

# SIMPLE PROCEDURES FOR DETERMINING THE AGE OF FISSION PRODUCTS

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## Summary

Rapid and simple age determination procedures important for civil defense were developed. The model substances for the explosion products used in experimental work were  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  fission products. The sensor was a Si(Li) semiconductor detector. The procedure based on the quotient of overall pulses measured above two different discrimination levels yielded satisfactory accuracy of age determination in the interval of 20 to 600 hours. The procedure based on continuous observation of dose rate decrease is serviceable in the interval of at least 2 to 1000 hours, but presumable longer on. The two portable instruments built at Gamma Works, Budapest allow to perform measurements on site.

## Introduction

Besides peaceful utilization of nuclear energy, our age unfortunately also has to reckon with the possibility that nuclear weapons will be put into action. The peace movement increasingly gaining strength in our days expresses the pronounced wish of humanity, when — to avoid world-wide cataclysm — it fights for blocking manufacture and deployment of nuclear weapons. One must, however, adopt a realistic view in the question that the vast amounts of nuclear weapons accumulated at this stage in the world necessarily require the manifold organization of the defense of the population, since such measures largely increase the probability of survival in the case of a nuclear catastrophe.

Within the frame of civil defense, an important group of preventive measures consists in the development of measuring techniques and instruments serving reliable estimation of the extent of contamination of the stricken areas and objects.

One of the important parameters of estimation is age of the fission products, which — as a first approach — can be identified with the time passed since the nuclear fission. In nuclear measurements, techniques serving to determine the age of a substance by measuring its  $^{14}\text{C}$  content are widely known. It is equally known that from the activity ratio of the parent element

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and the daughter element one can determine the time passed since the moment when the parent element was formed.

In this paper we report on a relatively new domain of age determination: a simple method will be presented serving to determine the ages of fission products originating from a nuclear catastrophe.

It is known that the isotope composition of fission products will change with time, nor can the rate of activity decrease and dose rate decrease be characterized with the usually applied half-life. Hence, to estimate both internal and external dose load it is indispensable to know the age of the fission products. In the general case this has to be determined by measurement

### The significance of age

The share of the residual radioactive products in the energy released in a nuclear explosion amounts to some percents. The products form particles varying in composition and size, which fall out from the radioactive cloud and contaminate the ground. The significance of fall-out consists in its damaging effect on living organisms, which it exerts continuously, over long periods, on large areas.

Regarding its origin, fall-out consists of the following components:

- residual fuel that did not suffer fission,
- fission products, and
- activated environmental substances.

The share of these components in total activity depends on the type of the explosion (one-, two- or three-phase bomb), on the share of energies released in nuclear fission and nuclear fusion, resp., on the mode of explosion (ground, underground, air, underwater) and on the time passed since the explosion, that is, on age. (The energy of the one-phase bomb is supplied by nuclear fission of  $^{233}\text{U}$ ,  $^{235}\text{U}$  or  $^{239}\text{Pu}$ , while in the two-phase bomb (hydrogen bomb) the neutrons emitted in the fission reaction and the heat energy evolved are utilized to activate the fusion reaction of tritium as well. The three-phase bomb has a cover containing natural uranium, and fission of  $^{238}\text{U}$  is achieved by 14 MeV neutrons set free in the fusion reaction.)

All authors [1–7] agree that the predominant share of fall-out activity — independently of other circumstances — is supplied by fission products. For this reason, its biological danger is defined by the average dosimetric properties of the fission products.

Regarding external dose load, this is expressed by the empiric Way-Wigner equation [8] describing the decrease of the dose rate of the fission products:

$$P_i = P_0 \left[ \frac{t_i}{t_0} \right]^{-n} \quad (1)$$

where  $P_i$  is the actual value of dose rate,  $P_0$  is the initial value of dose rate (usually related to the first hour after the explosion),  $t_i$  is the time passed since the explosion, i.e. age, and  $t_0$  is the initial value of the time passed since the explosion, which in generally accepted practice is  $t_0 = 1$  hr, and  $n$  is an empirical value.

