \{ \overline{\nu_I(\nu_I-1)} \ (1-p)^2 p \} / (1-p\bar{\nu}_I)^3 \qquad (4)$$

By inserting Eq. (3) and (4) in the elaboration of Eq. (1) and (2) we have

$$\bar{\nu} = \bar{\nu}_{Cf}[(1-k)M+k] \quad (5)$$

$$\overline{\nu(\nu-1)} = k^2\,\overline{\nu_{Cf}(\nu_{Cf}-1)} + (1-k)^2 M^2\,\overline{\nu_{Cf}(\nu_{Cf}-1)} + \\ + (1-k)\,M^2(M-1)\,\bar{\nu}_{Cf}\left[\overline{\nu_I(\nu_I-1)}/(\bar{\nu}_I-1)\right] + 2k(1-k)M\,\overline{\nu_{Cf}(\nu_{Cf}-1)} \quad (6)$$

As we previously pointed out "add-a-source" technique is based on the measurement of the perturbation, induced by an external Cf source, in the coincidence and total rate of a multiplication sample.

Such a perturbation has been defined as followed:

$$(T_{Cf+s} - T_s - T_{Cf})/T_{Cf} = Y_T$$

$$(R_{Cf+s} - R_s - R_{Cf})/R_{Cf} = Y_R$$

We can also define the two variables Y_T and Y_R as:

$$Y_T = (\bar{\nu}_{Cf+s} - \bar{\nu}_s - \bar{\nu}_{Cf})/\bar{\nu}_{Cf}$$

$$Y_R = \left[\overline{\nu_{Cf+s}(\nu_{Cf+s}-1)} - \overline{\nu_s(\nu_s-1)} - \overline{\nu_{Cf}(\nu_{Cf}-1)}\right] / \overline{\nu_{Cf}(\nu_{Cf}-1)}$$

where the differences $[\bar{\nu}_{Cf+s} - \bar{\nu}_s]$ and $[\overline{\nu_{Cf+s}(\nu_{Cf+s}-1)} - \overline{\nu_s(\nu_s-1)}]$ are expressed by the Eq. (5) and (6) respectively.

Finally we can get the following equations:

$$Y_T = (1-k)(M-1)$$

$$Y_R = k^2 + (1-k)^2M^2 + (1-k)M^2(M-1)A + 2k(1-k)M - 1$$

where

$$A = \left[\overline{\nu_I(\nu_I-1)}\;\bar{\nu}_{Cf}\right] / \left[(\bar{\nu}_I-1)\;\overline{\nu_{Cf}(\nu_{Cf}-1)}\right]$$

The resolution of such a system allows for the determination of the two unknown values M and k. The term A is evaluated on the basis of the sample isotopic composition and Cf nuclear data.

MATHEMATICAL MODEL VALIDATION

A simple computer simulation program has been developed in order to test the mathematical model and evaluate the sensitivity of the technique.

For the simulation program, the samples were assumed to be composed of 100% Pu^{239}, hence the spontaneous emission of the even Pu isotopes was not simulated. The calculation produced, for different values of k and M, the factorial moments of the first and second order ν and $\nu(\nu-1)$ of the probability distribution relative to two different phenomena, namely:

- neutrons which leave a system constituted by a Cf source and a multiplication sample without spontaneous emissions;
- neutrons emitted by the Cf source.

The difference of these two moments can represent the perturbation induced in the multiplication sample by a Cf source.

We have then used these values to evaluate k and M by the system of two equations. Table I and fig. 1 summarize the results obtained.

We have also plotted in fig. 2 the behaviour of Y_R versus (M-1) for different k values, as evaluated by the theoretical formulation, along with the values obtained by the simulation program.

These results show that it is possible to determine the multiplication factor by the system of equations at least for values of M higher than 1.05.

Below such a limit the perturbation induced by the external Cf source is so small that the errors introduced by the simulation, in the calculation of Y_T and Y_R become not negligible.

It must be pointed out that the simulation did not take into account the errors due to the three different measurements which must be carried out for evaluating Y_T and Y_R .

