

OPTIMIZATION OF IRRADIATION, DECAY  
AND COUNTING TIMES IN NUCLEAR ACTI-  
VATION ANALYSIS USING SHORT-LIVED  
NUCLIDES

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REPORT 80-26

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## 1. INTRODUCTION

### 1.1 General comments

A standard problem in activation analysis is the choice of analytical conditions with respect to the experimental times : the irradiation time,  $t_i$ , the decay time,  $t_d$ , and the counting time,  $t_c$ . For a given analytical system every sample has a special optimal combination of these experimental times.

Till now this fact is payed little attention by the analysts. The main reason is, presumably, that to obtain a proper answer to the questions formulated for the majority of the analyses performed, there has been no real lack of activity. Besides, a formal treatment of this optimization problem is rather laborious. Therefore, since there would be no practical improvement of significance in the final results the problem of optimizing has been of minor interest. The choice of  $t_i$ ,  $t_d$  and  $t_c$  has mainly been governed by thumb-rules like : "irradiate one half-life and count two halflives" or "irradiate and count long enough to obtain 10000 net counts".

However, nowadays the analysts experience a strong wish to accurately characterize still lower concentrations of various elements. And in order to handle such low quantities accurately an optimization of the analytical procedure becomes increasingly more important. This makes necessary a real optimal choice of  $t_i$ ,  $t_d$  and  $t_c$ .

### 1.2 Optimization criteria

The "goodness" of an optimization criterium formulated is closely connected to its eventual relationship with parameters that either are important or might be of significant value in the analysis like for instance the precision, the total analysis time or the total analysis cost. Therefore, depending on the present requirements any of the four criteria shortly formulated below (and variations of these), are considered to be fully acceptable :

- a) lowest possible (minimum) relative standard deviation;
- b) minimum detection limit ( $L_D$ -value as defined by Currie [Cur 68]);
- c) lowest possible analysis total-time in order to achieve a relative standard deviation lower than a preselected value;
- d) lowest possible analysis total-cost to achieve a specified relative standard deviation value.

Criterion c) and d) might be regarded as variations of a), but express so frequently arising practical wishes that they deserve to be mentioned separately.

### 1.3 Former work, a critical discussion

The problem of optimizing the experimental times,  $t_i$ ,  $t_d$  and  $t_c$  is sparingly studied and is formally treated by only a few authors. Their approach to the problem will briefly be reviewed and discussed in chronological order in this section.

The reported optimization procedures all have in common a proposal of an algorithm that should be either maximized or minimized with the experimental times as running variables. Uncommon are the optimization criteria used, and even the expressions formulated using the same criterium may vary. The results obtained will of course depend strongly upon the chosen optimization expressions. And for this reason before utilizing a proposed procedure one should be fully aware of its basic algorithm.

The first published article concerning these problems appeared in the literature in 1961 [Oka 61], written by M. Okada. His intention is to describe a method for calculating an optimum decay time,  $t_d$ , with the goal of minimizing possible interfering activity. His approach to the problem is however not sufficiently adequate. The parameter  $t_i/t_d$  used does not enable one to choose the experimental conditions since  $t_i$  and  $t_d$  are independent values; moreover, the chosen conditions are not related to the half-lives of the isotope to be determined and the interfering ones.

Izenhour et al. [Ise 64, Ise 65] have made a comprehensive computer program to calculate the optimum irradiation and decay times for in principle any element in any mixture regardless of its complexity. The program utilizes a library tape where physical parameters like atomic weights, isotopic abundancies, half-lives,  $\gamma$ -ray energies,  $\gamma$ -ray intensities, reference  $\gamma$ -ray spectra and reaction cross sections are stored. As additional information the program needs input data like the elements to be considered with their possible nuclear reactions and approximate weights, the flux of the bombarding particle, the experimentally convenient limits of irradiation and decay time, and a minimum acceptable activity level.

The calculations are based upon maximizing the value of the counting rate ratio  $A$  as a function of irradiation time,  $t_i$ , and decay time,  $t_d$ , when

$$A = \frac{R_i}{\sum_{j=1}^n R_j} \quad (1)$$

where  $R_i$  = the count rate of the nuclide to be determined and the denominator represent the sum of the countrates of all the  $n$  nuclei contributing to the energy range considered.

Limits to the general application of this method are of two kinds : 1) The minimization expression is not properly chosen because it does not fulfil the requirements given in section 1.2. This objection is the most serious. 2) The utilization of the program demands access to a relatively large computer with external devices for the physical parameter library. Besides, the program does not offer the possibility of calculating also the optimum counting time,  $t_c$ , which might be of great importance in specific situations.

A somewhat different approach is made by Yu.A. Tsirlin et al. [Tsi 66]. They construct a so-called quality factor

$$D = \frac{R_1^2}{(R_{VB} + R_{GB})} \quad (2)$$

where  $R_1$  = the net countrate of the nuclide to be determined,  
 $R_{VB}$  = the countrate of the variable background,  
 $R_{GB}$  = the general background count rate.

By maximizing D general trends in the variation of optimal irradiation and cooling times are correctly derived. However, since objection No 1 for the preceeding method also is applicable to this approach, the procedure cannot be adapted for calculation of real optimum times of sufficiently acceptable validity.

Quittner and Montvai [Qui 67] use the relative standard deviation as optimization criterion, expressed by maximizing the inverse formula

$$\delta = \frac{1}{\sigma_{rel}} = \frac{S_1}{(S_1 + 2S_{VB} + 2S_{GB})^{1/2}} \quad (3)$$

where  $S_1$  = the net number of counts from the nuclide to be determined,  
 $S_{VB}$  = the net number of counts from the interfering nuclei,  
 $S_{GB}$  = the number of counts from the general background.  
 Simple formulas for calculating the optimum  $t_i$  and  $t_d$  are given as approximate solutions for the derivative of  $\delta$  with respect to  $t_i$  and  $t_d$ .

Examples of curves relating the optimum  $t_i$  and  $t_d$  to various easily determinable dimensionless parameters are shown. No serious objections can be raised against this treatment, but its main limitations is its nonability to calculate also the counting time  $t_c$ , and that the solutions given are partly approximate.

The most comprehensive study so far is performed by Fedoroff [Fed 71, Fed 73], who has treated the problem both in terms of optimum sensitivity and minimum standard deviation. Concerning the first of these two possible approaches, he has adopted the formalism given by Currie [Cur 68]. As minimization expression the formula for the minimum detectable mass is used :

$$M_{LD} = \frac{m_s}{S_s} \cdot [k^2 + 2k \cdot (S_{GB} + \sigma_{GB}^2 + \sum_i S_{VB_i} + \sum_i \sigma_{VB_i}^2)^{1/2}] \quad (4)$$

Here  $\frac{m_a}{S_a}$  (or  $\frac{S_a}{m_a}$ ) is an overall calibration factor relating the detector response to the mass present (and is determined by a standard sample where  $m_a$  is well known),  $k$  is a constant related to the preselected probability for concluding correctly that activity is present and the predetermined degree of confidence for detecting its presence. If the two probabilities are both equal to 0.95 (which is a common case), one has a confidence level of 95% in the detection, and this corresponds to  $k = 1.645$ .  $S_{GB}$  is the total number of counts originated from the general background, and  $\sigma_{GB}$  the corresponding standard deviation,  $S_{VB_i}$  is the number of counts due to the interfering nuclide  $i$ , and  $\sigma_{VB_i}$  the corresponding standard deviation.

In the standard deviation approach the minimization expression used is :

$$\sigma_{rel} = \frac{(S_1 + S_B + \sigma_B^2)^{\frac{1}{2}}}{S_1} \quad (5)$$

where  $S_1$  is the net number of counts of the nuclide to be determined, and  $\sigma_{rel}$  the corresponding relative standard deviation,  $S_B$  is the number of counts from all the background components including the general background, and  $\sigma_B$  the corresponding standard deviation.

For both these possible minimization expressions the optimal values of  $t_i$ ,  $t_d$  and  $t_c$  are calculated separately, and represented graphically as functions of dimensionless parameters, under the supposition of one variable background component in addition to the general background.

This last outlined approach is friendly to the used with respect to the efforts needed for a proper calculation of optimal values of  $t_i$ ,  $t_d$  and  $t_c$ . This can relatively easy be performed on a small table calculator. However, the procedures do not include calculation of optimal combinations of the various experimental times.

Zikovsky [Zik 74] proposed methods for calculating optimal values of  $t_i$ ,  $t_d$  and  $t_c$  by minimization of the two expressions

$$F_1 = \left( \sum_{j=2}^n N_j \right) - N_1 \quad (6)$$

and

$$F_2 = F_1 \cdot \left( \sum_{j=1}^n N_j / N_1 \right) \quad (7)$$

where  $N_j$  is the number of disintegrations for the interfering component  $j$  within the counting time  $t_c$ , and  $N_1$  the corresponding counting number of the nuclide to be determined.

This approach is based on somewhat artificially constructed expressions, and the optimum times calculated has no direct connection to parameters that are important or might be of significant value in the analysis as pointed out in section 1.2. The general use of this method is therefore seriously limited.

Watterson [Wat 75] treats especially the problem of determination of the optimum values for  $t_i$  and  $t_d$  when selecting a specific photopeak in a complex spectrum. Under the supposition that the background can be represented by a straight line he uses the following minimization expression :

$$\frac{\sigma_1}{S_1} = \frac{(S_1 + \frac{k}{2} \cdot \sum_{j=1}^n B_j)^{\frac{1}{2}}}{S_1} \quad (8)$$

where  $\sigma_1/S_1$  is the relative standard deviation of the nuclide to be determined with  $S_1$  net number of counts,  $k$  equals the number of channels in the photopeak +2 and  $B_j$  is the number of counts from the background component  $j$ .

The discussion is restricted to counting times that are short in comparison with the half-life. This means in practice that the calculations are restricted to relatively longlived nuclides. No general and directly applicable results are given, neither in tables nor curves. Altogether, this treatment is limited, both theoretically and with respect to the applicability, to special cases only.



The most recently published approach to this optimization problem is worked out by Bourmistenko et al. [Bou 77]. A computer program is constructed to automatically perform the following operations : 1) determination of a list of nuclides and their  $\gamma$ -ray energies formed during irradiation of a sample (with well known composition) by bransstrahlung, 2) optimization of the experimental times and the maximum energy of the bremsstrahlung and 3) choice of the "best"  $\gamma$ -line for analysis of an element.

To execute these operations a rather comprehensive catalogus containing various physical constants for the different nuclides directly accessible by the computer program is necessary.

Minimization of the relative standard deviation represented by the formula :

$$\sigma_{rel} = \frac{(S_1 + 2 \cdot \lambda \cdot (S_{\phi_1} + S_{\phi_2}))^{\frac{1}{2}}}{S_1} + m \cdot \frac{N_{\phi_2}}{S_1} \quad (9)$$

as the best optimalization criterium is chosen.

Here  $\lambda$  = coefficient depending on the spectrum computer-treatment method,

$m$  = coefficient depending on the multiplet treatment method,

$S_{\phi_1}, S_{\phi_2}$  = number of counts from the  $i$ -th and  $j$ -th lines determined with the conditions  $|E - E_i| > k \cdot \sigma(E)$  and  $|E - E_j| < k \cdot \sigma(E)$ , respectively. Here  $E$ ,  $E_i$  and  $E_j$  are the energies of the  $\gamma$ -line under study and the interfering ones,  $\sigma(E)$  a parameter of the Gaussian distribution, and  $k$  a coefficient depending on the evaluation of the  $\gamma$ -spectrum.

It is obvious that in order to make full use of this procedure, the spectrum statistics must be good enough to give a rather smooth spectrum structure. It is not suitable in cases where summation over a relatively wide energy range gives only a few hundreds of counts and even lower. Besides, the procedure is specially designed for solving optimization problems in bremsstrahlung activation. This will also limit the general use.

Neither curves nor tables are given showing general trends in the variation experimental times as functions of suitable parameters.

In summary, it will be seen from this review, none of the approaches discussed can be generally applied. Especially when dealing with analysis utilizing short-lived nuclei and low concentrations there is a need for a more thorough treatment taking into consideration both the ideal wishes for such analysis (possibility of being able to choose all the varying parameters freely) and the somewhat more unavoidable limitations that might arise in the practical world, for instance an imposed least possible delay-time and other practical limitations concerning the experimental equipment).

The following discussion is an attempt to meet these requirements.

## 2. THEORETICAL BASIS

### 2.1 Principal criterion

The relative standard deviation of an analysis is chosen as the best criterion for optimization of  $t_i$ ,  $t_d$  and  $t_c$ . Usually when necessary precautions are taken, the precision in the analytical result from an NAA-analysis is governed by the uncertainty in the counting number, normally expressed by the standard deviation. By  $\dot{\gamma}$ -counting in an energy window selected by an upper and a lower discriminator level, the counting number obtained,  $S_T$ , has three main sources: the nuclide to be determined,  $S_1$ , the general background,  $S_{GB}$ , and the variable background,  $S_{VB}$ .  $S_{VB}$  includes the radioactivity from decaying nuclides produced from matrix or trace elements during irradiation, and the activity from possible radioactive daughter nuclides. This is expressed as:

$$S_T = S_1 + S_{GB} + S_{VB} \quad (10)$$

Assuming that radioactive decay is ruled by Poisson statistics the standard deviation in a counting number  $S$  is:

$$\sigma = \sqrt{S} \quad (11)$$

The relative standard deviation in  $S_1$  is then :

$$\begin{aligned} \frac{\sigma_1}{S_1} &= \frac{[S_T + S_{GB} + S_{VB}]^{\frac{1}{2}}}{S_1} \\ &= \frac{[S_1 + 2S_{GB} + 2S_{VB}]^{\frac{1}{2}}}{S_1} \end{aligned} \quad (12)$$

Eq. (12) shows that  $\sigma_1/S_1$  may be lowered by decreasing the contribution from the background. This may be accomplished by a proper choice of the experimental times. One therefore needs explicit expressions for  $S_1$ ,  $S_{GB}$  and  $S_{VB}$  as functions of the variable parameters.

## 2.2 Derivation of some mathematical expressions for single cycle activation/counting.

### 2.2.1 Mother activity

The desintegration rate of a primary formed nuclide at the end of irradiation ( $t_d = 0$ ) is described by the well known formula :

$$D(0) = \sigma \cdot \phi \cdot N' \cdot (1 - e^{-\lambda t_i}) \quad (13)$$

where  $\sigma$  = reaction cross section (in barn),

$\phi$  = neutron flux ( $n \cdot cm^{-2}s^{-1}$ ),

$N'$  = number of target nuclei,

$\lambda$  = desintegration constant,  $\ln 2/t_{1/2} (s^{-1})$ ,

$t_i$  = irradiation time (s).