In the knowledge of the parameters and the independent variables, the definite integral of Eq. (1) over the period between entrance and egress will yield the dose absorbed during the period in question.

Regarding internal dose load, the amount of fall-out entering the organism with inhaled air, food and drink is expressed in terms of maximum permissible incorporated activity [1, 9]. The theoretically admissible value decreases with age, owing to dramatic initial activity decrease and accumulation of highly radiotoxic isotopes of long half-life. The legally admitted limit values approach it stepwise from below, using suitable safety factors.

Consequently, age is of importance, because it is one of the basic parameters for estimating both external and internal dose load. Since, however, composition of fall-out — owing to the multiplicity of explosion conditions — may differ largely from average, and since in the case when fall-out originating from explosions carried out at different moments mixes the knowledge of average or resultant age defined by mixing ratio and time between explosions is needed, age in dosimetric evaluation cannot be identified with the time passed since the explosion, and its value can — in the general case — only be determined reliably by measurement.

This statement appears contradictory in the case when the moment of explosion is known. The contradiction is lifted by Eq. (1), since we are much more interested in the dose absorbed than in dose rate itself, and the dose absorbed is obtained by the integral of the equation. Integration will, however, yield a generally utilizable expression only if the value of the exponent is accepted as known and constant in time. In the literature one meets most frequently with the value  $n = 1.2$ , and simple evaluation tables used in practice are also based on this value. However, such standard fall-out will hardly occur in reality, due to the differing conditions mentioned above. For this reason, accepting a fixed value for  $n$ , the dose absorbed, that is, the area below the curve expressed by Eq. (1) is best approached in the manner that the slope of the curve is determined experimentally for the actual case and compared with the standard curve of standard fall-out; one may then accept the value  $t$  corresponding to the point of the standard curve in which slope is identical with that of the experimental curve as the resultant age of the product in question. This procedure appears rather sophisticated at first sight; however, it has the advantage that the simple estimation methods developed for the standard fall-out are applicable with greater reliability in possession of the factual measurement results.

If the above is complemented by the possibility that fall-out may well be a mixed product originating from several explosions carried out in different moments, whose resultant activity decrease curve will follow from the weighed summation of the curves of the two generating products, it is evident that in the general case age cannot be identified with the time passed since the explosion. It follows from what has been said that the age of the fall-out — together with dose rate — should be measured at the given site and in the given moment, with the simplest possible procedure and instrument, in view of the decisive importance of time of action. The fundamental objective of our work was therefore to simplify the procedure and means of age determination to the greatest possible extent.

### Known procedures

#### *Measurement of activity decrease*

The oldest procedure of age determination which still should be considered simplest of all is based on the Way–Wigner equation (Eq. 1). The rate of dose rate decrease is provided by the derivative of Eq. (1). In practice this is determined usually by two measurements taken at different moments, with the value of the exponent accepted as known and constant. The age of the product in the moment of the second measurement is then expressed by the following equation:

$$t = \frac{P_1^{1/n}}{P_1^{1/n} - P_2^{1/n}} \Delta t \quad (2)$$

where  $\Delta t$  is the period of time between the two measurements.

The great advantage of this procedure is simplicity; its disadvantage is that it requires two measurements at different moments of time. To obtain a result that could be evaluated significantly, a rather long period of time may be necessary in the case of older products. However, only the second measurement will give information on the age of the product.

#### *$\beta$ -absorption procedure*

Soviet authors developed a very simple procedure for age determination, based on the  $\beta$ -spectra varying in time of explosion products [1]. Its essence consists in the followings: they constructed a graph for the explosion products, based on the absorption coefficients characterizing the given energy level at different moments after the explosion (reported e.g. by Nelms and Cooper [6])

for the total  $\beta$ -spectrum divided into 22 energy intervals and percentage composition. They calculated how the ratio of counts measured without absorbent and with a  $260 \text{ mg/cm}^2$  interposed aluminium layer, that is, the attenuation coefficient of the arrangement changes with time. Calculations demonstrated that the attenuation coefficient *versus* time relationship has a maximum on the sixth day. Thus, to each of the values two ages can be assigned (except, of course, to the maximum), and therefore one can decide only by repeated measurement whether the age of the product in question is located on the ascendent or descendent portion of the curve.