In this context it is worth mentioning that Y_T and Y_R are evaluated with different precisions (better in the case of Y_T) and that the multiplication signal is enhanced by the coincidence rates (Y_R is in the range of 0-0.3 while Y_T is in the range 0-0.08).

ERROR PROPAGATION ANALYSIS

In order to study the errors propagation in determining the multiplication factor by the add-a-source approach, a computer simulation was used.

Such a simulation starts with the "true" values of Y_T and Y_R, obtained from the system of two equations with M and k known.

The measured values Y_T^* and Y_R^* are given by :

$$Y_T^* = Y_T(1+\varepsilon)$$
$$Y_R^* = Y_R(1+\eta)$$

where ε and η represent the relative errors caused by instrument imprecision.

Both errors are assumed to be independent and to be normally distributed with mean zero and variances σ_ε^2 and σ_η^2, respectively.

These measured values Y_T^* and Y_T^* are then used to recalculate the multiplication factor.

The whole calculation is repeated, with the same set of variances, in order to produce a distribution of the M values, which simulates the statistical density function of the actual measurements.

Fig. 3 shows the relative standard deviation of M versus the relative precision of Y_T and Y_R; fig. 4 shows the behaviour of the relative standard deviation of M as a function of the multiplication factor for various figures of relative precisions of Y_T and Y_R.

CONCLUSIONS

Theoretical formulation of "^{252}Cf add-a-source", using both induced perturbation on real and total rate for solving a system of two equations, has been confirmed by computer simulation.

Simulation results indicate the possibility to perform measurements of the multiplication coefficient at least for values of M higher than 1.05, for realistic values of (1-k) in the range 0.25÷0.5.

This approach could be very promising for measuring sample whose

chemical composition is not known for the presence of light impurities or large moisture quantities.

In fact, after determining the M value by the "add-a-source technique", the multiplication equation can be rewritten, to evaluate the (α,n) to spontaneous fission ratio, in the following form:

$$\alpha = \frac{M}{(R/T)/\rho_0 - 2.074\,(M^2-M)} - 1$$

where ρ_0 is the ratio (R/T) for a small nonmultiplying reference sample.

Both α and M are then used to calculate the multiplication-corrected coincidence ratio.

This add-a-source technique has the desirable feature that, although it requires an additional measurement with the Cf source, no further calibrations are needed for the multiplication factor determination.

The disadvantage of the method is that Y_T and Y_R must be determined with high precision so that counting times should be long.

The analysis of the statistical error propagation pointed out that relative precisions of 0.1% and 0.5% for Y_T and Y_R respectively are needed to obtain a relative precision on M value better than 1%.

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2 H.O. Menlove "Calibration and Error Reduction in Neutron Coincidence Counting", J. of Nuclear Material Management (October 1987), pp.20-28

3 K. Bohnel "The Effect of Multiplication on the Quantitative Determination of Spontaneously Fissioning Isotopes by Neutron Correlation Analysis", Nucl. Sci. Eng. 90 (May 1985), pp. 75-82

M_{True} \ $(1 - K)_{True}$	0.25	0.50	0.75	1.00
1.01	1.1202	1.0474	1.0489	1.0000
1.02	1.0776	1.0477	1.0349	1.0154
1.03	1.0168	1.0495	1.0364	1.0169
1.04	1.0487	1.0453	1.0439	1.0389
1.05	1.0508	1.0672	1.0552	1.0504
1.10	1.0992	1.1034	1.1025	1.0974
1.15	1.1518	1.1525	1.1498	1.1498
1.20	1.1979	1.2042	1.2031	1.1995

Table I - M values determined by the system of two equations using Y_T and Y_R obtained with the simulation program. (A = 1.1336)