The desintegration rate for a  $\beta$ -radioactive nuclide at any time  $t_d$  after the end of irradiation is given by :

$$D(t_d) = D(0) \cdot e^{-\lambda t_d}$$

Since

$$0 = - \frac{dN}{dt} = \lambda \cdot N$$

where  $N$  is the number of radioactive nuclei at any time  $t$ , we have similarly that

$$N(t_d) = N(0) \cdot e^{-\lambda t_d} \quad (16)$$

Let the nuclide be counted in the time period  $t_d$  to  $t_d + t_c$ , and let the total counting efficiency be  $\epsilon$ . The number of recorded counts is then expressed by :

$$\begin{aligned} S &= \epsilon \cdot (N(0) \cdot e^{-\lambda t_d} - N(0) \cdot e^{-\lambda(t_d+t_c)}) \\ &= \epsilon \cdot N(0) \cdot e^{-\lambda t_d} \cdot (1 - e^{-\lambda t_c}) \end{aligned} \quad (17)$$

Combination of eqs. (13), (15) and (17) gives

$$\begin{aligned} S &= \epsilon \cdot K \cdot (1 - e^{-\lambda t_i}) \cdot e^{-\lambda t_d} \cdot (1 - e^{-\lambda t_c}) \\ K &= \frac{1}{\lambda} \cdot \sigma \cdot \phi \cdot N' \end{aligned} \quad (18)$$

### 2.2.2 Daughter activity

Let the index combination  $ij$  mean the decay-chain number  $i$  of the primary formed nuclide  $j$ . The desintegration rate of a radioactive daughter nuclide is then

$$\frac{dN_{21}(t)}{dt} = \lambda_{11} \cdot N_{11}(0) \cdot e^{-\lambda_{11}t} - \lambda_{21} \cdot N_{21}(t) \quad (19)$$

$$(\text{when } N_{21}(0) = 0)$$

The number of radioactive daughter nuclei at any time  $t_d$  is then found by integration of eq. (19)

$$N_{21}(t_d) = \frac{\lambda_{11} \cdot N_{11}(0)}{\lambda_{21} - \lambda_{11}} \cdot (e^{-\lambda_{11}t_d} - e^{-\lambda_{21}t_d})$$

Let the absolute counting efficiencies for the mother and daughter nuclides be  $\epsilon_{11}$  and  $\epsilon_{21}$  respectively. The totally recorded number of counts from the daughter nuclide in a counting period  $t_d$  to  $t_d + t_c$  is then proportional to the number of produced daughter-daughter nuclides,  $N_{31}$  :

$$\begin{aligned}
 S_{21} &= \epsilon_{21} \cdot [N_{31}(t_d+t_c) - N_{31}(t_d)] & (21) \\
 &= \epsilon_{21} \cdot [N_{11}(0) - (N_{11}(t_d+t_c) + N_{21}(t_d+t_c)) - K_{11}(0) + \\
 &\quad + (N_{11}(t_d) + N_{21}(t_d))] \\
 &= \epsilon_{21} \cdot [(N_{11}(t_d) + N_{21}(t_d)) - (N_{11}(t_d+t_c) + N_{21}(t_d+t_c))]
 \end{aligned}$$

Introduction of the expressions for  $N_{11}$  and  $N_{21}$  in eq. (21) and rearrangements gives :

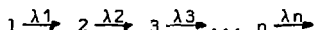
$$\begin{aligned}
 S_{21} &= \epsilon_{21} \cdot K_{11}(0) \cdot [e^{-\lambda_{11}t_d} \cdot (1 - e^{-\lambda_{11}t_c}) \cdot (1 + \frac{\lambda_{11}}{\lambda_{21} - \lambda_{11}}) - \\
 &\quad - \frac{\lambda_{11}}{\lambda_{21} - \lambda_{11}} \cdot e^{-\lambda_{21}t_d} \cdot (1 - e^{-\lambda_{21}t_c})] & (22)
 \end{aligned}$$

Combination of eqs. (13), (15) and (22) gives :

$$\begin{aligned}
 S_{21} &= \epsilon_{21} \cdot K_{11} \cdot (1 - e^{-\lambda_{11}t_i}) \cdot [e^{-\lambda_{11}t_d} \cdot (1 - e^{-\lambda_{11}t_c}) \cdot \\
 &\quad \cdot (1 + \frac{\lambda_{11}}{\lambda_{21} - \lambda_{11}}) - \frac{\lambda_{11}}{\lambda_{21} - \lambda_{11}} \cdot e^{-\lambda_{21}t_c}] & (23)
 \end{aligned}$$

### 2.3.3. The case of n successively decaying nuclei

In NAA the primary formed activation products give rise to none or a single radioactive daughter product as the most common case, and these situations are treated explicitly in section 2.2.1 and 2.2.2. However, there are examples where the decay chain contains several members, and it might be of interest to consider a series of n successive decays :



The desintegration rate of the  $i$ -th member is given by an equation similar to eq. (19). However, the substitution of the explicit expression of  $i$ -th member into the expression for the  $(i+1)$ -th member is rather cumbersome. Assuming that at  $t_d = 0$  the parent fraction alone is present, an expression for the number of nuclides of the  $n$ -th member at any time  $t_d$  is given by Bateman [Bat 10] :

$$N_{n1}(t_d) = \lambda_{11} \cdot \lambda_{21} \cdot \lambda_{31} \cdots \lambda_{(n-1)1} \cdot N_{11}(0) \cdot \sum_{i=1}^n F_{i1} \cdot e^{-\lambda_{i1} t_d} \quad (24)$$

where

$$F_{i1} = \frac{\lambda_{i1}}{\lambda_{i1} - \lambda_{(i-1)1}} \cdot \frac{1}{\lambda_{i1}}, \quad (i = p)$$

The number of nuclei of the  $(n+1)$ -th member produced up to time  $t$  equals the total number of the  $n$ -th member decayed from time  $t_d = 0$  to  $t_d = t_d$  :

$$N_{(n+1)1}(t_d) = N_{n1}(0) - \sum_{i=1}^n N_{i1}(t_d) \quad (25)$$

The total number of counts of the  $n$ -th member in the time period  $t_d$  to  $t_d + t_c$  is then

$$S_{n1} = \epsilon_{n1} \cdot [N_{(n+1)1}(t_d + t_c) - N_{(n+1)1}(t_d)] \quad (26)$$

Substitution of eqs. (25) and (24) into eq. (26), and rearrangements, gives :

$$S_{n1} = \epsilon_{n1} \cdot N_{11}(0) \cdot \sum_{i=1}^n \lambda_{i1} \cdot \lambda_{21} \cdot \lambda_{31} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^n F_{i1} \cdot e^{-\lambda_{i1} t_d} \cdot (1 - e^{-\lambda_{i1} t_c}) \quad (27)$$

Combination of eqs. (13) and (15) and substitution into eq. (27) gives :

$$S_{nj} = \epsilon_{nj} \cdot K_{1j} \cdot (1 - e^{-\lambda_{1j} t_i}) \cdot \prod_{i=1}^n \lambda_{1j} \cdot \lambda_{2j} \cdot \lambda_{3j} \cdots \lambda_{(i-1)j}$$

$$\sum_{i=1}^n F_{ij} \cdot e^{-\lambda_{1j} t_d} \cdot (1 - e^{-\lambda_{1j} t_c}) \quad (28)$$

#### 2.2.4 Explicit minimization formula

When the counting time,  $t_c$ , is not negligible compared to the half-life of the nuclide to be determined, and the counting efficiency is neither unity nor very small, the standard deviation (for no background present) is [Fri 64] :

$$\sigma = [S \cdot (1 - \epsilon + \epsilon \cdot e^{-\lambda t_c})]^{1/2} \quad (29)$$

For the variable background components  $\epsilon$  can be assumed to be very low, and for these nuclides the standard deviation is represented by eq. (11). Let the constant background count rate be  $R_{GB}$  (in counts per  $t_d$  for the nuclide to be determined). Combination of eqs. (12), (18), (28) and (29) gives the relative standard deviation for the case where the variable background is composed only of the daughters of the nuclide to be determined, i.e.  $j = 1$  :

$$\sigma_{11}/S_{11} = [\epsilon_{11} \cdot K_{11} \cdot (1 - e^{-\lambda_{11} t_i}) \cdot e^{-\lambda_{11} t_d} \cdot (1 - e^{-\lambda_{11} t_c})]^{-1} \cdot (30)$$

$$\cdot [\epsilon_{11} \cdot K_{11} \cdot (1 - e^{-\lambda_{11} t_i}) \cdot e^{-\lambda_{11} t_d} \cdot (1 - e^{-\lambda_{11} t_c}) \cdot (1 - \epsilon_{11} + \epsilon_{11} \cdot e^{-\lambda_{11} t_c}) + 2 \cdot K_{11} \cdot (1 - e^{-\lambda_{11} t_i}) \cdot \sum_{i=2}^{n1} \epsilon_{i1} \cdot \prod_{i=1}^{n1} \lambda_{11} \cdot \lambda_{21} \cdot \lambda_{31} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^{n1} F_{i1} \cdot e^{-\lambda_{11} t_d} \cdot (1 - e^{-\lambda_{11} t_c}) + 2 \cdot R_{GB} \cdot t_c]^{1/2}$$

For simplicity the following abbreviations are introduced :

$$(1 - e^{-\lambda_{ij}t_i}) = P_{iij} \cdot$$

$$e^{-\lambda_{ij}t_d} = P_{dij} \cdot$$

$$(1 - e^{-\lambda_{ij}t_c}) = P_{cij} \cdot$$

$$(1 - \epsilon_{ij} + \epsilon_{ij} e^{-\lambda_{ij}t_c}) = P_{\epsilon ij}$$

Then eq. (30) might be written as :

$$\begin{aligned} \sigma_{11}/S_{11} = & [\epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot P_{c11}]^{-1} \cdot [\epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot P_{c11} \cdot P_{\epsilon 11} \cdot \\ & + 2 \cdot K_{11} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{11} \cdot \lambda_{21} \cdot \lambda_{31} \cdots \lambda_{(i-1)1} \cdot \\ & \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{d11} \cdot P_{c11} + 2 \cdot R_{GB} \cdot t_c]^{1/2} \quad (30a) \end{aligned}$$

If the variable background is composed of activity from n primary formed nuclei only,  $\sigma_{11}/S_{11}$  is given by :

$$\begin{aligned} \sigma_{11}/S_{11} = & [\epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot P_{c11}]^{-1} \cdot [\epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot \\ & \cdot P_{c11} \cdot P_{\epsilon 11} + 2 \cdot \sum_{j=2}^n \epsilon_{1j} \cdot P_{i1j} \cdot P_{d1j} \cdot P_{c1j} + 2 \cdot R_{GB} \cdot t_c]^{1/2} \quad (31) \end{aligned}$$

Assuming that each of the n primary formed nuclei gives rise to a decay chain composed of  $n_1, n_2 \dots n_n$  members respectively, a further generalization is achieved. The explicit expression for  $\sigma_{11}/S_{11}$  will then be :

$$\begin{aligned} \sigma_{11}/S_{11} = & [\epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot P_{c11}]^{-1} \cdot [\epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot \\ & \cdot P_{c11} \cdot P_{\epsilon 11} + 2 \cdot K_{11} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{11} \cdot \lambda_{21} \cdot \\ & \cdot \lambda_{31} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{d11} \cdot P_{c11} + 2 \cdot \sum_{j=2}^n K_{1j} \cdot P_{i1j} \cdot \\ & \cdot \sum_{i=1}^{n_j} \epsilon_{ij} \cdot \sum \lambda_{ij} \cdot \lambda_{2j} \cdot \lambda_{3j} \cdots \lambda_{(i-1)j} \cdot \sum_{i=1}^{n_j} F_{ij} \cdot P_{dij} \cdot P_{cij} + 2 \cdot R_{GB} \cdot t_c]^{1/2} \quad (32) \end{aligned}$$



### 2.3 The relative standard deviation for an arbitrary number of accumulation cycles

If the specific count rate of the nuclide to be determined is low after one irradiation, one might have to repeat the irradiation/counting sequence (accumulation cycle) several times to obtain sufficiently good statistics. This section shows the evaluation of a general expression for the relative standard deviation of a number of net counts when an arbitrary number of background components and accumulation cycles are considered.

The duration of the accumulation cycles is assumed constant with constant  $t_i$ ,  $t_d$  and  $t_c$ , and denoted by  $r$ . Then  $r = t_i + t_d + t_c + t_x$ , where  $t_x$  is an extra "waiting" or "handling" time. Assume that the contribution from a nuclide (ij) will be zero after  $10 \cdot (t_i)_{ij}$ , and let

$$\frac{10 \cdot (t_i)_{ij}}{r} = m'_{ij}.$$

In addition, let  $m_{ij}$  be the natural number lying closest to  $m'_{ij}$  so that  $m_{ij} > m'_{ij}$ .

Let us first consider the simple case of one primary formed variable background component. The total number of counts in the first, second, third ... cycle is denoted  $S_{11,12}^{(1)}$ ,  $S_{11,12}^{(2)}$ ,  $S_{11,12}^{(3)}$  ... respectively. Then :

$$S_{11,12}^{(1)} = \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{d11} \cdot P_{c11} + \epsilon_{12} \cdot K_{12} \cdot P_{i12} \cdot P_{d12} \cdot P_{c12} + R_{GB} \cdot t_c$$

When the counting after the second irradiation starts there might still be activity left from the first irradiation, depending upon the  $m_{11}$  and  $m_{12}$  values. The number of counts from the second cycle is therefore :

$$S_{11,12}^{(2)} = \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{c11} \cdot (P_{d11} + e^{-\lambda_{11}(t_d+r)}) + \epsilon_{12} \cdot K_{12} \cdot P_{i12} \cdot (P_{d12} + e^{-\lambda_{12}(t_d+r)}) + R_{GB} \cdot t_c$$

By introducing the following abbreviation

$$e^{-\lambda_{ij}(t_d + x \cdot r)} = P_{dij}^{(x)}$$

the number of counts from the third cycle will be :

$$S_{11,12}^{(3)} = \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{c11} \cdot (P_{d11}^{(0)} + P_{d11}^{(1)} + P_{d11}^{(2)}) + \epsilon_{12} \cdot K_{12} \cdot P_{i12} \cdot P_{c12} \cdot (P_{d12}^{(0)} + P_{d12}^{(1)} + P_{d12}^{(2)}) + R_{GB} \cdot t_c$$

After  $m_{11}$  accumulation cycles :

$$S_{11}^{(m_{11})} = S_{11}^{(m_{11}+1)} = \dots = S_{11}^{(q)}$$

if  $q$  is the total number of accumulation cycles. Similarly after  $m_{12}$  cycles :

$$S_{12}^{(m_{12})} = S_{12}^{(m_{12}+1)} = \dots = S_{12}^{(q)}$$

Therefore, the number of counts accumulated after the  $q$  cycle is :

$$\begin{aligned} S_{11,12}^{(q)} &= \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{c11} \cdot (P_{d11}^{(0)} + P_{d11}^{(1)} + \dots + P_{d11}^{(m_{11}-1)}) + \\ &+ \epsilon_{12} \cdot K_{12} \cdot P_{i12} \cdot P_{c12} \cdot (P_{d12}^{(0)} + P_{d12}^{(1)} + \dots + P_{d12}^{(m_{12}-1)}) + R_{GB} \cdot t_c \\ &= \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{c11} \cdot \sum_{x=0}^{m_{11}-1} P_{d11}^{(x)} + \epsilon_{12} \cdot K_{12} \cdot P_{i12} \cdot P_{c12} \cdot \sum_{x=0}^{m_{12}-1} P_{d12}^{(x)} + \\ &+ R_{GB} \cdot t_c, \quad x \leq q \end{aligned}$$

The total number of counts accumulated after  $q$  cycles is:

$$\begin{aligned} S_{11,12}^{(\Sigma q)} &= S_{11,12}^{(1)} + S_{11,12}^{(2)} + \dots + S_{11,12}^{(q)} \\ &= \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{c11} \cdot \sum_{x=0}^{m_{11}-1} (q-x) \cdot P_{d11}^{(x)} + \epsilon_{12} \cdot K_{12} \cdot P_{i12} \cdot P_{c12} \cdot \sum_{x=0}^{m_{12}-1} (q-x) \cdot P_{d12}^{(x)} + \\ &+ q \cdot R_{GB} \cdot t_c, \quad x \leq q \end{aligned} \quad (33)$$

In the case of  $n$  primary formed nuclides the total number of counts after  $q$  accumulation cycles will be:

$$S_{\Sigma 1j}^{(\Sigma q)} = \sum_{j=1}^n \epsilon_{1j} \cdot K_{1j} \cdot P_{i1j} \cdot P_{c1j} \cdot \sum_{x=0}^{m_{i1j}-1} (q-x) \cdot P_{d1j}^{(x)} + q \cdot R_{GB} \cdot t_c, \quad x \leq q \quad (34)$$

Consider now the special case of no primary formed variable background component, i.e. the variable background is composed of decay products from the nuclide to be determined. The number of counts after the successive accumulation cycles will then be:

$$S_{\Sigma i1}^{(1)} = \epsilon_{i1} \cdot K_{i1} \cdot P_{i11} \cdot P_{c11} \cdot P_{d11}^{(0)} + K_{i1} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{i1} \cdot \lambda_{21} \cdot \dots \cdot \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{c11} \cdot P_{d11}^{(0)} + R_{GB} \cdot t_c$$

$$S_{\Sigma i1}^{(2)} = \epsilon_{i1} \cdot K_{i1} \cdot P_{i11} \cdot P_{c11} \cdot (P_{d11}^{(0)} + P_{d11}^{(1)}) + K_{i1} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{i1} \cdot \lambda_{21} \cdot \dots \cdot \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{c11} \cdot (P_{d11}^{(0)} + P_{d11}^{(1)}) + R_{GB} \cdot t_c$$

After a time corresponding to  $m_{i1}$  accumulation cycles:

$$S_{i1}^{(m_{i1})} = S_{i1}^{(m_{i1}+1)} = \dots = S_{i1}^{(q)}$$

Therefore:

$$S_{\Sigma i1}^{(q)} = \epsilon_{i1} \cdot K_{i1} \cdot P_{i11} \cdot P_{c11} \cdot \sum_{x=0}^{m_{i1}-1} P_{d11}^{(x)} + K_{i1} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{i1} \cdot \lambda_{21} \cdot \dots \cdot \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{c11} \cdot \sum_{x=0}^{m_{i1}-1} P_{d11}^{(x)} + R_{GB} \cdot t_c$$

The total number of counts accumulated after  $q$  cycles is:

$$S_{\Sigma i1}^{(\Sigma q)} = S_{\Sigma i1}^{(1)} + S_{\Sigma i1}^{(2)} + \dots + S_{\Sigma i1}^{(q)}$$

which explicitly gives:

$$S_{\Sigma 11}^{(\Sigma q)} = \epsilon_{11} \cdot K_{11} \cdot P_{i11} \cdot P_{c11} \cdot \sum_{x=0}^{m_{11}-1} (q-x) \cdot P_{d11}^{(x)} + K_{11} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{i1} \cdot \lambda_{21} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{c11} \cdot \sum_{x=0}^{m_{i1}-1} (q-x) \cdot P_{d11}^{(x)} + q \cdot R_{GB} \cdot t_c \quad (35)$$