### $\gamma$ -spectrometric procedures

These procedures determine the composition of the fall-out by analysis of its  $\gamma$ -spectrum, and compare it with calculated or measured spectra of products with known ages. In this manner one can conclude on age by means of isotope composition, or rather by the presence and share of some characteristic isotopes.

Detailed data on changes in  $\gamma$ - and  $\beta$ -spectral composition of the fission products of uranium-235 were reported by Nelms and Cooper [6], Dolan [10] and Miller [11]. Their data are cited in the comprehensive paper by Cook [12], who compared his own experimental results with the mean  $\gamma$ -energy per photon of uranium fission products computed by Miller [11] (Fig. 1).

It is interesting to observe in the figure that the measured mean energy values of the fission products of  $^{238}\text{U}$  deviate from the calculated curve in the negative direction. This is particularly distinct around the 100-th hour. The

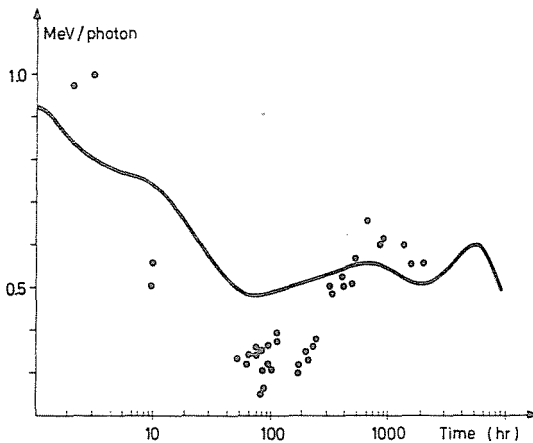


Fig. 1. Mean gamma energy/photon values vs. time for  $^{235}\text{U}$ , determined experimentally by Cook [12] and computed by Miller [11]

finding is explained by the nuclear reaction  $^{238}\text{U}(n, \gamma)^{239}\text{U}$ . One part of the natural uranium jacket of the three-phase bomb will subsequently, by  $\beta$ -disintegration, produce the isotope  $^{239}\text{Np}$  with a half-life of 2.33 days; its soft  $\gamma$ -lines will substantially reduce the mean  $\gamma$ -energy of the product.

$\gamma$ -spectrum analyses were first performed using a NaI(Tl) scintillator and an amplitude analyzer. Several techniques limited to the quantitative

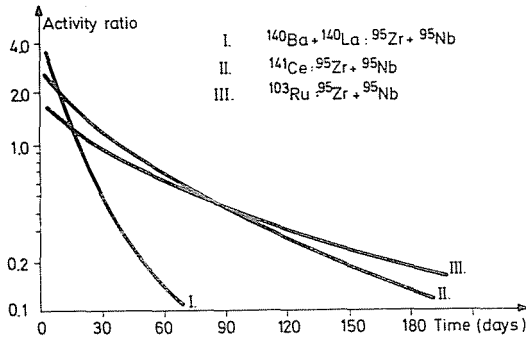


Fig. 2. Age determination by activity ratios of isotopes (Svensson and Lidén [13])

determination of a few characteristic radioactive isotopes only were developed. For instance Svensson and Lidén [13] determined the time passed since the explosion from the activity ratio of  $^{103}\text{Ru}$ ,  $^{141}\text{Ce}$  and  $^{140}\text{Ba} + ^{140}\text{La}$ , resp., to  $^{95}\text{Zr} + ^{95}\text{Nb}$  (Fig. 2).

The curves in the figure indicate that the procedure is suited for age determination within maximum about 180 days. The results published demonstrate that age determination of the explosion products within the interval of 23 to 170 days is possible with an accuracy of  $\pm 23$ –26%. Using high-resolution semiconductor detectors, isotope composition can be studied with still higher accuracy.