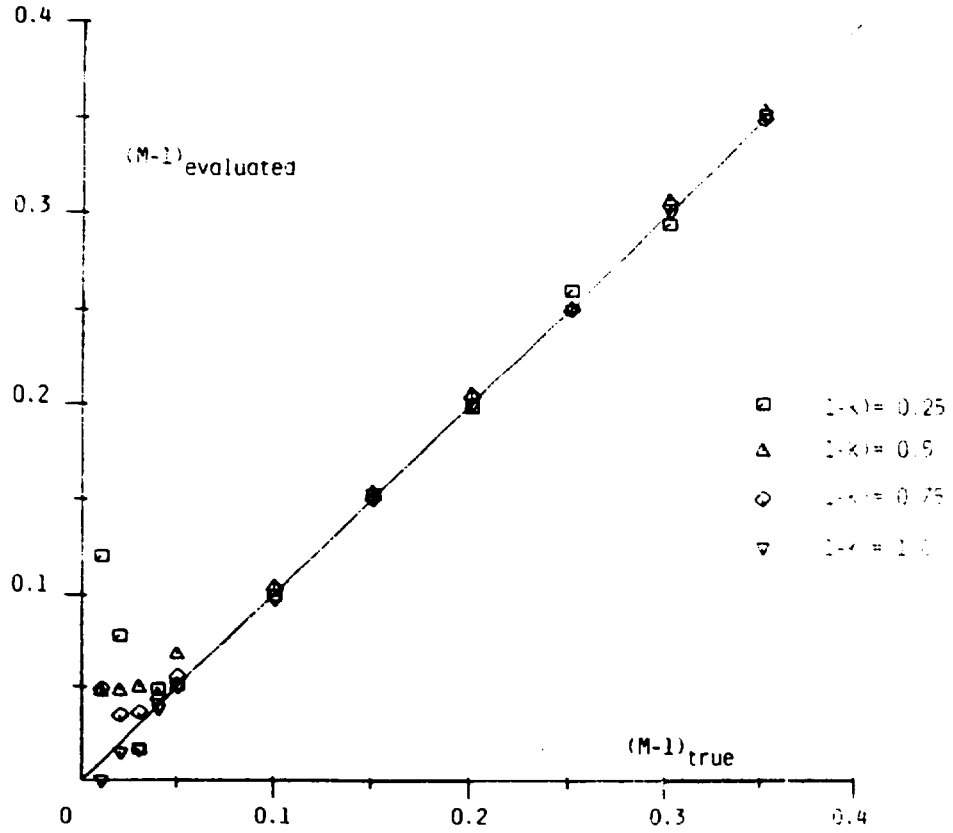

Fig.1 - Comparison between (M-1) calculated by the system of two equations and true (M-1) values.