A more general expression is obtained if one assumes a total of  $n$  primary formed nuclides with decay chains of  $n_1, n_2, \dots, n_n$  numbers respectively. The total accumulated number of counts is then:

$$S_{\Sigma ij}^{(\Sigma q)} = S_{\Sigma i1}^{(\Sigma q)} + \sum_{j=2}^n K_{1j} \cdot P_{i1j} \cdot \sum_{i=1}^{n_j} \epsilon_{ij} \cdot \sum_{i=1}^{n_j} \lambda_{i1} \cdot \lambda_{21} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_j} F_{ij} \cdot P_{c1j} \cdot \sum_{x=0}^{m_{ij}-1} (q-x) \cdot P_{dij}^{(x)} \quad (36)$$

The explicit expression for the relative standard deviation based on this last described situation will then be:

$$\sigma_{11}/S_{11}^{(\Sigma q)} = \left[ K_{11} \cdot \epsilon_{11} \cdot P_{i11} \cdot P_{c11} \cdot \sum_{x=0}^{m_{11}-1} (q-x) \cdot P_{d11}^{(x)} \right]^{-1} \cdot \left[ K_{11} \cdot \epsilon_{11} \cdot P_{i11} \cdot P_{c11} \cdot P_{e11} \cdot \sum_{x=0}^{m_{11}-1} (q-x) \cdot P_{d11}^{(x)} + 2 \cdot K_{11} \cdot P_{i11} \cdot \sum_{i=2}^{n_1} \epsilon_{i1} \cdot \sum_{i=1}^{n_1} \lambda_{i1} \cdot \lambda_{21} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_1} F_{i1} \cdot P_{c11} \cdot \sum_{x=0}^{m_{i1}-1} (q-x) \cdot P_{d11}^{(x)} + 2 \cdot \sum_{j=2}^n K_{1j} \cdot P_{i1j} \cdot \sum_{i=1}^{n_j} \epsilon_{ij} \cdot \sum_{i=1}^{n_j} \lambda_{i1} \cdot \lambda_{21} \cdots \lambda_{(i-1)1} \cdot \sum_{i=1}^{n_j} F_{ij} \cdot P_{c1j} \cdot \sum_{x=0}^{m_{ij}-1} (q-x) \cdot P_{dij}^{(x)} + 2 \cdot q \cdot R_{GB} \cdot t_c \right]^{1/2}, \quad x \leq q \quad (37)$$

### 3. RESULTS AND DISCUSSION.

#### 3.1. Calculations.

The function of  $\sigma/S$  (given by eqn.s (32) and (37) has no maximum and only one minimum. When varying the parameters  $t_i$ ,  $t_d$  and  $t_c$  this minimum value can be reached. Likewise, when fixing the value of one or two of the parameters, variation of the third one will give a curve of one minimum. If for instance  $t_i$  and  $t_d$  are fixed, the optimum counting time will be the  $t_c$ -value that satisfy the eqn.:

$$\frac{\partial(\sigma/S)_{t_i, t_d}}{\partial t_c} = 0$$

However, an exact solution of  $t_i$ ,  $t_d$  or  $t_c$  from the respective partial derivatives of eqn.s (32) or (37) is hard to achieve. Therefore, as will be briefly outlined in the points 1-10 below, the problem is most conveniently solved by direct computer calculations utilizing an iteration procedure:

1. Choose  $t_c$  to be freely variable in steps of  $\Delta t_c$ , and fix the  $t_d$ - and  $t_i$ -values.
2. Calculate three successive values of  $\sigma/S$  for  $t_c$ -values of  $t_c$ ,  $t_c + \Delta t_c$  and  $t_c + 2\Delta t_c$ .
3. Test which of the situations illustrated in fig.1a-c is present.
4. If 1a (resp. 1c) represents the situation then a new value for  $\sigma/S$  is calculated with the counting time  $t_c + 3\Delta t_c$  (resp.  $t_c - \Delta t_c$ ), and the test is repeated for the  $\sigma/S$ -values corresponding to the counting times  $t_c + \Delta t_c$ ,  $t_c + 2\Delta t_c$  and  $t_c + 3\Delta t_c$  (resp.  $t_c - \Delta t_c$ ,  $t_c$  and  $t_c + \Delta t_c$ ).
5. The sequence "add  $\Delta t_c$  (resp. subtract  $\Delta t_c$ )- calculate  $\sigma/S$  - test" is repeated until the situation in fig.1b is obtained. The values of  $\sigma/S$ ,  $t_i$ ,  $t_d$  and  $t_c$  are then stor-

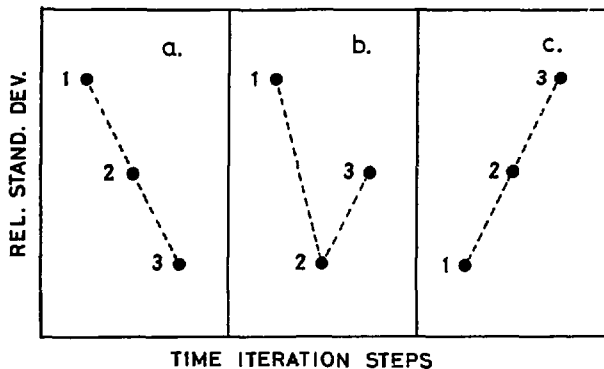


Fig.1. Qualitative illustration of the three situations possible (a, b and c) when calculating the value of the relative standard deviation for three successive values of one of the experimental times (the two others are kept konstant).

- ed in matrix (c). The  $t_c$ -value obtained in this way is the optimum counting time for the preselected  $t_i$ - and  $t_d$ -values.
- Then keep the value of  $t_d$  fixed and repeat the procedure in points 1-5 for the irradiation times  $t_i + \Delta t_i$  and  $t_i + 2\Delta t_i$ .
  - Test the stored matrix-(c) values for the situations in fig.1a-c.
  - If 1a (resp. 1c) represents the situation, then a new sequence of points 1-5 are ran through with the irradiation time  $t_i + 3\Delta t_i$  (resp.  $t_i - \Delta t_i$ ), and the test according to fig.1 is repeated for the  $\sigma/S$ -values in matrix (c) corresponding to the irradiation times  $t_i + \Delta t_i$ ,  $t_i + 2\Delta t_i$  and  $t_i + 3\Delta t_i$  (resp.  $t_i - \Delta t_i$ ,  $t_i$  and  $t_i + \Delta t_i$ ).
  - The sequence "add  $\Delta t_i$  (resp. subtract  $\Delta t_i$ ) - run through the points 1-5 - test" is repeated until the matrix-(c) values of  $\sigma/S$  show the situation in fig.1b. The values of  $\sigma/S$ ,  $t_i$ ,  $t_d$  and  $t_c$  corresponding to the point labeled 2 are then stored in matrix (i). The  $t_c$ - and  $t_i$ -values obtained in this way constitute the optimum combination of irradiation and counting time for a preselected  $t_d$ -value.
  - Now, the procedure in points 1-9 is repeated for the decay times  $t_d + \Delta t_d$  and  $t_d + 2\Delta t_d$ . A minimum value of the  $\sigma/S$ -values in matrix (i) is then found in a similar way as described in the points 7-9 above. This  $\sigma/S$ -value is the absolute minimum for all combinations of the experimental times. Accordingly the corresponding values of  $t_i$ ,  $t_d$  and  $t_c$  constitute the absolute optimum combination.

In order to calculate these optimum values a computer program in the language BASIC, COMB1, is written for use on a computer of modest size. It is intended to be easily understandable and simple in use. This program is included in Appendix I.

Results calculated with this program are given below.

### 3.2. Single cycle activation.

#### 3.2.1. Optimization of the counting time, $t_c$ .

Frequently the most easily variable parameter is the counting time,  $t_c$ . An example where  $t_c$  is freely variable and  $t_d$  and  $t_d$  has to be preselected is treated in this chapter.

In order to make the treatment more general the results calculated are presented (in tables and figures) as functions of the following three dimensionless parameters:

$$C_{11}/C_{12}, \lambda_{12}/\lambda_{11} \text{ and } C_{11}/R_{GB}$$

Here:

$$C_{ij} = K_{1j} \cdot P_{i1j} \cdot \epsilon_{ij} \cdot \sum_{i=1}^{n_i} \lambda_{1j} \cdot \lambda_{2j} \cdot \dots \cdot \lambda_{(i-1)j} \cdot \sum_{i=1}^{n_i} F_{ij} \cdot \sum_{x=0}^{m_{ij}-1} (q-x) \cdot P_{dij}(x)$$

and expresses the maximum number of counts obtainable (infinite counting time) of the nuclide (ij).

For a specific experiment all these three parameters are either known ( $\lambda_{ij}$ ) or can easily be determined ( $R_{GB}$ )/estimated ( $C_{ij}$  by determining  $\epsilon_{ij}$  and estimating  $N_{ij}^0$ ).

The frequently arising case where the main contribution in  $S_{VB}$  comes from one primary formed nuclide only, is specially discussed. Eqn. (32) then turns into the simple expression:

$$\frac{\sigma_{11}}{S_{11}} = \frac{[C_{11} \cdot P_{c11} \cdot P_{\epsilon 11} + 2C_{12} \cdot P_{c12} + 2R_{GB} \cdot t_c]^{\frac{1}{2}}}{C_{11} \cdot P_{c11}}$$

Some calculated results for the optimum  $t_c$  are listed in Appendix II (table I), in units of  $(t_{\frac{1}{2}})_{11}$ . Trends in the vari-



ation of  $t_c(\text{opt})$  with the parameters  $C_{11}/C_{12}$ ,  $\lambda_{12}/\lambda_{11}$  and  $C_{11}/R_{GB}$  are illustrated in the fig.s 2-7. The span in the parameter values used allows one to draw curves covering most of the practical needs that might arise on the basis of the present data.

3.2.2. The optimal combination of the irradiation time,  $t_i$ , and the counting time,  $t_c$ .

In many situations the decay time,  $t_d$ , can be freely chosen only within certain limits (mainly above a lower limit). For instance the transfer time in the rabbit tube will constitute a lower limit for those analysis where such an apparatus can be used. Dealing with very short-lived nuclides this delay time may amount to some hundreds of milliseconds, and for longer-lived nuclides to hours and days (when the radioactive substance has to be sent by car or plane).

However, in such cases where a delay time is dictated, one usually has the opportunity to vary the irradiation time in order to achieve the optimum combination of  $t_i$  and  $t_c$ . Two cases must be separately regarded:

- a.  $\lambda_{12}/\lambda_{11} > 1$ . The  $t_i(\text{opt})$ -value will obviously be "infinite", and the  $t_c(\text{opt})$ -value will be limited only by the general background,  $R_{GB}$ .
- b.  $\lambda_{12}/\lambda_{11} < 1$ . There exists a real optimum combination of  $t_i$  and  $t_c$ . Such optimal combinations are calculated for a number of values of the parameters  $C_{11}/C_{12}$ ,  $\lambda_{12}/\lambda_{11}$  and  $C_{11}/R_{GB}$ , and listed in Appendix II (table II).

It should be noted that here  $C_{ij}$  is given as:

$$C_{ij} = K_{1j} \cdot \epsilon_{ij} \cdot \sum_{i=1}^{n_i} \lambda_{1j} \cdot \lambda_{2j} \cdot \dots \cdot \lambda_{(i-1)j} \cdot \sum_{i=1}^{n_i} F_{ij} \cdot \sum_{x=0}^{m_{ij}-1} (q-x) \cdot P_{dij}^{(x)}$$

The table shows that in the cases where the possible contribution from the variable background,  $S_{VB}$ , is low, the irradi-

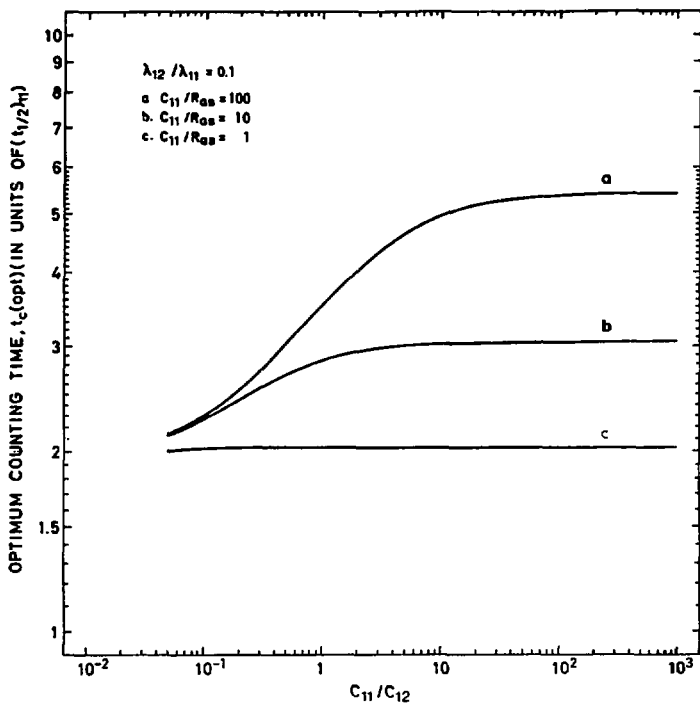


Fig.2. Illustration showing general trends in the variation of the optimum counting time as a function of the dimensionless parameter  $C_{11}/C_{12}$  ( $C_{ij}$ , defined in section 3.2.1). In this special case  $\lambda_{12}/\lambda_{11}$  is constant and  $C_{11}/R_{GB}$  parameter.

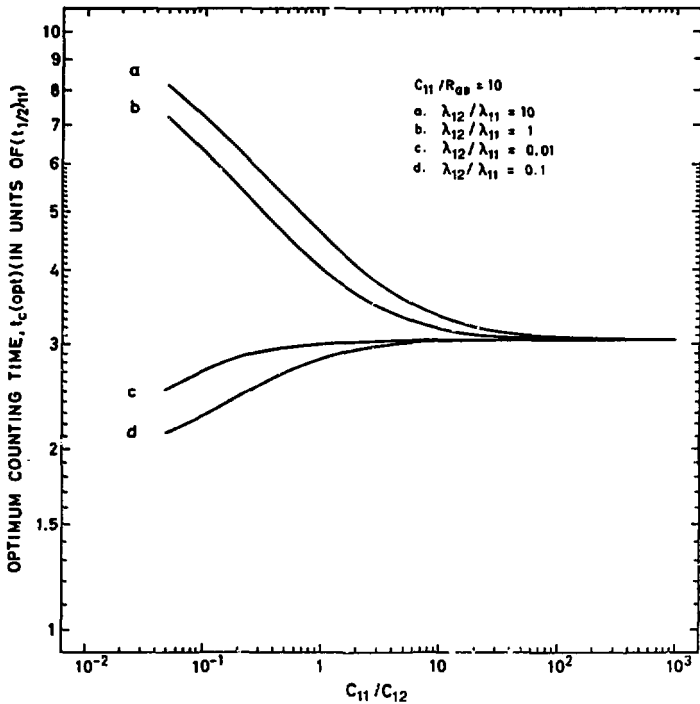


Fig.3. General trends in the optimum counting time as a function of  $C_{11}/C_{12}$ , where  $C_{11}/R_{GB}$  is constant and  $\lambda_{12}/\lambda_{11}$  parameter.

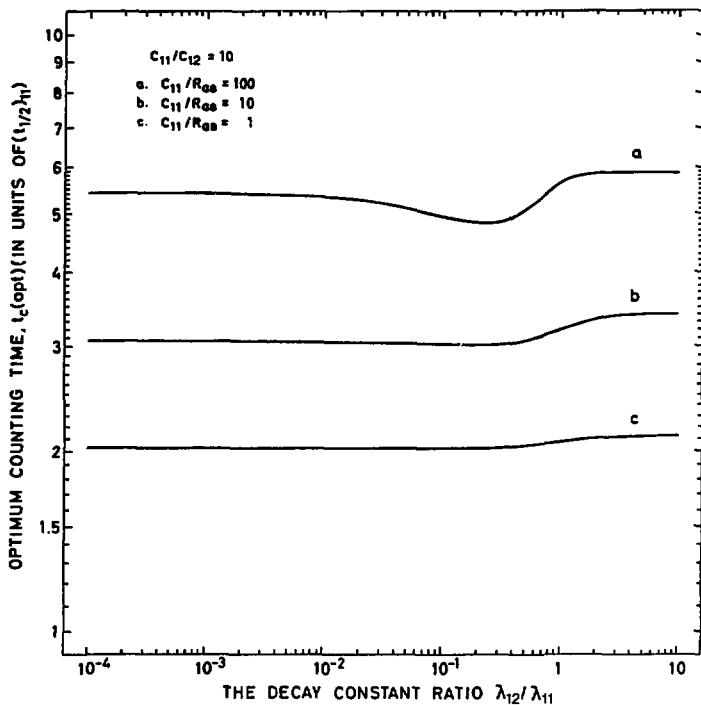


Fig.4. General trends in the optimum counting time as a function of  $\lambda_{12}/\lambda_{11}$  where  $C_{11}/C_{12}$  is constant and  $C_{11}/R_{GB}$  parameter.