However, the fundamental inadequacies of these methods should also be mentioned. Already Cook [12] noted that the true composition of the explosion products may differ significantly from the calculated data. Later [14] it was stated that the difference is due above all to the phenomenon termed fractionation: at the high temperature accompanying explosion, part of the fission products — particularly volatile elements like bromine, iodine, crypton, xenon — will evaporate and will not be incorporated in the condensed fall-out particles. Hence, these isotopes and their disintegration chain will partly or wholly be absent in fall-out.

Fractionation was studied above all by Japanese authors. Mamuro and co-workers [16–18] gave the following expression for the fractionation coefficient:

$$f(i) = \left[ \frac{A(i)}{A(^{95}\text{Zr} + ^{95}\text{Nb})} \right]_{\text{exp.}} \left[ \frac{A(^{95}\text{Zr} + ^{95}\text{Nb})}{A(i)} \right]_{\text{theor.}}$$

where  $A(i)$  is the activity of the nuclide in question, and  $A(^{95}\text{Zr} + ^{95}\text{Nb})$  is the joint activity of these two isotopes.

Their results display that the isotope composition of fall-out varies significantly with explosion conditions. The two extreme values for  $^{90}\text{Sr}$  may, e.g. vary by a factor of 1200, and hence this phenomenon may cause large errors in age determination based on isotope composition.

Procedures which — within a chosen decay chain — investigate activity of a single isotope or overall activity of the radioactive chain *versus* time are exempt of the error caused by fractionation. Since activity ratios within a given decay chain are strictly defined by the known half-lives of the parent element and the daughter elements, eventual fractionation of the first member of the chain will interfere with measurement to a far lower degree (insofar, of course, its activity is above the measurable level).

Silantev [15] measured the activity of the second member of the disintegration chain  $^{140}\text{Ba} + ^{140}\text{La}$  at two different moments and calculated age from the extent of activity change. The advantage of this method is that the 1.6 MeV photopeak of  $^{140}\text{La}$  in its  $\gamma$ -spectrum is separable even if a scintillation detector is being used. Its disadvantage is that two measurements must be made at different moments. It follows from half-lives that the procedure can only be used successfully till ages of about 2 months.

Within the first period of about 100 days, the age of the fission products can be determined with relatively high accuracy from the  $\gamma$ -spectrum obtained with a Ge(Li) semiconductor detector by calculating the activity quotient  $^{95}\text{Zr}/^{95}\text{Nb}$  [19].  $^{95}\text{Nb}$  is not formed as a primary product of nuclear fission, but only in the decay chain, as daughter element of  $^{95}\text{Zr}$ . In this manner one could, on principle, determine age till transient equilibrium is established. However, the quotient curve, after the 200th day, flattens to such an extent that no estimation of age is possible.

The greatest advantage of the procedure is that one single measurement will yield age. It can, however, only be applied in laboratories and requires expensive equipment. For civil defense purposes, one needs rapid and simple procedures that can be carried out under rough conditions, preferably with a portable instrument. The more recent procedures to be discussed in the followings were developed to satisfy these demands.

## Recent procedures for age determination

### *Age determination based on integral spectra quotients of fission products*

This procedure was born from the recognition [20] that if, on the one hand, activity and dose rate decrease of the fall-out within some defined period is describable by the Way–Wigner equation, and on the other hand, the disintegration of individual isotopes or, in the case of daughter elements, their concentration increase will be reflected in the decrease or relative increase of the corresponding photopeaks in the  $\gamma$ -spectrum, at least two threshold values must exist within the  $\gamma$ -energy interval studied, above which integral  $\gamma$ -spectra will change with time according to the Way–Wigner equation, with the values of the exponents differing in the following manner: if the intensities of the integral spectrum are