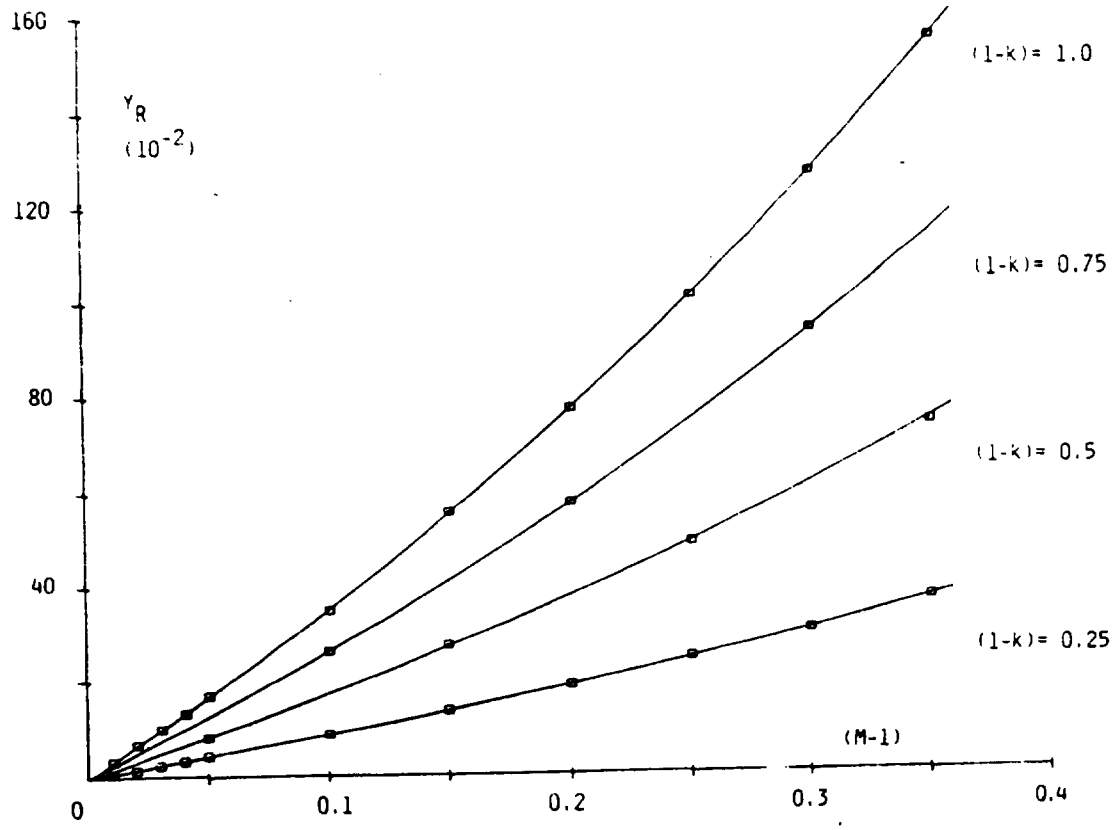

Fig.2 - Y_R as a function of (M - 1) for various (1-K) values.

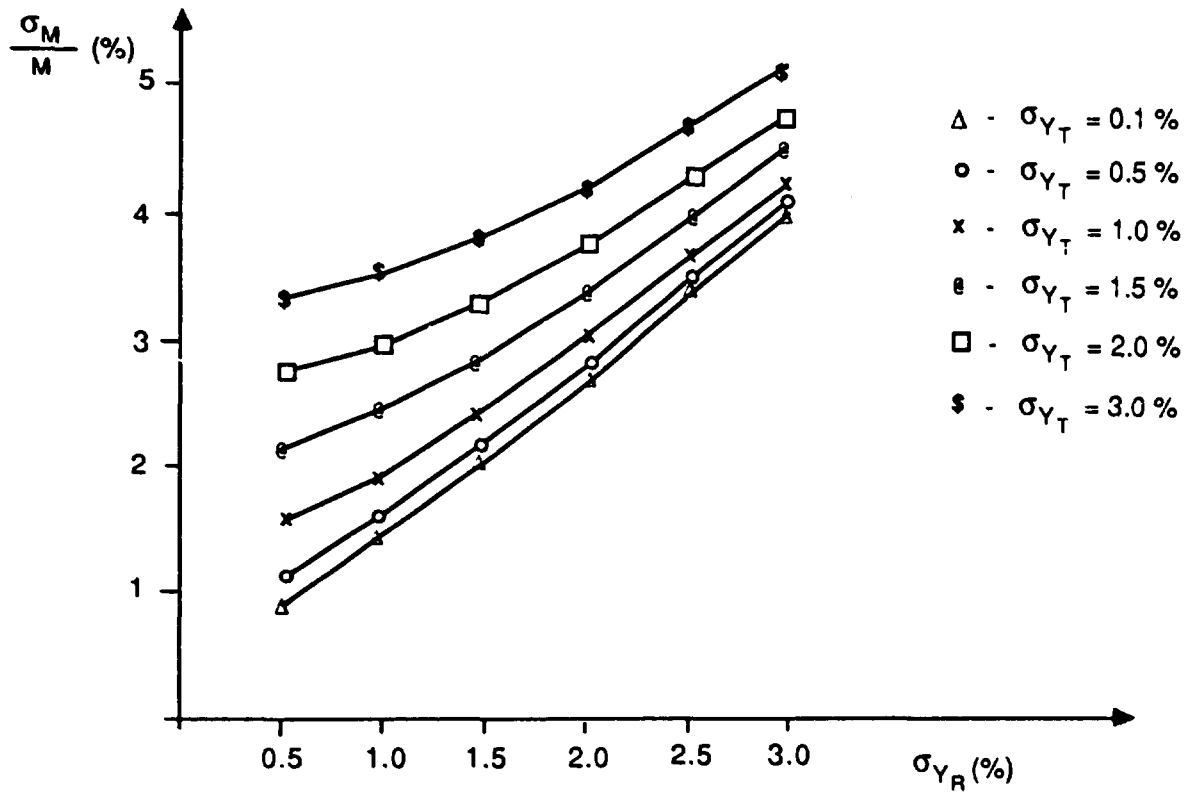

Fig. 3 - Relative standard deviation (%) of M as a function of relative precision of Y_T and Y_R.