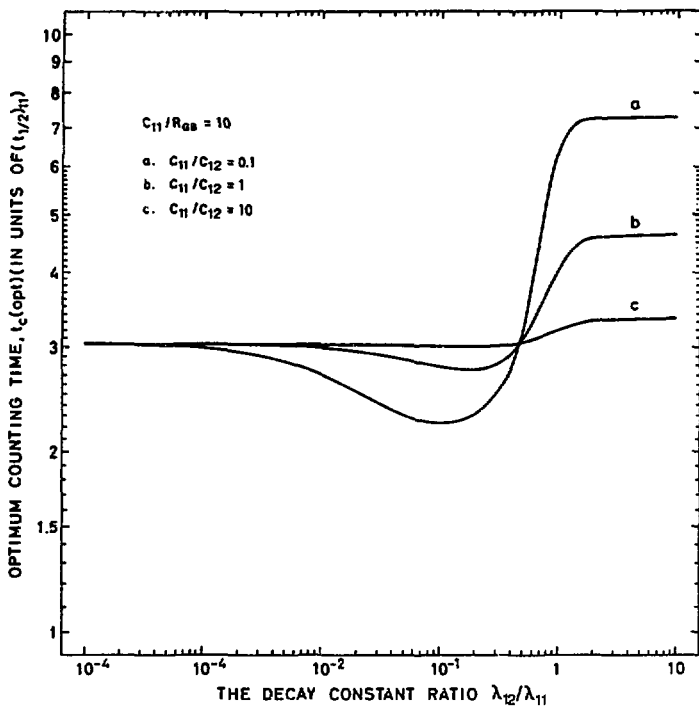


Fig.5. General trends in the optimum counting time as a function of  $\lambda_{12}/\lambda_{11}$  where  $C_{11}/R_{GB}$  is constant and  $C_{11}/C_{12}$  parameter.

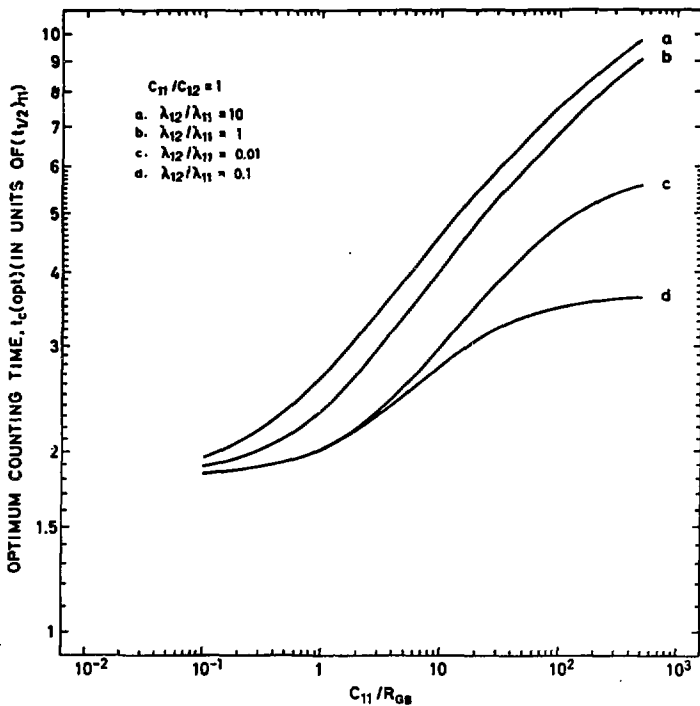


Fig.6. General trends in the optimum counting time as a function of  $C_{11}/R_{GB}$  where  $C_{11}/C_{12}$  is constant and  $\lambda_{12}/\lambda_{11}$  parameter.

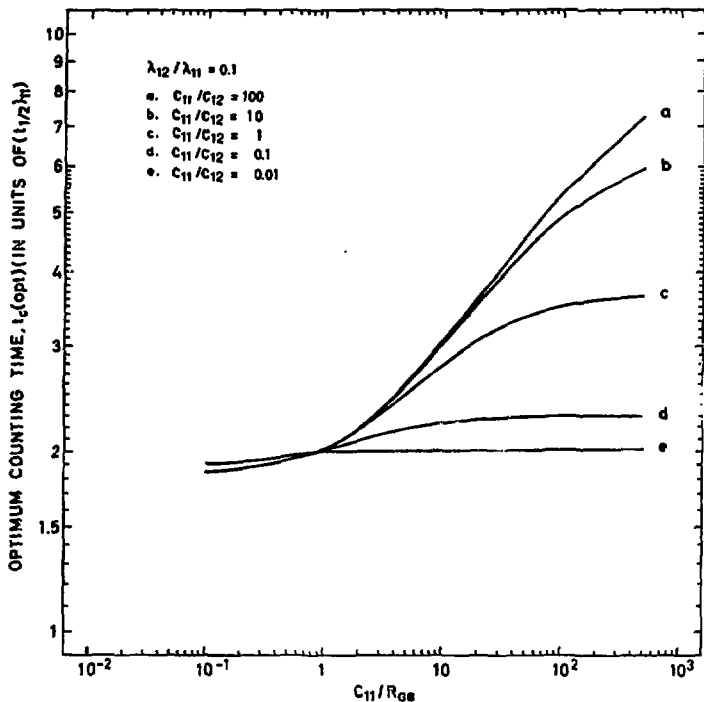


Fig. 7. General trends in the optimum counting time as a function of  $C_{11} / R_{GB}$  where  $\lambda_{12} / \lambda_{11}$  is constant and  $C_{11} / C_{12}$  parameter.

ation time might be high. In order to reduce the cost and the time needed for the analyses an increase in the relative standard deviation of a certain amount may be accepted. A reasonable increase is 10%, and "the best" values of  $t_i$  and  $t_c$  calculated on the basis of this assumption are likewise listed in table II below the real optimal values. The results may be graphically represented either by three-dimensional plots like fig.8 where the optimal combination of  $t_i$  and  $t_c$  constitute a space curve, or by the two-dimensional plots in fig.9 and 10. The latter types of plot is to be preferred because it is simpler and contains more information than the one in fig.8.

3.2.3. The optimal combination of the irradiation time,  $t_i$ , the decay time,  $t_d$ , and the counting time,  $t_c$ .

A significant gain in the analysis quality (expressed by the relative standard deviation) may be obtained if the decay time,  $t_d$ , in addition to  $t_i$  and  $t_c$ , can be freely chosen to give the "best" combination of all three parameters. In this treatment  $C_{ij}$  is given by:

$$C_{ij} = K_{1j} \cdot \epsilon_{ij} \cdot \sum_{i=1}^{n_i} \lambda_{1j} \cdot \lambda_{2j} \cdot \dots \cdot \lambda_{(i-1)j} \cdot \sum_{i=1}^{n_i} F_{ij} \cdot \sum_{x=0}^{m_{ij}-1} (q-x) \cdot [P_{dij}^{(x)} / P_{dij}^{(0)}]$$

Three cases must be separately regarded:

- a.  $\lambda_{12} / \lambda_{11} < 1$ . In this case it is obvious that the optimal decay time is 0, and the "best" combination of  $t_i$  and  $t_c$  can be found in table II. (Mind the meaning of the  $C_{11}$  and  $C_{12}$  values!).
- b.  $1 \leq \lambda_{12} / \lambda_{11} \leq 2$ . By letting the variable background be composed of one primary formed nuclide only and differentiating eqn. (32) with respect to  $t_d$ , it can be shown that  $t_d(\text{opt})=0$ . (This result is of course valid even for several primary formed nuclides contributing to  $S_{VB}$  as long as the assumption for the decay constant ratio holds).



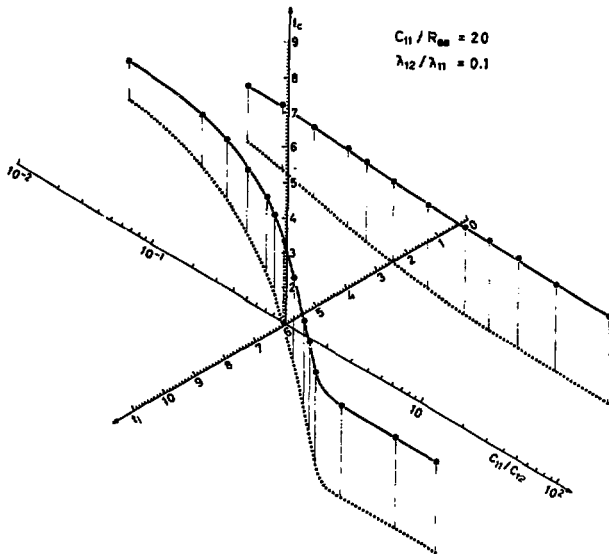


Fig.8. Space curve showing general trends in the optimum combination of  $t_i$  and  $t_c$  as a function of  $C_{11}/C_{12}$  ( $C_{ij}$  defined in section 3.2.2.) for constant value for both  $C_{11}/R_{GB}$  and  $\lambda_{12}/\lambda_{11}$ . Besides, the space curve for the "reasonable" optimum combination (as defined at p.24) of  $t_i$  and  $t_c$  is shown. The dotted lines are the projections of the space curves into the  $t_i$ - $C_{11}/C_{12}$  plane.

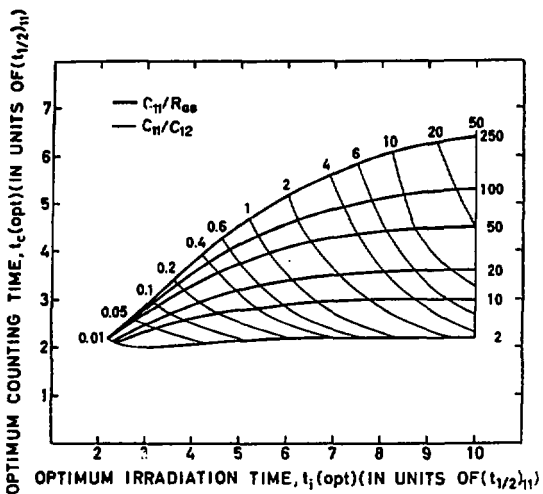


Fig.9. General trends in the optimum combination of  $t_i$  and  $t_c$  exemplified by the case where  $\lambda_{12}/\lambda_{11}=0.1$  and  $C_{11}/C_{12}$  and  $C_{11}/R_{GB}$  are both parameters.

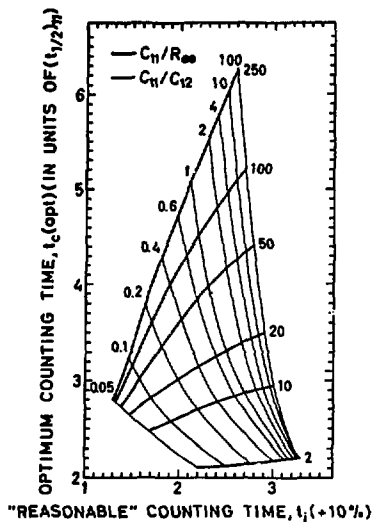


Fig.10. Combination of the "reasonable" ("best practical") optimum value of  $t_i$  and the corresponding optimum value of  $t_c$  exemplified by the case where  $\lambda_{12}/\lambda_{11} = 0.1$  and  $C_{11}/C_{12}$  and  $C_{11}/R_{GB}$  are both parameters.

Besides it is obvious that there is no optimum value of  $t_i$ . But the "best practical" values (defined, like above, as those corresponding to 10% increase in the relative standard deviation calculated with  $t_i(\text{opt})=10 \cdot (t_i)_{11}$ ) are listed in table III (Appendix II) together with the corresponding  $t_c$ -values.

- c.  $\lambda_{12}/\lambda_{11} > 2$ . In this case there is no limited optimum value of  $t_i$ . But by using the "best practical" value as defined above, the corresponding "best combinations" of  $t_d$  and  $t_c$  can be calculated. Table III in Appendix II shows values calculated with stepping length of  $0.1 \cdot (t_i)_{11}$  in  $t_i$  and  $t_c$ , and  $0.01 \cdot (t_i)_{11}$  in  $t_d$ .

In more complex systems where the variable background is composed of several components, the decay constant ratios,  $\lambda_{ij}/\lambda_{11}$ , may be widely spread in values. Then a real optimum combination of all three parameters  $t_i$ ,  $t_d$  and  $t_c$  may be found (the program COMB1 takes care of that!).

A synoptic graphical illustration of the optimal combination of these three experimental times as function of the three dimensionless parameters is not easy to achieve when much compressed information is wanted. It is however possible to make illustrations like fig.11 that might be satisfactorily simple for most of the actual  $C_{11}/R_{GB}$  values.

### 3.3. Accumulation cycles.

When the desirable induced activity is low after one irradiation, the relative standard deviation will be correspondingly high. It might then be necessary or desirable to continue with several successive irradiation-counting cycles, so-called accumulation cycles. The number of necessary cycles,  $q$ , for a special case will then depend upon the specific test-criterion chosen.

When increasing the number of cycles the relative standard deviation of the accumulated net number of counts will continue to decrease infinitely. Accordingly, when using the

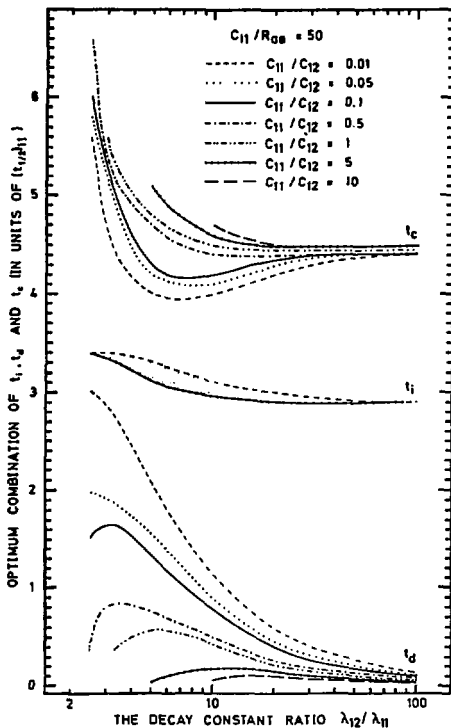


Fig.11. General trends in the optimum combinations of  $t_1$ ,  $t_d$  and  $t_c$  as a function of  $\lambda_{12}/\lambda_{11}$  exemplified by the case where  $C_{11}/R_{GB}=50$  and  $C_{11}/C_{12}$  parameter with values ranging from 0.01 to 10.

relative standard deviation as a test criterion without any additional demands, there is no real optimum number of cycles to be found. However, in practice there has to be a limit to the number of cycles reasonable to perform. Let us consider a case where there is one dominant background component with half-life  $(t_{1/2})_{ij}$ . Then two main situations must be separately regarded:

- a.  $(t_{1/2})_{ij} \leq (t_{1/2})_{11}$ . If the total cycle time  $r \geq 10 \cdot (t_{1/2})_{11}$ , as is a common case, then the contribution from the variable background,  $S_{VB}$ , will be constant from cycle to cycle. Hence, the limiting factor to the number of cycles is one of the two kinds: Desired precision or analysis total time (cost), whichever is the more important.
- b.  $(t_{1/2})_{ij} \gg (t_{1/2})_{11}$ . Suppose that  $r \ll (t_{1/2})_{ij}$ . The variable background activity will then continue to build up for each successive cycle. After  $n$  cycles the following situation may be reached: The total number of counts from the background for the next cycle,  $S_{VB}^{n+1} + S_{GP}$ , become so large that the net number of counts for the nuclide of interest,  $S_{11}^{n+1}$ , is less than the detection limit,  $L_D^{n+1}$ , for the nuclide (1,1). An appropriate definition of  $L_D$  is the one given by Currie (Cur68) where

$$L_D = k^2 + 2L_C \quad (\text{for } k_\alpha = k_\beta)$$

(for the def. of  $k$ ,  $k_\alpha$ ,  $k_\beta$  and  $L_C$  see ref. (Cur68)).

For such cases it may be valuable to introduce the idea of "the reasonable optimum number of accumulation cycles" denoted by  $\bar{q}$ . An acceptable choice will be  $\bar{q} = n$  where  $S_{11}^n > L_D > S_{11}^{n+1}$ . (Alternative ways of choosing the value of  $\bar{q}$  are of course possible, and to a great extent a matter of opinion).

On the basis of the previous discussion it is now possible to construct some applicable formulations to be used in order to select this "reasonable optimum" number of  $\bar{q}$ , all based upon

the relative standard deviation criterion or variations of this. The formulations in points a-d below describe frequently arising situations where certain parameter values are to be calculated in order to fulfil one or more preselected demands:

- a. Preselection:  $\sigma_{11}(\text{rel}) < \text{a preselected value}$ . The values of  $t_i$ ,  $t_d$  and  $t_c$  (average for the  $q$  cycles) are more or less arbitrarily chosen.

To be calculated:

The necessary number of accumulation cycles in order to fulfil the preselections.

Comment:

It is here demanded that the derived value of  $q \leq \bar{q}$ . If it is found that  $q > \bar{q}$  the preselected value of  $\sigma_{11}(\text{rel})$  is unrealistic (too small).