$$I_1 = I_{0.1} t_i^{-n_1} \quad (3)$$

$$I_2 = I_{0.2} t_i^{-n_2}$$

and their quotient function  $H_{1.2}$  is

$$H_{1.2} = \frac{I_{0.1}}{I_{0.2}} t_i^{-(n_1 - n_2)} \quad (4)$$

or, by using simpler symbols

$$H_{1.2} = (H_0)_{1.2} i^{-n_{1.2}} \quad (5)$$

this function will have a slope steep enough to allow estimation of the age  $t_i$ ,  $H_{1.2}$  being the actual value of the quotient of the integral spectra measured above the two threshold energy values in the moment  $t_i$ ,  $(H_0)_{1.2}$  is the initial value of this quotient (preferably related to one hour after the explosion),  $t_i$  is age, that is, the time passed since the explosion, and  $n_{1.2}$  an empirical value. (In all equations, time is considered in the sense that  $t_0 = 1$  hr.)

In Eq. (5), the parameters  $(H_0)_{1.2}$  and  $n_{1.2}$  are values characteristic for the given experimental arrangement and equipment. Their determination is equivalent to calibration of the system. For this purpose, a series of measurements is performed in different moments after fission with a standard fission product at a fixed geometrical arrangement, with some energy-selective detector, at two different discrimination settings, and the above two parameters are determined from the results by some curve fitting technique, e.g. by the method of least squares.



With the calibrated equipment, the unknown age of fission products is obtained, after readjustment of Eq. (5), by the following expression:

$$t_i = \left[ \frac{(H_0)_{1.2}}{H_{1.2}} \right]^{1/n_{1.2}} \quad (6)$$

where the symbols are identical with those in Eq. (5).

Since the parameters  $(H_0)_{1.2}$  and  $n_{1.2}$  are known from calibration and constant for the given equipment, age determination is traced back in Eq. (6) to the quotient  $H_{1.2}$  of the overall intensities  $I_1$  and  $I_2$  of the integral spectrum measured at one and the same moment above two different  $\gamma$ -energy values, namely the two discrimination thresholds.

The physical meaning of the procedures will be easy to understand by the following example. By selecting the two discrimination levels in a manner to coincide with the two base points of a photopeak changing with time, the curves expressed by Eq. (3) (straight lines in log-log plots) will diverge at a relative increase of the photopeak, and converge at its decrease. In the first case the exponent in the quotient function Eq. (6) will be positive, and in the second case it will be negative.

To select the preferential energy levels is, of course, no simple task, since the behaviour of photopeaks occurring in the spectra of fission products, usually of a complex nature, is very difficult to anticipate. Experimental investigations therefore have the purpose to find the optimum quotient function (5), that is, the function with the steepest slope for the given equipment. In a later chapter we shall present the course of choosing the function.

*Age determination based on continuous measurement  
of dose rate decrease*

The preceding method is primarily suited for age determinations in various samples prepared from materials contaminated with fall-out (e.g. foods, water, etc.). It is, however, unsuited for direct age determination from dose rates in the open ground, since  $\gamma$ -spectra will be largely distorted due to scattering. Such determinations are, however, indispensable for estimating dose rates originating from external irradiation. In the followings, the procedure developed for this purpose [20] will be presented.

We have seen in the foregoings that age can be calculated by Eq. (2) derived from the Way-Wigner equation by means of measuring dose rates at two different moments with equipment identical in its geometrical arrangement.

Let us now reverse the order of the investigation. Let the quotient of the dose rates  $K_p = P_2/P_1$  be a preset value. Then, at a constant value of the

exponent  $n$ , the term

$$\left(\frac{P_2}{P_1}\right)^{1/n} = K_1 \quad (7)$$

will also be constant and Eq. (2) can be written in the form

$$t_2 = \frac{\Delta t}{1 - K_1} \quad (8)$$

or, reducing the constants,

$$K_2 = \frac{1}{1 - K_1} \quad (9)$$

and

$$t_2 = K_2 \Delta t = t_i. \quad (10)$$

Thus, age  $t_i$  can be determined by a linear equation readily solved electrically, so that instrumental implementation will be largely simplified.