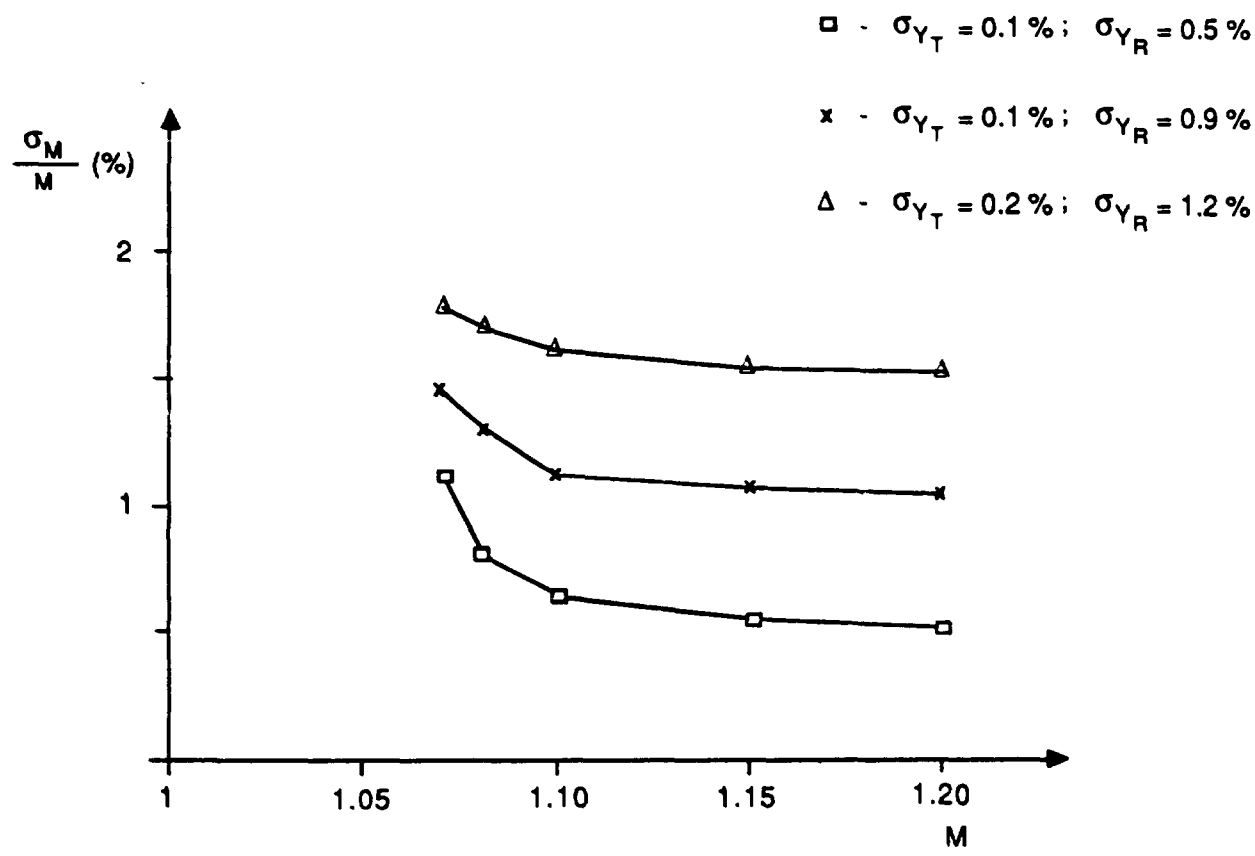

Fig. 4 - Relative standard deviation (%) of M as a function of the M value.

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