- b. Preselection:  $\sigma_{11}(\text{rel}) < \text{a preselected value}$ , and the analysis total time as small as possible.

To be calculated:

The "best" combination of  $q$  and the average values of  $t_i$ ,  $t_d$  and  $t_c$  (for the  $q$  cycles) in order to fulfil the preselections.

Comment:

As for point a.

- c. Preselection:  $\sigma_{11}(\text{rel})$  at the "reasonable" minimum value (corresponding to  $\bar{q}$  cycles).

To be calculated:

The optimum combination of average values of  $t_i$ ,  $t_d$  and  $t_c$  for the  $\bar{q}$  cycles.

Comment:

This preselection is only to be applied in cases where  $(t_i)_{ij} \gg (t_i)_{11}$ .

If the electronic part of the analysis system offers the possibility of preselecting (programming) the optimum combination of the experimental times for each of the individual accumulation cycles, the following point may be of interest:

- d. Preselection:  $\sigma_{11}(\text{rel})$  at the "reasonable" minimum val-

us (corresponding to  $\bar{q}$  cycles).

To be calcu-

lated: The optimum combination of  $t_1$ ,  $t_d$  and  $t_c$   
for each of the individual cycles  $\bar{q}$ .

Comment: As for point c.

It should be emphasized that the value of  $\bar{q}$  will depend upon the calculated optimum combination values of the experimental times. However, in points c and d  $\bar{q}$  will be determined simultaneously with, and correlated to, these experimental times.



#### 14. EXAMPLES OF APPLICATION.

Among the unlimited number of examples possible, two cases that partly need special treatment are chosen: Analysis of oxygen in aluminium samples, and analysis of boron in organic samples containing oxygen.

##### 4.1. 14 MeV neutron activation analysis of oxygen in aluminium samples.

When irradiating aluminium with 14 MeV neutrons, the two radioactive nuclides  $^{27}\text{Mg}$  and  $^{24}\text{Na}$  are formed through the reactions  $^{27}\text{Al}(n,p)^{27}\text{Mg}$  and  $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ . 99% of the  $^{24}\text{Na}$  decay passes through the  $\gamma$ -cascade 2.7539 MeV + 1.3685 MeV. If the counting geometry is rather close (as it is in the author's laboratory), the risk for summing of the two  $\gamma$ -rays is considerable. A sum peak will appear at 4.1224 MeV. Accidental coincidences may then occur between this sum peak and the rest of the  $\gamma$ -ray spectrum, thus yielding counts within the normally gated energy area of 4.5 MeV - 7.0 MeV. A half-life measurement of the variable background contribution in this area gives values that can be attributed to  $^{27}\text{Mg}$  (9.5m) and  $^{24}\text{Na}$  (15h) as expected [Bj578]. The relative contribution of the two nuclides depends of course strongly upon the irradiation and decay times. Therefore the "counting efficiencies", as input parameters to the program COMB1, has to be recalculated for each combination of irradiation and decay times.

Table 1. Oxygen in aluminium: actual physical constants for 14 MeV n-activation analysis.

Element	Isotope of interest	Nuclear reaction	Cross-section (mb)	Half-life (s)	Dominant $\gamma$ -rays (MeV)
O	$^{16}\text{O}$ (99.76%)	$^{16}\text{O}(n,p)^{16}\text{N}$	$39 \pm 4$	7.13	6.130, 7.117, SE:5.619, 6.606 DE:5.108, 6.095
Al	$^{27}\text{Al}$ (100%)	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	$75 \pm 8$	567.6	0.844, 1.014
		$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	$116 \pm 3$	54000	1.369, 2.754

Table 2. Experimental parameters (for O in Al) commonly fixed in the author's laboratory.

Sample dimensions (l=50.8mm, r=6.25mm):	w=16.5g
Number of $^{27}\text{Al}$ target nuclides pr sample:	$3.68 \cdot 10^{23}$ atoms
Average neutron flux across the sample:	$4.5 \cdot 10^8 \text{ n/s.cm}^2$
Decay time:	3 s
General background count rate:	0.21 cps

#### 4.1.1. Optimum counting time.

Besides the values given in table 1 and 2 above, the irradiation time is also usually fixed to 10 s (due to inflexibility in the apparatus adjustment). On the basis of these constants the K-values (see eqn.(18)) of the variable background components can be calculated (table 3). Measurements of the contribution to the gated energy region from each of the contributing components allow us in addition to calculate the respective counting efficiencies  $\epsilon_{ij}$  (table 3). The calculations are based on one accumulation cycle and 10 s irradiation time.

Table 3. Calculated K-values and counting efficiencies for the variable background components, and the counting efficiency of the  $^{16}\text{N}$ -activity.

$K_{12}(^{27}\text{Mg})$	$K_{13}(^{24}\text{Na})$	$\epsilon_{11}(^{16}\text{N})$	$\epsilon_{12}(^{27}\text{Mg})$	$\epsilon_{13}(^{24}\text{Na})$
$6.8 \cdot 10^8$	$1.497 \cdot 10^{12}$	$4.5 \cdot 10^{-2}$	$4.25 \cdot 10^{-7}$	$3.92 \cdot 10^{-6}$

For oxygen contents in the range 10-100 ppm, and for one accumulation cycle, the calculated optimum counting times are given in table 4.

Table 4. Optimum and "best practical" values of the counting time (and corresponding relative standard deviation) for Al-samples containing 10-100 ppm oxygen.

Oxygen-content (ppm)	Corresp. K-value	Optimum $t_c$ in units of $(t_{\frac{1}{2}})_{11}$	Corresp.*) rel.stand. dev. (%)	"Best practic." $t_c$ in units of $(t_{\frac{1}{2}})_{11}$
10	1122	3.16	27.5	1.47
20	2244	3.77	17.4	1.70
30	3366	4.18	13.5	1.83
40	4488	4.50	11.4	1.92
50	5610	4.75	10.0	1.99
60	6732	4.96	9.0	2.04
70	7854	5.15	8.3	2.08
80	8976	5.31	7.7	2.12
90	10098	5.46	7.2	2.15
100	11220	5.59	6.8	2.17

\*) The relative standard deviation corresponding to the "best practical" values are obtained by adding 10% to the respective optimum ones.

#### 4.1.2. Irradiation and counting times,- optimum and "best practical" combinations.

The counting efficiencies  $\epsilon_{12}$  and  $\epsilon_{13}$  are not absolute systemdependant constants, but their values increase with the desintegration rate of  $^{24}\text{Na}$  according to the formulas [Bj678]:

$$\epsilon_{12} = k_{12} \cdot D_{13}$$

$$\epsilon_{13} = k_{13} \cdot D_{13}$$

where  $k_{12}$  and  $k_{13}$  are systemdependant constants and  $D_{13}$  the desintegration rate of  $^{24}\text{Na}$ . In the author's laboratory these constants usually take the values  $1.43 \cdot 10^{-12}$  and  $1.327 \cdot 10^{-11}$  respectively. This dependance of the variable background efficiency upon  $D_{13}$  will contribute largely to the limitation of the number of accumulation cycles, especially for samples with low oxygen content (see next chapter). In order to reduce the variable background contribution to an acceptable level, the cycles have to be rather widely spaced in time. Hence, one

will be obliged to find a compromise between the analysis total time and the acceptable standard deviation.

A total cycle time of 2 hours is necessary to avoid the building up of the  $^{27}\text{Mg}$ -activity. According to this, the optimum combination (average values) of  $t_i$  and  $t_c$ , and the "best practical"  $t_i$  and the corresponding  $t_c$  are calculated for samples containing 10-100 ppm oxygen, and for 1 -10 accumulation cycles. (For this calculation a slightly modified procedure has to be used, described in the last section of Appendix I). The results are illustrated in fig 12. The curves show that the "best practical"  $t_i$  will reduce the irradiation cost 40-50% relative to the optimum  $t_i$ -value (since the cost in the first approximation is proportional to the irradiation time). This reduction becomes increasingly more important with decreasing number of cycles and increasing oxygen content.

#### 4.1.3. More about accumulation cycles.

Besides fig.12 (showing the variation in the experimental times with the number of cycles as parameter), fig. gives the corresponding optimum relative standard deviation. The curves are step functions with the number of accumulations on the abscissa and the oxygen content as a parameter. Due to the functional connection between the counting efficiencies and the desintegration rate of  $^{24}\text{Na}$ , these curves show minimum values. For a sample containing 10 ppm oxygen the minimum is reached after 5 cycles, for a sample containing 50 ppm after 10 cycles. In the same figure the levels corresponding to 10% increase in the relative standard deviation are indicated (dashed lines) for the five samples containing 10, 20, 30, 40 and 50 ppm oxygen. In order to creep under these levels, 3,3,4,4 and 5 cycles are needed for the five mentioned samples respectively.

Since samples of pure aluminium normally contain less than 50 ppm oxygen, a number of 4 cycles would be, in average, recommended. Corresponding average values of the experi-

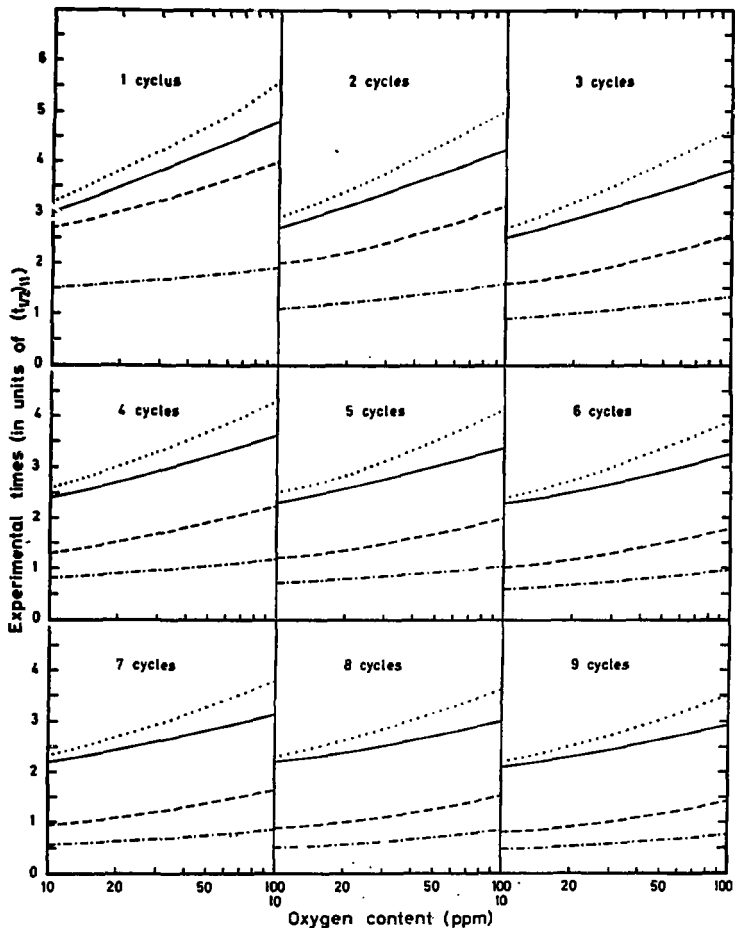


Fig.12. Average optimum combination of  $t_i$  (-----) and  $t_c$  (——), and the "reasonable" value of  $t_i$  (····) with the corresponding optimum  $t_c$  (-·-·-) as a function of the oxygen content with the number of accumulation cycles as parameter.

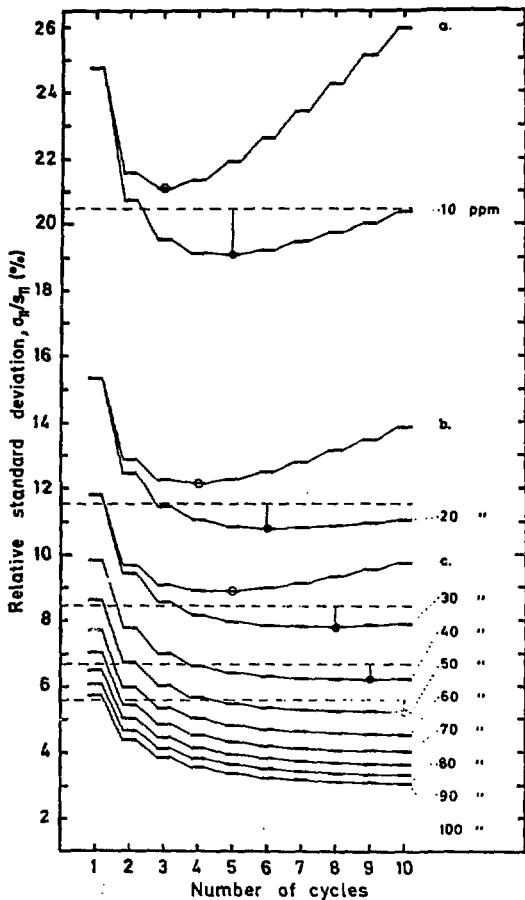


Fig.13. Relative standard deviations (corresponding to the optimum combination values of  $t_i$  and  $t_c$ ) as a function of the number of cycles, with the oxygen content as a parameter. The total cycle time is 7200s. For the curves a., b. and c. the total cycle time is 600s. The circles (• or o) marks the minimum of each curve.

mental times will then be (from fig.12):  $t_i(\text{opt}) \approx 11.5\text{s}$ ,  $t_c(\text{opt}) \approx 20\text{s}$ ,  $t_i(\text{best}) \approx 6.5\text{s}$  and  $t_c(\text{corresp}) \approx 23\text{s}$ .

As mentioned earlier, due to inflexibility in the present counting system in the author's laboratory, the irradiation time is preset to 10s (the nearest possible value to the 11.5s derived above). The corresponding step function of the relative standard deviation (for  $t_i = 10\text{s}$ ) is shown in fig.14 for a sample containing 10 ppm oxygen. The minimum value is now shifted to the left, and reached after 4 cycles. According to fig.12 this number of cycles will for the present sample correspond to an optimum counting time of approximately 17s (provided that  $t_i$  has the corresponding optimum value of about 9s). The nearest presently possible adjustment value, 20s, is then used as the overall most reasonable value for the counting time.

As shortly mentioned above, the shorter the cycle time, the more serious is the affection of the relative standard deviation due to increase in the variable background activity. This is demonstrated by the curves denoted a., b. and c. in fig.13 (corresponding to an oxygen content of 10, 20 and 30 ppm). Here the total cycle time is 600s, and the building up of the  $^{27}\text{Mg}$ -activity becomes increasingly important.

#### 4.2. 14 MeV n-activation analysis of boron in samples containing oxygen.

It is supposed here in this example that the only contributors to the total sample activity is the reaction products of boron and oxygen. This will mainly restrict the samples to organic materials, excluding halogen compounds and a few other types of compounds (for instance organo-metallic compounds where the reaction products are short-lived).

An apparatus specially designed for cyclic activation is used for the boron analysis. The produced  $\beta$ -activity is detected by a thin (minimum  $\gamma$ -response) plastic scintillator (NE 102). Table 5 and 6 below give some parameter values necessary

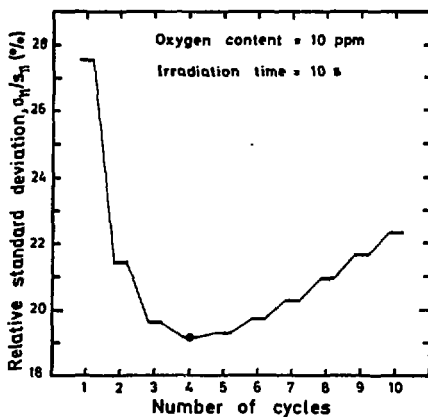


Fig.14. The relative standard deviation as a function of the number of cycles for  $t_i = 10s$  (and the corresponding optimum  $t_c$ ) and an oxygen content of 10 ppm.



for the calculations.

Table 5. Boron in oxygencontaining materials: actual physical constants for 14 MeV n-activation analysis.

Element	Isotope (natural abund.)	Nuclear reaction	Cross-section (mb)	Half-life (s)	Dominant $\beta$ -energies (MeV)
B	$^{11}\text{B}$ (80%)	$^{11}\text{B}(n,\alpha)^8\text{Li}$	30	0.84	13(100%)
O	$^{16}\text{O}$ (99.76%)	$^{16}\text{O}(n,p)^{16}\text{N}$	39±4	7.13	10.422(26%) 4.291(69%)

Table 6. Experimental parameters commonly fixed for the boron analysis.