Based on the above, age is determined as follows: the dose rate of the radioactive fission products decreasing monotonously by the known law is measured continuously (or, instead of dose rate, intensity in the same experimental arrangement), and the period  $\Delta t$  is observed after which the dose rate at the start of the measurement  $P_1$  decreases by the preset factor  $K_p$ . The value of  $K_p$  is preferably chosen between 0.5 and 0.95. Age is obtained by multiplying  $\Delta t$  with a constant proportionality factor  $K_2$  (cf. Eq. (9)) according to Eq. (10).

The objective of experimental study is to determine the actual value of the exponent  $n$  valid in the system represented by the model material, radiation detector, electronics and measurement arrangement, and to find the conditions under which the value of the exponent may be considered constant within the time interval of interpretation.

#### *Forecasting of the integral dose during total decay time*

The preceding method is of increased importance for practice, since it can readily be utilized to forecast the integral dose during the total decay period, i.e. the dosimetric parameter derived from measured dose rate and age.

Integral dose forecasts are of significance in determining the advisable mode of conduct for inhabitants (e.g. the population) in the area. If a single measurement allows to determine the dose to which an individual in open location will be subjected during total decay time (which, for fall-out, may be estimated to one year in practice), authorities may immediately take decisions on total or partial evacuation (e.g. women, children only), on the proportion of

time to be spent daily in shelters and in free location, resp. Measurement of dose rate and age provide realistic foundations for such measures.

For further simplification let us assume in the followings that the exponent in Eq. (1) is constant and equal to 1.2. This is the mean value generally accepted in the literature [1-3, 9, 14]. The dose  $D_{1,2}$  suffered from the entrance moment  $t_1$  to the exit moment  $t_2$  will then be obtained by the definite integral of Eq. (1):

$$D_{1,2} = 5P_0(t_1^{-0.2} - t_2^{-0.2}). \quad (11)$$

Hence the dose suffered during total decay time, if  $t_2 = \infty$ , is

$$D_{1,\infty} = 5Pt. \quad (12)$$

In this expression we have only the actual measured values of  $P$  and  $t$  (in the last step we substituted the value  $P_0$  from Eq. (1)).

If  $n \neq 1.2$ , the generalized form of Eq. (12) is

$$D_{1,\infty} = \frac{Pt}{n-1} \quad (13)$$

This function must hence be implemented by the given equipment. Determination of the age  $t$  is performed by means of the linear equation (10).

A further precondition for simple instrumental implementation is that the product  $Pt$  should be replaceable by some other parameter that can be measured in a simple way, since otherwise the extremely high dynamic demand to the multiplier loop will render practical implementation impossible (unless microprocessors are utilized).

One can prove mathematically that Eq. (12) can be written in the following form:

$$D_{1,\infty} = K_0 D_{1,2} \quad (14)$$

where the value of the constant  $K_0$  is defined by  $K_1$  (cf. Eq. (7)), that is, by the selected values of the exponent  $n$  and dose rate decrease, resp., and by the unit applied.

### Experimental

The model materials for explosion products were fission products of  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  prepared by irradiating the fissile material in a nuclear reactor for maximum several times ten minutes. The properties of the products obtained in this manner closely approached the characteristics of fall-out reported in the literature [1-7].

To confirm this assumption, we performed gamma-spectrometric investigations: we determined the isotope composition and its change *versus*

cooling time, under equal conditions, using an apparatus consisting of a Ge(Li) semiconductor detector, a preamplifier type NV 800 (manufactured by ATOMKI), an analogue signal processing electronic unit type NV 804 (also by ATOMKI) and a 4096-channel amplitude analyzer type ICA 70 (manufactured by KFKI). Figs 3, 4 and 5 represent, by way of example, the gamma spectra of fission products and transuranium elements formed from uranium measured after cooling periods of 8, 14 and 23 days, resp.

Subsequently we studied the products for about two months, at strictly identical geometry of measurement, using a Si(Li) semiconductor detector coupled to the multichannel amplitude analyzer over a charge-sensitive preamplifier. From the differential  $\gamma$ -spectra obtained, we computed the gross pulse counts measured above individual energy levels by numerical integration.

Since energy calibration of the detector system (proportionality between pulse height and gamma energy) could be considered constant during the series of measurements, we could fit the function described by Eq. (3) to the integral

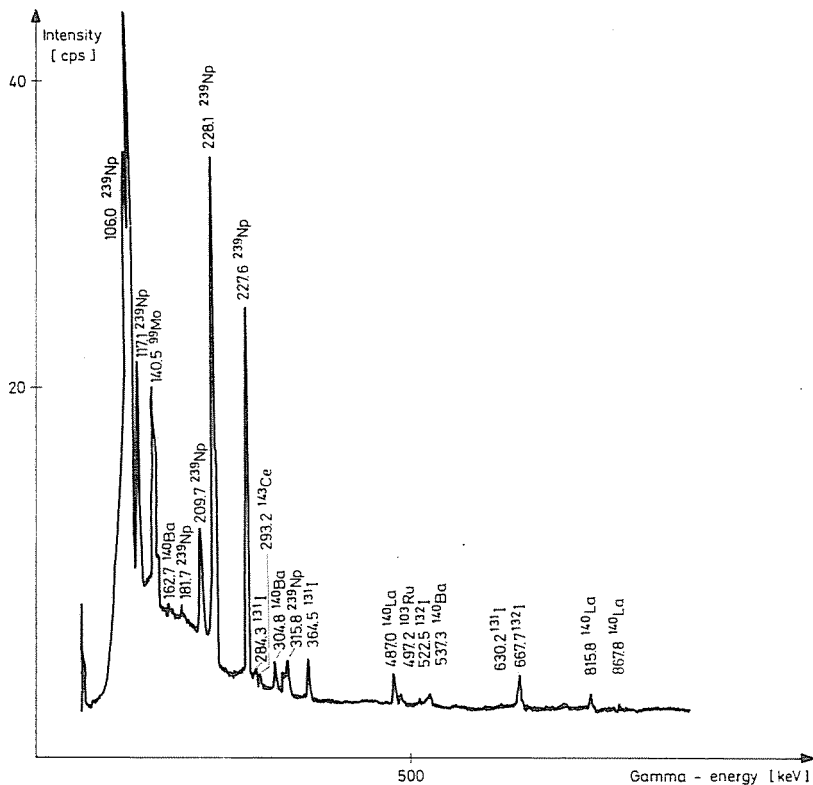


Fig. 3. Gamma spectrum of uranium fission and activation products. Cooling period: 8 days  
Sensor: Ge(Li) detector