Sample dimensions: l=15mm, r=2.5mm $\implies$	$V = 0.29 \text{ cm}^3$
Average neutron flux:	$1.0 \cdot 10^9 \text{ n/cm}^2\text{s}$
Decay time:	0.6s
General background count rate:	50 cps

The counting efficiencies of boron,  $\epsilon_{11}$ , and oxygen,  $\epsilon_{12}$ , can be found in the following way: Irradiate a sample of well known composition of boron and oxygen. Measure the  $\beta$ -activity with a multichannel analyzer in the multiscaling mode. The counting rates of the components (mainly  $^8\text{Li}$  and  $^{16}\text{N}$ ) follow immediately from the resolution of the decay curve. From these values and the data given in the tables 5 and 6, the efficiencies can be calculated. For the special experimental equipment utilized in the present laboratory the efficiencies are:

$$\epsilon_{11}(\text{boron}) = 0.065, \quad \epsilon_{12}(\text{oxygen}) = 0.041$$

#### 4.2.1. The counting time, - optimum and "best practical" values.

In addition to the data given in tables 5 and 6, the example treated here uses the following fixed parameter values:

irradiation time per cycle is 1s and the duration of one cycle is 5s. Fig.15 illustrates the trends in the mean optimum and "best practical" counting times for 10 accumulation cycles as a function of the boron content in the sample, and with the oxygen content as a parameter. As expected the oxygen content affects the results seriously. For a boron content of 0.1g the optimum  $t_c$ -value ranges from  $2.70 \cdot (t_i)_{11}$  to  $5.32 \cdot (t_i)_{11}$  when the oxygen content varies from 0.1g to 0.0001g. (To avoid misunderstanding it should be pointed out here that the real  $t_c$ -value used in an experiment is most often the "best practical" value. Then the total time consumed for one accumulation cycle is less than the 5s used in the calculations!).

#### 4.2.2. Irradiation and counting times,- the optimum and "best practical" combinations.

The variation of these parameter values as function of the oxygen content is shown in fig.16 for a sample containing 0.01g of boron, 10 accumulation cycles and a total cycle time of 5s. The curves for the optimum  $t_c$  and the  $t_c$ -value corresponding to the "best practical"  $t_i$  are not resolved in the figure.

The transfer time from counting position to irradiation position is 0.6s. Then the figure shows that for oxygen contents below some 0.005g the optimum combination of  $t_i$  and  $t_c$  will imply a total cycle time exceeding the 5s supposed in the calculations. Accordingly if 5s is an overall desirable cycle time (especially with regard to standardization of the analytical procedure), one has to accept a relative standard deviation higher than the minimum obtainable. The use of the "best practical"  $t_i$  and the corresponding  $t_c$  is suitable for keeping the total cycle time within the 5s for the whole range of oxygen contents shown.

#### 4.2.3. Accumulation cycles.

Fig.17 illustrates the influence of the number of accumulation cycles upon the relative standard deviation for some

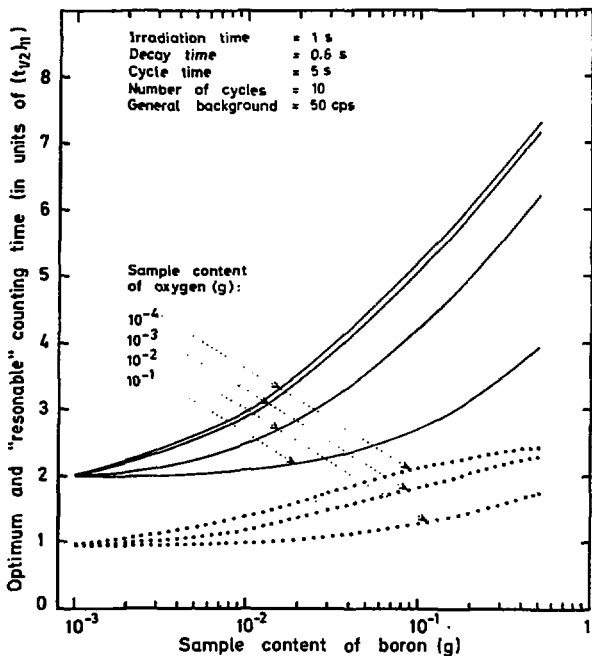


Fig.15. Trends in the average optimum and "reasonable"  $t_c$  for 10 accumulation cycles as a function of the sample content of boron. The oxygen content is parameter.

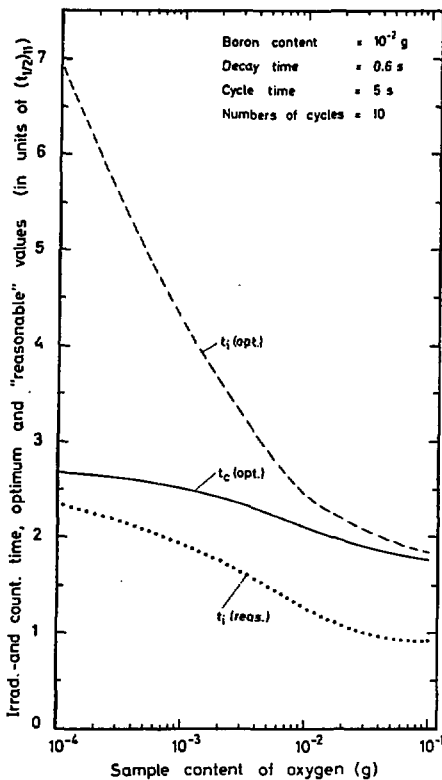


Fig.16. Trends in the variation of the optimum and "reasonable" combinations of  $t_i$  and  $t_c$  as a function of the oxygen content for a sample containing 0.01g boron.

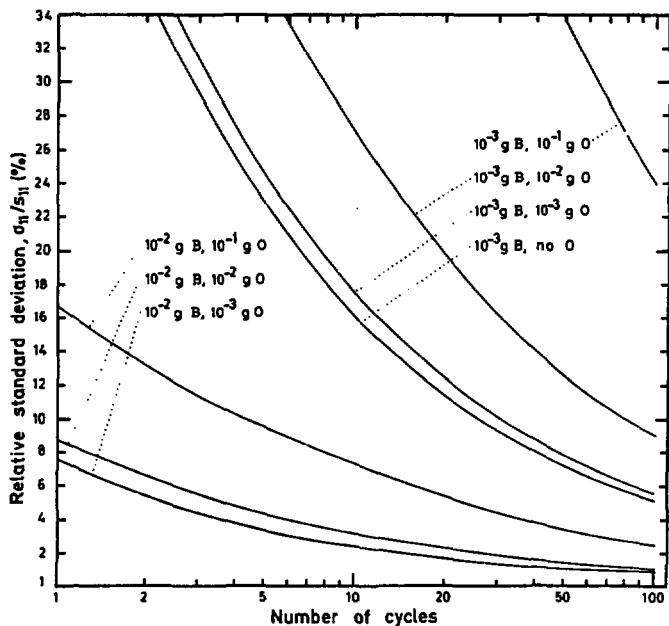


Fig.17. Relative standard deviation as a function of the number of cycles for some samples with arbitrarily chosen contents of boron and oxygen, when  $t_i = 1$  s,  $t_d = 0.6$  s,  $t_c = 2.8$  s and the cycle total time is 5 s.

samples with arbitrarily chosen contents of boron and oxygen. If, for instance, highest allowed value of the relative standard deviation is preset to 10%, the curves show that for a sample containing 0.001g of boron and 0.001g of oxygen 30 cycles are needed. However, if the oxygen content raises to 0.01g a number of 80 cycles are needed.

In this way, by employing the program COMB1 (see Appendix I) curves for the experimental times related to special problems (defined type of sample and experimental setup) may be calculated and used for selecting the parameter values according to the present needs.

## 5. CONCLUDING REMARKS.

This work describes a method and outlines a procedure for optimization of an activation analysis with respect to the experimental times irradiation time,  $t_i$ , decay time,  $t_d$  and counting time,  $t_c$ . The method is based on the "minimum relative standard deviation criterion", and specially designed for the use on short-lived nuclides. A computer program, COMB1, is written in the BASIC language in order to make the calculations easier and faster. It is intended to be understandable, and easily applicable on a computer of modest size.

If applied properly this method can guide us to make full benefit out of the experimental apparatus at hand by selecting the real optimum combinations of the experimental times. However, time and cost are important factors, especially for routine analysis on a service basis. In those cases one can often allow a controlled reduction in the analysis result quality (through a higher relative standard deviation). Therefore, with respect to such wishes, the outlined procedure can also help us to find acceptable conditions by calculation of the "best practical" (or "reasonable") experimental time values, and the minimum number of accumulation cycles necessary to fulfil the demands given.

This procedure constitutes a tool that every analyst should have in his tool box. He should frequently be reminded of its presence, and told: Take it, - if necessary sharpen it, - but anyhow make use of it!

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APPENDIX I.

COMB1 - users manual.

Introduction.

COMB1 is a program for calculation of optimum values of the experimental times irradiation time,  $t_i$ , decay time,  $t_d$  and counting time,  $t_c$ , in activation analysis using short-lived nuclides. In addition the "best practical" (or "reasonable") values (defined at p.24 in the text) of  $t_i$  and  $t_c$  are calculated. The calculations are based on the "minimum relative standard deviation criterion" expressed by the formula given in eqn.(37). The working principles are given at p.20-21 in the text. The program is written in the BASIC computer language for interactive use on a computer of modest size (but does not claim to be the work of a programming specialist!).

Practical use.

The program is intended to be self-explaining, but some additional comments are anyhow needed.

When commanding to run the program a series of questions are successively displayed on the output device (data screen or teletype). The answer of one question initiates the next. The questions are as follows:

Question 1: GIVE THE VALUE OF V:?

Comment : The parameter V can take any of the three values 1, 2 or 3. The values define the three subprograms TOPT1 (calculation of optimum  $t_c$ ), TOPT2 (optimum combination of  $t_i$  and  $t_c$ ) and TOPT3 (optimum combination of  $t_i$ ,  $t_d$  and  $t_c$ ), respectively.

Question 2: NUMBER OF PRIMARY FORMED NUCLIDES CONTRIBUTING:?

Comment : The answer must be the total number of nuclides (j) formed in nuclear reactions during irradiation that directly (or indirectly via daughter products) contribute to the accumulated number of counts.

Question 3: GENERAL BACKGROUND COUNT RATE:?

Comment : The answer may be given in cps, cpm or for instance  $cp(t_{1/2})_{11}$  (half-life of the nuclide to be determined). But once the time unit is defined mind to use it throughout the calculations.

Question 4: NUMBER OF ACCUMULATION CYCLES:?

Comment : The answer (q-value) must be a positive integer. If  $q=1$  the program jumps to question 6. If  $q \geq 1$  the program will proceed to the next question.

Question 5: DURATIDN OF ONE ACCUMULATION CYCLE:?

Comment : The answer can be freely chosen among the real numbers, and expresses the total time for the following steps in the procedure: transfer to the irradiation position, irradiation, decay (including transfer to the counting position), counting, and time for some extra handling. NB! Make sure that the value given corresponds to the time unit chosen in the answer of question 3.

Question 6: NUMBER OF NUCLIDES IN DECAY CHAIN 1:?

Comment : All the contributing nuclides are defined by the index  $(i,j)$ , where  $j$  means the primary formed nuclide no.  $j$ , and  $i$  the decay chain member no.  $i$  in the decay chain starting with the nuclide  $j$ . (The equivalent notation in the text is the abbreviated form  $ij$ ). The answer of this question must therefore be the number of members in decay chain no. 1 that contribute to the recorded number of counts.

Question 7: VALUE DF  $K(1,1)$ :?

Comment :  $K$  is defined by eqn.(18) at p.11 in the text, and must be precalculated or estimated.

Question 8: HALF-LIFE AND EFFICIENCY:?

Comment : The half-life of the nuclide defined by the index  $(1,1)$  and its absolute counting efficiency

within the gated energy region must be given. (it is supposed here that the counting efficiency,  $\epsilon$ , is a constant figure throughout the experiment. In cases where  $\epsilon$  can be considered as a variable, for instance for the variable back ground components  $^{24}\text{Na}$  and  $^{27}\text{Mg}$  in the 14 MeV n-activation analysis of oxygen in aluminium, the figure given as answer here is a dummy one. Accordingly any number can be used. This special case is closer described in the last section of Appendix I). The values are separated by a comma. If the values of j (given as answer to question 2) and/or i (given as answer of question 6) is greater than 1, questions 6, 7 and 8 are repeated until all the contributing nuclides are defined by their K-value, half-life and efficiency.

If V=1 (2 or 3) the following text is now displayed:

TOPT1 (2 or 3) ON LINE

If V=1 the program will now jump to question 11, if V=2 to question 10 and if V=3 the program proceeds to the next question.

Question 9: DECAY TIME STEP LENGTH:?

Comment : This is the value by which the decay time should be changed for each iteration cycle in the calculation (defined by  $\Delta t_d$  at p.21 in the text). The minimum value of the step length is for practical reasons suggested to be  $0.01 \cdot (t_{1/2})_{11}$ .

Question 10: IRRADIATION TIME STEP LENGTH:?

Comment : As for question 9 (just change  $\Delta t_d$  into  $\Delta t_i$ ). If V=2 the program jumps to question 12, if V=3 to question 13 and if V=1 the program proceeds to the next question.

Question 11: IRRADIATION TIME:?

Comment : Any positive number is accepted.

Question 12: DECAY TIME:?

Comment : Any positive number is accepted.

Question 13: COUNTING TIME STEP LENGTH:?

Comment : The same as for question 9 (just change  $\Delta t_d$  to  $\Delta t_c$ ).

By this all the input data needed for the computation are completed, and the calculations start.

Examples of the program output.

All the calculated results for the optimum and "best practical" ("reasonable") experimental time values are here given in seconds. The calculated standard deviations are the relative values.

The first example is the calculation of optimum and "best practical" values of the counting time in 14 MeV n-activation analysis of a sample containing ~ 0.01g of both boron and oxygen (see point 4.2. in the text). The complete listing is as follows:

```
GIVE THE VALUE OF V: ?1
NUMBER OF PRIMARY FORMED NUCLIDES CONTRIBUTING: ?2
GENERAL BACKGROUND COUNT RATE: ?50
NUMBER OF ACCUMULATION CYCLES: ?10
DURATION OF ONE ACCUMULATION CYCLE: ?8
NUMBER OF NUCLIDES IN DECAY CHAIN 1: ?1
VALUE OF K(1,1): ?16200
HALF-LIFE AND EFFICIENCY: ?0.84,0.065
NUMBER OF NUCLIDES IN DECAY CHAIN 2: ?1
VALUE OF (1,2): ?151000
HALF-LIFE AND EFFICIENCY: ?7.13,0.041
```

TOPT1 ON LINE

```
IRRADIATION TIME: ?1.5
DECAY TIME: ?0.6
COUNTING TIME STEP LENGTH: ?0.0084
```

```
OPT.STAND.DEV.      = 2.74520E-2
OPT.COUNT.TIME     = 2.1252
OPT.STAND.DEV.+10% = 3.01972E-2
CORRESP.COUNT.TIME = 0.9912
```

The results for the optimum and "best practical" (here called

"corresp.") counting time values correspond to 2.53 and 1.18 units of  $(t_{\frac{1}{2}})_{11}$  respectively.

The following example is the display from the calculation of optimum and "best practical" combinations of the irradiation and counting times for the same sample:

```
GIVE THE VALUE OF V:?2
NUMBER OF PRIMARY FORMED NUCLIDES CONTRIBUTING:?2
GENERAL BACKGROUND COUNT RATE:?50
NUMBER OF ACCUMULATION CYCLES:?10
DURATION OF ONE ACCUMULATION CYCLE:?8
NUMBER OF NUCLIDES IN DECAY CHAIN 1:?1
VALUE OF K(1,1):?16200
HALF-LIFE AND EFFICIENCY:?0.84,0.065
NUMBER OF NUCLIDES IN DECAY CHAIN 2:?1
VALUE OF K(1,2):?151000
HALF-LIFE AND EFFICIENCY:?7.13,0.041

TOPT2 ON LINE

IRRADIATION TIME STEP LENGTH:?0.084
DECAY TIME:?0.6
COUNTING TIME STEP LENGTH:?0.084

OPT.STAND.DEV.   = 2.59622E-2
OPT.IRR.TIME    = 2.604
OPT.COUNT.TIME  = 2.1

OPT.STAND.DEV.+10% = 2.81347E-2
BEST IRR.TIME     = 1.344
CORRESP.COUNT.TIME = 2.1
```

The calculated optimum and "best practical" irradiation time values correspond to 3.1 and 1.6 units of  $(t_{\frac{1}{2}})_{11}$  respectively. The optimum counting time and the value of the counting time that corresponds to the "best practical" irradiation time are identical (within the counting time step length used). They correspond to 2.5 units of  $(t_{\frac{1}{2}})_{11}$ .

The last example shows the output from the calculation of the optimum combination of irradiation, decay and counting time values in a 14 MeV n-activation analysis of oxygen (content ~0.001g) in an organic matrix containing boron (content ~ 0.1g). The sample size and experimental technique are the same as for the boron analysis described in the text:

GIVE THE VALUE OF V: ?3  
NUMBER OF PRIMARY FORMED NUCLIDES CONTRIBUTING: ?2  
GENERAL BACKGROUND COUNT RATE: ?50  
NUMBER OF ACCUMULATION CYCLES: ?1  
NUMBER OF NUCLIDES IN DECAY CHAIN 1: ?1  
VALUE OF K(1,1): ?15100  
HALF-LIFE AND EFFICIENCY: ?7.13,0.041  
NUMBER OF NUCLIDES IN DECAY CHAIN 2: ?1  
VALUE OF K(1,2): ?162000  
HALF-LIFE AND EFFICIENCY: ? 0.64,0.065

TOPT3 ON LINE

DECAY TIME STEP LENGTH: ?0.2  
IRRADIATION TIME STEP LENGTH: ?1  
COUNTING TIME STEP LENGTH: ?0.1

OPT.STAND.DEV. = 0.155323  
OPT.IRR.TIME = .71.3  
OPT.DECAY TIME = 4.2  
OPT.COUNT.TIME = 16.93

Since the only interfering activity has a half-life less than the half-life of the nuclide to be determined, it is obvious that no optimum irradiation time exists, but in fact  $t_i(\text{opt}) \rightarrow \infty$ . Therefore, for practical reasons, the program has a build-in limit of  $10 \cdot (t_{1/2})_{11}$  both for the irradiation and counting times. These practical limit values will be printed if the real optimum values are larger than this limit. The calculated results in this example (in the order given above) therefore correspond to 10, 0.59 and 2.65 units of  $(t_{1/2})_{11}$  respectively.

As will be seen, TOPT3 does not give "best practical" values. If the "best practical" irradiation time is wanted, the program has to be restarted with V=2, and the calculated optimum decay time as input value for this parameter.

On the following pages a listing of the program is given.

Listing of the program COMB1.

```
192 DIM D(2,1000),C(3,1000),G(4,1000)
200 PRINT "GIVE THE VALUE OF V:?",
210 INPUT V
220 PRINT "NUMBER OF PRIMARY FORMED NUCLIDES CONTRIBUTING:?",
240 INPUT A
250 PRINT "GENERAL BACKGROUND COUNT RATE:?",
260 INPUT B
270 PRINT "NUMBER OF ACCUMULATION CYCLES:?",
272 INPUT Q
280 IF Q=1 GO TO 340
285 PRINT "DURATION OF ONE ACCUMULATION CYCLE:?",
287 INPUT R
340 FOR J=1 TO A
345 PRINT "NUMBER OF NUCLIDES IN DECAY CHAIN",J,":?",
350 INPUT N(J)
351 PRINT "VALUE OF K(1,"J,"):?",
352 INPUT A(1,J)
360 FOR I=1 TO N(J)
365 PRINT "HALF-LIFE AND EFFICIENCY:?",
370 INPUT H(I,J);E(I,J)
380 L(I,J)=LOG(2)/H(I,J)
382 IF Q>1 GO TO 390
384 R=10*H(I,J)
390 M(I,J)=INT((10*H(I,J)/R)+0.4)
400 NEXT I
410 NEXT J
420 ON V GO TO 500,900,1100
500 PRINT
501 PRINT "TOPT1 ON LINE"
502 PRINT
530 PRINT "IRRADIATION TIME:?"
535 INPUT T1
548 IF P6>1 GO TO 615
549 IF V=3 GO TO 590
550 PRINT "DECAY TIME:?"
555 INPUT T2
590 T3=1.0*H(1,1)'INITIAL VALUE OF THE COUNTING TIME
592 IF P8>1 GO TO 615
600 PRINT "COUNTING TIME STEP LENGTH:?"
602 INPUT T9
615 P4=0
620 FOR P1=T3 TO 10*H(1,1) STEP T9
630 P4=P4+1
640 GOSUB 1400
650 GOSUB 1600
660 J=1
670 N=2
675 IF N(J)>N GO TO 680
676 I3=1
680 GOSUB 1690
690 C1=B9
691 IF A<2 GO TO 748
```



```
700 FOR J=2 TO A
710 N=1
712 IF J>2 GO TO 720
715 C2=0
720 GOSUB 1690
730 C2=C2+89
740 NEXT J
747 GO TO 750
748 C2=0
750 D(1,P4)=(1/A5)*SQR(A5*A2+A1+C1+C2)
760 D(2,P4)=P1
770 IF P4<3 GO TO 850
780 IF D(1,P4-1)>D(1,P4) GO TO 850
790 IF D(1,P4-1)>D(1,P4-2) GO TO 840
792 ON V GO TO 800,960,960
800 PRINT
801 PRINT "OPT.STAND.DEV.          =" ;D(1,P4-1)
802 PRINT "OPT.COUNT.TIME         =" ;D(2,P4-1)
810 GOSUB 2000
820 PRINT "OPT=STAND.DEV.+10%    =" ;D(1,P5)
822 PRINT "CORRESP.COUNT.TIME    =" ;D(2,P5)
830 GO TO 2500
840 T9=-ABS(T9)
850 NEXT P1
900 PRINT "TOPT2 ON LINE"
902 IF V=2 GO TO 910
904 IF P8>1 GO TO 930
910 PRINT "IRRAQIATION TIME STEP LENGTH:?" ;
920 INPUT T7
930 T1=H(1,1)'INITIAL VALUE FOR THE IRRAQIATION TIME
935 P6=0
940 FOR T1=T1 TO 10*H(1,1) STEP T7
945 P6=P6+1
950 GO TO 548
960 C(1,P6)=D(1,P4-1)'OPT.STAND.DEV.
970 C(2,P6)=D(2,P4-1)'OPT.COUNT.TIME
980 C(3,P6)=T1'OPT.IRR.TIME
990 IF P6<3 GO TO 1070
1000 IF C(1,P6-1)>C(1,P6) GO TO 1070
1010 IF C(1,P6-1)>C(1,P6-2) GO TO 1070
1015 IF V=3 GO TO 1170
1028 PRINT
1030 PRINT "OPT.STAND.DEV.          =" ;C(1,P6-1)
1032 PRINT "OPT.IRR.TIME           =" ;C(3,P6-1)
1034 PRINT "OPT.COUNT.TIME          =" ;C(2,P6-1)
1040 GOSUB 2050
1048 PRINT
1050 PRINT "OPT.STAND.DEV.+10%       =" ;C(1,P7)
1052 PRINT "BEST IRR.TIME           =" ;C(3,P7)
1054 PRINT "CORRESP.COUNT.TIME    =" ;C(2,P7)
1060 GO TO 2500
1070 NEXT T1
1071 IF V=2 GO TO 1028
1072 GO TO 1170
```

```
1100 PRINT
1110 PRINT "TOPT3 ON LINE"
1120 PRINT "DECAY TIME STEP LENGTH:?" ;
1130 INPUT T8
1140 P8=0
1142 T2=0
1144 IF T2=0 GO TO 1308
1155 P8=P8+1
1160 GO TO 902
1170 G(1,P8)=C(1,P8-1)'OPT.STAND.DEV.
1180 G(2,P8)=C(3,P8-1)'OPT.IRR.TIME
1190 G(3,P8)=C(62,P8-1)'OPT.COUNT.TIME
1200 G(4,P8)=T2'OPT.DECAY TIME
1210 IF P8<3 GO TO 1300
1220 IF G(1,P8-1)>G(1,P8) GO TO 1300
1230 IF G(1,P8-1)>G(1,P8-2) GO TO 1292
1240 PRINT
1250 PRINT "OPT.STAND.DEV.           = ";G(1,P8-1)
1260 PRINT "OPT.IRR.TIME           = ";G(2,P8-1)
1270 PRINT "OPT.DECAY TIME         = ";G(4,P8-1)
1280 PRINT "OPT.COUNT.TIME         = ";G(3,P8-1)
1290 GO TO 2500
1292 T8=-ABS(T8)
1300 GO TO 1146
1308 PRINT
1310 PRINT "OPT.DECAY TIME=0, FOR CALCULATION OF OPT.IRR."
1312 PRINT "AND OPT.COUNT. TIMES USE T8 WITH DECAY TIME=0"
1314 GO TO 2500
1400 REM SUBROUTINE FOR CALCULATION OF POTENSES
1402 FOR J=1 TO A
1410 FOR I=1 TO N(J)
1430 Y(I,J)=L(I,J)*T2
1440 X(I,J)=L(I,J)*T1
1450 Z(I,J)=L(I,J)*P1
1460 NEXT I
1470 NEXT J
1480 RETURN
1600 REM SUBROUTINE FOR CALCULATION OF THE DENOMINATOR+ THE
1601 REM EFFICIENCY FACTOR AND THE CONTRIBUTION OF THE GENERAL
1602 REM BACKGROUND IN THE EXPRESSION FOR THE REL.STAND.OEV.
1603 A1=2*Q*B*P1
1610 A2=1-E(1,1)+E(1,1)*EXP(-Z(1,1))'EFFICIENCY FACTOR
1620 A4=0
1622 FOR X=0 TO M(1,1)-1
1630 A3=(Q-X)*EXP(-L(1,1))*(T2+X*R)
1640 A4=A4+A3
1650 NEXT X
1660 B(1,1)=A(1,1)*A4
1670 F(1,1)=B(1,1)*(1-EXP(-L(1,1)*T1))
1680 A5=F(1,1)*E(1,1)*(1-EXP(-Z(1,1)))
1686 RETURN
1690 REM SUBROUTINE TO CALCULATE THE CONTRIBUTION FROM THE
1691 REM VARIABLE BACKGR. IN THE EXPRESS. FOR THE REL.STAND.OEV.
1692 B8=0
1693 IF N>N(J) GO TO 1962
```

```
1695 FOR I3=N TO N(J)
1698 B6=0
1700 FOR I1=1 TO N(J)
1705 B2=0
1710 FOR I4=1 TO N(J)
1711 A7=0
1712 IF Q>=M(I4,J)-1 GO TO 1720
1713 M=Q
1714 GO TO 1721
1720 M=M(I4,J)-1
1721 FOR X=0 TO M
1730 A6=(Q-X)*EXP(-L(I4,J)*(T2+X*R))
1740 A7=A7+A6
1750 NEXT X
1755 A9=1
1760 FOR P3=1 TO N(J)
1765 IF N(J)=1 GO TO 1795
1770 IF P3=I4 GO TO 1800
1780 A8=ABS(1/(L(P3,J)-L(I4,J)))
1790 A9=A9*A8
1792 GO TO 1800
1795 A9=1
1800 NEXT P3
1810 B1=A7*A9*(1-EXP(-Z(I4,J)))
1820 B2=B2+B1
1830 NEXT I4
1840 B4=1
1845 IF I1=1 GO TO 1900
1850 FOR I2=1 TO I1-1
1860 B3=L(I2,J)
1870 B4=B4*B3
1880 NEXT I2
1900 B5=B4*B2
1910 B6=B6+B5
1920 NEXT I1
1930 B7=E(I3,J)*B6
1940 B8=B8+B7
1950 NEXT I3
1960 B9=2*B8*A(1,J)*(1-EXP(-X(1,J)))
1961 GO TO 1970
1962 B9=0
1970 RETURN
2000 REM SUBROUTINE FOR DETERMINATION OF THE COUNTING TIME
2001 REM FOR A STAND.DEV. <= TO THE MIN.REL.STAND.DEV.*10%
2005 FOR P5=1 TO P4-1
2010 E1=((D(1,P5)-D(1,P4-1))/D(1,P4-1))*100
2020 IF E1<=10 GO TO 2040
2030 NEXT P5
2040 RETURN
2050 REM SUBROUTINE FOR DETERMINATION OF THE IFR.TIME FOR
2060 REM A STAND.DEV. <= THE MIN.REL.STAND.DEV.*10%
2080 FOR P7=1 TO P6-1
2090 E2=((C(1,P7)-C(1,P6-1))/C(1,P6-1))*100
2100 IF E2<=10 GO TO 2120
2110 NEXT P7
2120 RETURN
2500 END
```

Special case.

As pointed out earlier, the program has to be slightly modified for calculation of optimum experimental times in the determination of oxygen in aluminium samples. The modifications are all carried out in the subroutine starting with statement no. 1690. This subroutine now takes the following form:

```
1690 REM SUBROUTINE TO CALCULATE THE CONTRIBUTION FROM THE
1691 REM VARIABLE BACKGR.IN THE EXPRESS.FOR THE REL.STAND.DEV.
1692 B8=0
1693 IF N>N(J) GO TO 1980
.
.
.
.
.
1925 IF J>1 GO TO 1935
1930 B7=E(I3,J)*B6
1932 GO TO 1940
1935 E7=B6
1940 B8=B8+B7
1950 NEXT I3
1952 B9=2*B8*A(1,J)*(1-EXP(-X(1,J,
1961 IF J<>2 GO TO 1970
1962 X1=B9*L(1,2)/(1-EXP(-Z(1,2)))
1963 E(1,2)=(1.327E-11)*X1
1964 B9=B9+E(1,2)
1966 GO TO 1995
1970 IF J=1 GO TO 1995
1972 E(1,3)=(1.43E-12)*X1
1974 B9=B9+E(1,3)
1976 GO TO 1995
1980 B9=0
1995 RETURN
```

All the statements between no. 1693 and 1925 are identical to those given at p.47 from no.1695 to 1920.

The special values that may be derived for the constants  $k_{12}$  and  $k_{13}$  (see page 31) must for each special counting arrangement be programmed into statement no.1963 and 1972 respectively. Note that the nuclide with index (1,2) is  $^{24}\text{Na}$ .

APPENDIX II

Table I. Optimum values of the counting time when only one primary formed variable background component is considered. The values are given as functions of the parameters  $C_{11}/C_{12}$ ,  $C_{11}/R_{GB}$  and  $\lambda_{12}/\lambda_{11}$ . Here C expresses the total number of counts obtainable by an "infinite" counting time from the number of radioactive nuclides present at the start of counting.  $C_{ij}$  is defined at p.22. The index (11) denotes the nuclide to be analyzed, and (12) the disturbing primary formed nuclide.  $\lambda$  is the normal decay constant and  $R_{GB}$  the general constant background count rate in counts per time unit used. Here the time unit is one half-life of the nuclide to be analyzed,  $(t_{\frac{1}{2}})_{11}$ . Accordingly, the results in the table are given in units of  $(t_{\frac{1}{2}})_{11}$ .

Table II. Optimum combinations of irradiation and counting times for some arbitrarily selected values of the above mentioned parameters (in units of  $(t_{\frac{1}{2}})_{11}$ ) when only one primary formed variable background component is considered.  $C_{ij}$  is defined at p.23.

Table III. Optimum combination of irradiation, decay and counting times for a few arbitrarily selected values of the above mentioned variable parameters (in units of  $(t_{\frac{1}{2}})_{11}$ ) when only one primary formed variable background component is considered.  $C_{ij}$  is defined at p.24.

Table I.

$C_{11}/C_{12}$	0.01	0.1	1.0	10	100	0.01	0.1	1.0	10	100
$\lambda_{12}/\lambda_{11}$	$C_{11}/R_{GB} = 4$					$C_{11}/R_{GB} = 10$				
0.0001	2.47	2.46	2.48	2.48	2.48	3.00	3.04	3.05	3.05	3.05
0.001	2.37	2.47	2.48	2.48	2.48	2.69	3.00	3.04	3.05	3.05
0.01	2.05	2.37	2.47	2.48	2.48	2.09	2.70	3.00	3.04	3.05
0.1	2.01	2.17	2.41	2.47	2.48	2.01	2.24	2.78	3.01	3.04
0.2	2.19	2.26	2.41	2.47	2.48	2.19	2.32	2.75	3.01	3.04
0.4	2.74	2.68	2.54	2.49	2.48	2.75	2.79	2.93	3.03	3.05
0.6	3.71	3.35	2.75	2.51	2.49	3.76	3.64	3.28	3.08	3.05
0.8	5.20	4.34	2.98	2.54	2.49	6.15	5.05	3.68	3.14	3.06
1.0	8.18	5.19	3.19	2.57	2.49	9.47	6.35	4.03	3.19	3.06
10.0	9.15	6.05	3.69	2.69	2.50	10	7.26	4.62	3.34	3.08
	$C_{11}/R_{GB} = 20$					$C_{11}/R_{GB} = 50$				
0.0001	3.51	3.62	3.63	3.63	3.63	4.26	4.54	4.58	4.58	4.58
0.001	2.92	3.51	3.62	3.63	3.63	3.14	4.26	4.54	4.58	4.58
0.01	2.10	2.94	3.52	3.62	3.63	2.12	3.16	4.27	4.55	4.58
0.1	2.01	2.27	3.07	3.55	3.62	2.01	2.29	3.36	4.35	4.56
0.2	2.20	2.34	2.98	3.52	3.62	2.20	2.36	3.20	4.28	4.55
0.4	2.75	2.83	3.20	3.56	3.63	2.76	2.86	3.47	4.35	4.56
0.6	3.77	3.76	3.69	3.64	3.63	3.78	3.84	4.16	4.51	4.57
0.8	6.31	5.53	4.29	3.73	3.64	6.42	6.03	5.14	4.67	4.59
1.0	10	7.28	4.79	3.81	3.65	10	8.55	5.91	4.79	4.60
10.0	10	8.21	5.43	3.98	3.67	10	9.50	6.59	4.98	4.63
	$C_{11}/R_{GB} = 75$					$C_{11}/R_{GB} = 100$				
0.0001	4.57	4.99	5.05	5.05	5.05	4.77	5.32	5.40	5.40	5.40
0.001	3.20	4.57	4.99	5.05	5.05	3.23	4.77	5.32	5.40	5.40
0.01	2.12	3.22	4.58	5.00	5.05	2.12	2.26	4.79	5.32	5.40
0.1	2.01	2.29	3.45	4.70	5.01	2.01	2.29	3.50	4.94	5.35
0.2	2.20	2.36	3.27	4.61	5.00	2.20	2.36	3.30	4.83	5.33
0.4	2.76	2.86	3.55	4.70	5.01	2.76	2.86	3.59	4.94	5.35
0.6	3.78	3.86	4.33	4.92	5.04	3.78	3.87	4.43	5.21	5.38
0.8	6.45	6.19	5.52	5.13	5.06	6.47	6.29	5.78	5.46	5.41
1.0	10	9.13	6.44	5.27	5.08	10	9.53	6.83	5.63	5.43
10.0	10	10	7.14	5.47	5.10	10	10	7.53	5.83	5.49
	$C_{11}/R_{GB} = 200$					$C_{11}/R_{GB} = 500$				
0.0001	5.19	6.12	6.27	6.28	6.29	5.55	7.12	7.47	7.52	7.52
0.001	3.29	5.19	6.12	6.27	6.29	3.32	5.55	7.12	7.48	7.52
0.01	2.12	3.31	5.22	6.13	6.27	2.12	3.35	5.59	7.13	7.48
0.1	2.01	2.30	3.58	5.46	6.18	2.01	2.30	3.63	5.96	7.27
0.2	2.20	2.37	3.36	5.30	6.15	2.20	2.37	3.40	5.74	7.23
0.4	2.76	2.87	3.66	5.49	6.19	2.76	2.87	3.71	6.06	7.33
0.6	3.79	3.89	4.60	5.92	6.25	3.79	3.90	4.74	6.80	7.44
0.8	6.49	6.46	6.37	6.30	6.29	6.50	6.59	7.04	7.45	7.51
1.0	10	10	7.77	6.53	6.31	10	10	9.06	7.77	7.55
10.0	10	10	8.49	6.74	6.34	10	10	9.78	7.99	7.58

Table II.

$C_{11}/C_{12}$		100		10		1		0.6		0.2		0.1		0.05		0.01		0.005		0.001		
$C_{11}/R_{DB}$		$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	
$\lambda_{12}/\lambda_{11} = 0.01$																						
250	opt	10	6.6			10	6.4	9.7	6.4	8.2	6.1	7.3	5.8	6.4	5.4	4.6	4.3	4.0	3.8	2.8	2.7	
	+10%	2.6	6.3			2.6	6.3	2.6	6.3	2.5	6.2	2.5	6.0	2.4	5.7	2.0	4.7	1.8	4.2	1.4	3.0	
100	opt	10	5.4							8.5	5.2	7.6	5.0	6.7	4.8	4.8	4.0	4.1	3.6	2.9	2.7	
	+10%	2.7	5.2							2.6	5.1	2.5	5.0	2.4	4.9	2.1	4.3	1.9	3.9	1.4	2.9	
50	opt	10	4.6										7.9	4.4	7.0	4.3	5.0	3.8	4.3	3.4	3.0	2.7
	+10%	2.8	4.4										2.6	4.3	2.5	4.2	2.2	3.9	2.0	3.6	1.5	2.8
20	opt	10	3.6										6.5	3.6	7.6	3.5	5.5	3.3	4.7	3.1	3.2	2.5
	+10%	2.9	3.5										2.8	3.5	2.7	3.4	2.4	3.3	2.2	3.1	1.6	2.6
10	opt	10	3.0										9.1	3.0	8.1	3.0	6.0	2.9	5.1	2.8	3.5	2.4
	+10%	3.0	3.0										3.0	2.9	2.9	2.9	2.6	2.8	2.4	2.6	1.8	2.5
2	opt	10	2.2												9.9	2.2	7.6	2.2	6.6	2.2	4.7	2.1
	+10%	3.3	2.2												3.3	2.2	3.1	2.2	2.9	2.1	2.4	2.1

$C_{11}/C_{12}$		100		10		4		1		0.4		0.1		0.05		0.01	
$C_{11}/R_{DB}$		$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$	$t_i$	$t_c$
$\lambda_{12}/\lambda_{11} = 0.1$																	
250	opt	10	6.5	8.2	6.1	6.9	5.6	5.2	4.7	4.2	3.9	3.3	3.3	3.3	3.2	3.2	3.2
	+10%	2.6	6.3	2.5	6.1	2.4	5.8	2.1	5.1	1.8	4.3	1.4	3.3	1.4	3.3	1.4	3.2
100	opt	10	5.4	8.5	5.2	7.2	4.9	5.4	4.3	4.3	3.8	3.4	3.3	3.3	3.2	3.2	3.2
	+10%	2.7	5.2	2.6	5.1	2.5	5.0	2.2	4.5	1.9	4.0	1.4	3.3	1.4	3.3	1.4	3.3
50	opt	10	3.6	9.5	3.6	8.1	3.5	6.1	3.4	4.9	3.2	3.3	3.3	3.3	3.2	3.2	3.2
	+10%	2.8	4.4	2.7	4.4	2.6	4.3	2.3	4.0	2.0	3.7	1.5	3.3	1.4	3.2	1.4	3.2
20	opt	10	3.6	9.5	3.6	8.1	3.5	6.1	3.4	4.9	3.2	3.5	3.2	3.3	3.2	3.2	3.2
	+10%	2.9	3.5	2.8	3.5	2.8	3.5	2.5	3.3	2.2	3.2	1.5	3.2	1.4	3.2	1.4	3.2
10	opt	10	3.0	10	3.0	8.7	3.0	6.7	2.9	5.4	2.8	3.6	3.1	3.4	3.1	3.2	3.2
	+10%	3.0	3.0	3.0	2.9	2.9	2.9	2.7	2.9	2.4	2.6	1.6	3.1	1.5	3.1	1.4	3.2
2	opt	10	2.2	10	2.2	10	2.2	8.4	2.2	7.1	2.2	4.2	2.7	3.8	2.9	1.5	3.0
	+10%	3.3	2.2	3.3	2.2	3.3	2.2	3.1	2.2	3.0	2.2	1.9	2.6	1.7	2.8	1.5	3.0
$\lambda_{12}/\lambda_{11} = 0.5$																	
250	opt	10	6.5	8.0	5.9	6.2	5.3	4.4	4.2	3.8	3.7	3.1	3.0	2.7	2.6	2.2	2.2
	+10%	2.6	6.3	2.4	5.9	2.2	5.4	1.8	4.3	1.6	3.8	1.5	3.2	1.3	3.0	1.2	2.2
100	opt	10	5.4	8.3	5.1	6.4	4.8	4.5	4.0	3.8	3.6	3.2	2.9	2.7	2.6	2.2	2.2
	+10%	2.7	5.2	2.5	5.0	2.2	4.7	1.8	4.1	1.6	3.7	1.5	3.1	1.2	2.8	1.2	2.2
50	opt	10	4.6	8.7	4.4	6.7	4.3	4.7	3.8	3.9	3.5	3.3	2.9	2.8	2.6	2.2	2.2
	+10%	2.8	4.4	2.6	4.3	2.3	4.1	1.9	3.8	1.8	3.2	1.6	3.0	1.4	2.7	1.2	2.2
20	opt	10	3.6	9.5	3.6	7.4	3.6	5.0	3.4	4.1	3.3	3.5	2.7	3.0	2.5	2.3	2.1
	+10%	2.9	3.5	2.7	3.5	2.5	3.4	2.1	3.3	1.8	3.2	1.7	2.8	1.5	2.6	1.2	2.2
10	opt	10	3.0	10	3.1	8.1	3.1	5.5	3.1	4.4	3.1	3.8	2.6	3.2	2.4	2.4	2.1
	+10%	3.0	3.0	2.9	3.0	2.7	2.9	2.2	2.9	1.9	3.0	1.9	2.6	1.7	2.4	1.2	2.1
2	opt	10	2.2	10	2.2	10	2.2	7.5	2.3	5.9	2.5	5.2	2.1	4.3	2.1	3.0	2.0
	+10%	3.3	2.2	3.2	2.2	3.2	2.2	2.8	2.3	2.5	2.4	2.5	2.1	2.2	2.1	1.6	2.0

Table III.

$C_{11}/C_{12}$	0.01			0.05			0.1			0.5			1			5			100				
	$\lambda_{12}/\lambda_{11}$	$t_i$	$t_d$	$t_c$	$t_i$	$t_d$	$t_c$	$t_i$	$t_d$	$t_c$	$t_i$	$t_d$	$t_c$	$t_i$	$t_d$	$t_c$	$t_i$	$t_d$	$t_c$	$t_i$	$t_d$	$t_c$	
$C_{11}/R_{GB} = 5$																							
100	3.2	0.13	2.5	3.2	0.11	2.5	3.2	0.10	2.5	3.2	0.07	2.6	3.2	0.06	2.6	3.2	0.04	2.6	<0.01				
20	3.3	0.52	2.6	3.3	0.41	2.6	3.3	0.36	2.6	3.3	0.24	2.7	3.3	0.19	2.7	3.3	0.07	2.7	<0.01				
10	3.4	0.93	2.7	3.4	0.67	2.8	3.3	0.59	2.8	3.3	0.36	2.8	3.3	0.25	2.9	3.3	0.01	3.0	<0.01				
5	3.4	1.55	3.2	3.4	1.07	3.3	3.4	0.87	3.3	3.4	0.38	3.4	3.4	0.17	3.4	<0.01	<0.01	<0.01	<0.01				
3.3	3.5	1.89	4.0	3.5	1.16	4.0	3.4	0.86	4.1	3.4	0.13	4.2	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01				
2.5	3.5	1.80	5.3	3.5	0.83	5.4	3.5	0.42	5.4	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01				
$C_{11}/R_{GB} = 10$																							
100	3.1	0.14	2.9	3.1	0.11	2.9	3.1	0.10	2.9	3.1	0.08	2.9	3.1	0.07	3.0	3.1	0.04	3.0	<0.01				
20	3.2	0.56	2.9	3.2	0.44	2.9	3.2	0.39	3.0	3.2	0.27	3.0	3.2	0.22	3.0	3.2	0.10	3.1	<0.01				
10	3.3	1.01	2.9	3.3	0.77	3.0	3.3	0.66	3.1	3.2	0.42	3.2	3.2	0.32	3.2	3.2	0.07	3.3	<0.01				
5	3.4	1.73	3.3	3.4	1.24	3.4	3.4	1.03	3.5	3.3	0.54	3.6	3.3	0.32	3.7	<0.01	<0.01	<0.01	<0.01				
3.3	3.5	2.18	4.0	3.4	1.45	4.1	3.4	1.13	4.2	3.4	0.37	4.4	3.4	0.04	4.5	<0.01	<0.01	<0.01	<0.01				
2.5	3.5	2.18	5.3	3.5	1.20	5.5	3.5	0.55	6.0	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01				
$C_{11}/R_{GB} = 20$																							
100	3.0	0.14	3.5	3.0	0.12	3.5	3.0	0.11	3.5	3.0	0.08	3.5	3.0	0.07	3.5	3.0	0.05	3.5	3.0	0.01	3.5		
20	3.1	0.59	3.3	3.1	0.47	3.4	3.1	0.41	3.4	3.1	0.29	3.5	3.1	0.24	3.5	3.0	0.12	3.6	<0.01				
10	3.2	1.07	3.3	3.2	0.83	3.4	3.2	0.72	3.5	3.1	0.48	3.6	3.1	0.37	3.7	3.1	0.13	3.8	<0.01				
5	3.4	1.83	3.7	3.3	1.39	3.7	3.3	1.17	3.8	3.3	0.62	4.1	3.2	0.44	4.1	<0.01	<0.01	<0.01	<0.01				
3.3	3.4	2.44	4.1	3.4	1.69	4.3	3.4	1.37	4.4	3.4	0.43	5.0	3.3	0.24	4.8	<0.01	<0.01	<0.01	<0.01				
2.5	3.5	2.55	5.4	3.4	1.55	5.6	3.4	1.11	5.7	3.4	0.04	6.1	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01				
$C_{11}/R_{GB} = 50$																							
100	2.9	0.14	4.4	2.9	0.11	4.4	2.9	0.10	4.4	2.9	0.08	4.4	2.9	0.08	4.5	2.9	0.04	4.5	2.8	0.01	4.4		
20	3.0	0.59	4.2	3.0	0.46	4.3	3.0	0.40	4.4	3.0	0.26	4.5	3.0	0.21	4.6	2.9	0.15	4.4	<0.01				
10	3.1	1.14	4.0	3.0	0.89	4.1	3.0	0.76	4.2	3.0	0.53	4.4	3.0	0.43	4.4	3.0	0.18	4.6	<0.01				
5	3.3	2.06	4.0	3.2	1.55	4.2	3.2	1.33	4.3	3.2	0.75	4.7	3.1	0.58	4.7	3.1	0.04	5.1	<0.01				
3.3	3.4	2.76	4.4	3.4	1.87	4.9	3.3	1.65	4.6	3.3	0.83	5.2	3.3	0.37	5.6	<0.01	<0.01	<0.01	<0.01				
2.5	3.4	3.02	5.6	3.4	1.59	5.6	3.4	1.52	6.0	3.4	0.37	6.0	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01				
$C_{11}/R_{GB} = 100$																							
100	2.8	0.15	5.1	2.8	0.12	5.1	2.8	0.11	5.2	2.8	0.09	5.2	2.8	0.08	5.2	2.8	0.06	5.2	2.8	0.01	5.2		
20	2.9	0.62	4.9	2.9	0.48	5.0	2.9	0.42	5.1	2.9	0.29	5.2	2.9	0.24	5.2	2.8	0.15	5.2	<0.01				
10	3.0	1.17	4.6	3.0	0.89	4.9	3.0	0.76	5.0	2.9	0.57	5.0	2.9	0.46	5.1	2.9	0.21	5.3	<0.01				
5	3.2	2.16	4.4	3.1	1.65	4.7	3.1	1.42	4.9	3.1	0.88	5.2	3.1	0.61	5.5	3.0	0.10	5.8	<0.01				
3.3	3.4	2.87	4.9	3.3	2.18	5.0	3.3	1.82	5.2	3.2	0.97	5.8	3.2	0.59	6.0	<0.01	<0.01	<0.01	<0.01				
2.5	3.4	3.36	5.7	3.4	2.28	6.1	3.4	1.79	6.3	3.3	0.58	7.1	3.3	0.02	7.5	<0.01	<0.01	<0.01	<0.01				
$C_{11}/R_{GB} = 200$																							
100	2.7	0.15	6.0	2.7	0.12	6.0	2.7	0.11	6.0	2.7	0.09	6.0	2.7	0.08	6.0	2.7	0.06	6.0	2.7	0.01	6.1		
20	2.9	0.56	5.9	2.8	0.52	5.7	2.8	0.46	5.8	2.8	0.34	5.9	2.8	0.29	6.0	2.8	0.16	6.1	<0.01				
10	2.9	1.20	5.3	2.9	0.95	5.5	2.8	0.83	5.7	2.8	0.59	5.8	2.8	0.48	5.9	2.9	0.17	6.3	<0.01				
5	3.1	2.26	5.0	3.1	1.70	5.4	3.1	1.43	5.7	3.0	0.94	5.9	3.0	0.71	6.1	3.0	0.14	6.6	<0.01				
3.3	3.3	3.16	5.0	3.3	2.21	5.8	3.2	1.96	5.8	3.2	1.07	6.4	3.2	0.61	6.9	<0.01	<0.01	<0.01	<0.01				
2.5	3.4	3.68	5.9	3.4	2.54	6.5	3.3	2.02	6.8	3.3	0.74	7.7	3.3	0.15	8.1	<0.01	<0.01	<0.01	<0.01				
$C_{11}/R_{GB} = 500$																							
100	2.7	0.14	7.2	2.7	0.11	7.3	2.7	0.10	7.3	2.7	0.08	7.3	2.7	0.07	7.3	2.7	0.04	7.3	2.6	0.02	7.3		
20	2.8	0.60	7.0	2.8	0.47	7.1	2.8	0.42	7.1	2.7	0.35	7.1	2.8	0.24	7.3	2.8	0.11	7.5	<0.01				
10	2.9	1.15	6.7	2.8	0.87	6.7	2.8	0.86	6.8	2.8	0.61	7.0	2.8	0.50	7.1	2.9	0.13	7.7	<0.01				
5	3.0	2.35	5.9	3.0	1.80	6.3	3.0	1.56	6.5	3.0	0.95	7.1	2.9	0.75	7.2	2.9	0.19	7.7	<0.01				
3.3	3.2	3.38	5.7	3.2	2.49	6.3	3.2	2.04	6.8	3.2	1.00	7.8	3.1	0.77	7.8	<0.01	<0.01	<0.01	<0.01				
2.5	3.4	4.03	6.4	3.4	2.52	7.7	3.4	1.83	8.4	3.3	0.88	8.6	3.3	0.26	9.2	<0.01	<0.01	<0.01	<0.01				
2.1	3.4	3.43	9.0	3.4	1.85	10	3.4	1.10	10	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01				
2.01	3.4	1.10	10	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01			