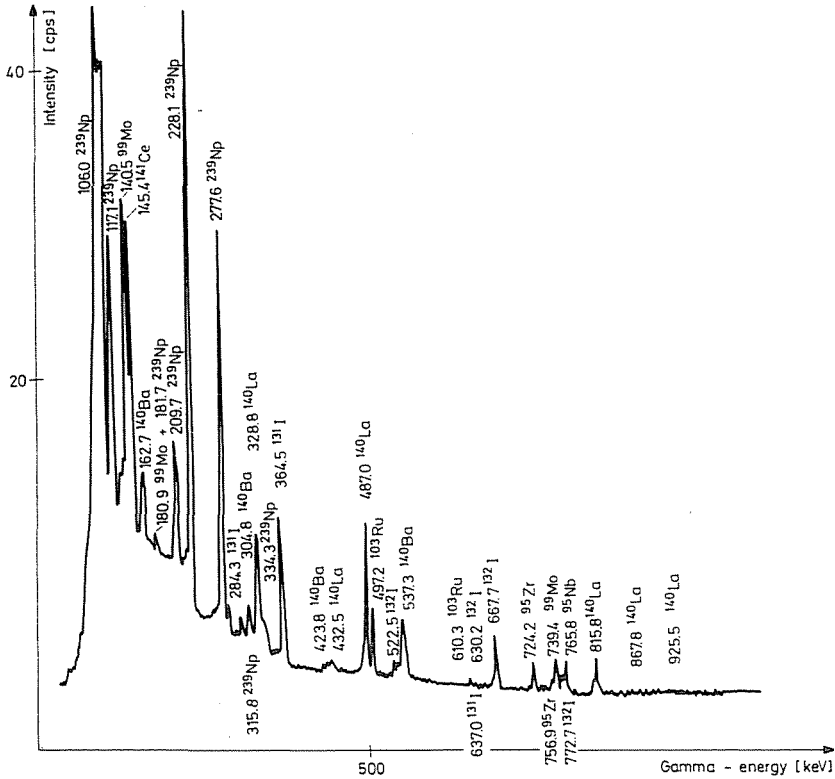


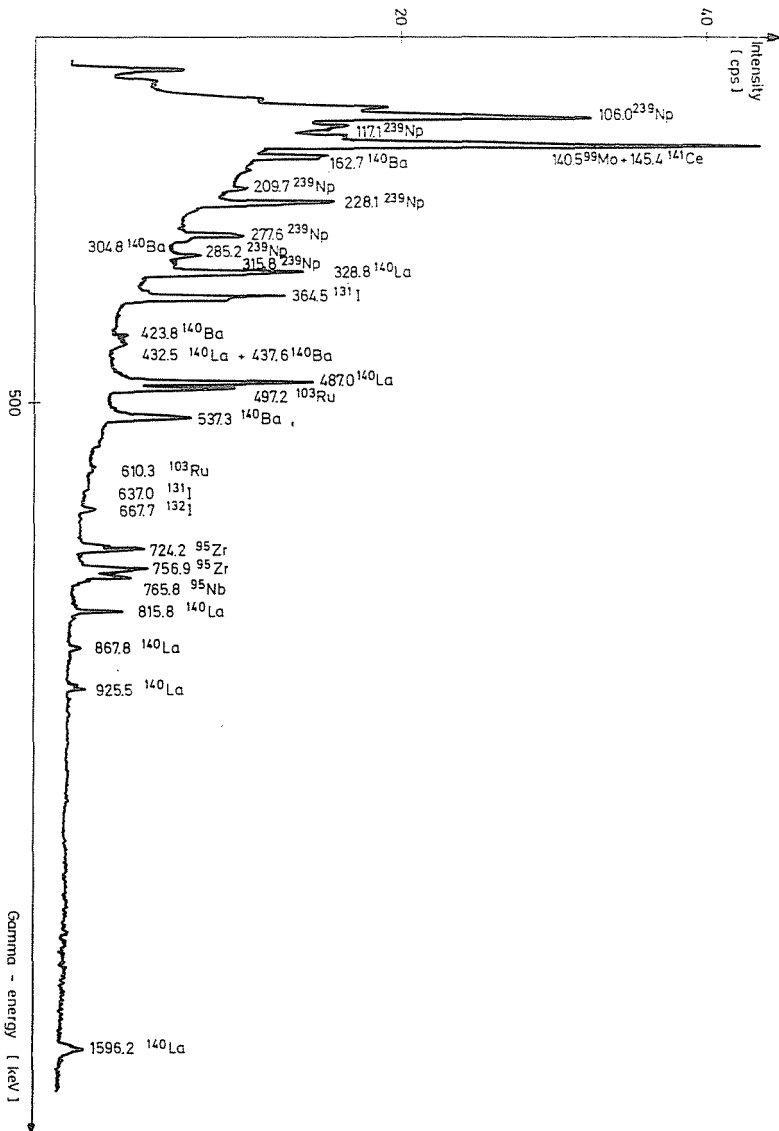
Fig. 4. Gamma spectrum of uranium fission and activation products. Cooling period: 14 days

intensity values assigned to one and the same channel of the spectra taken in different moments of time. The curve fitting program utilized regression linearized by logarithmation.

Reliability of results was promoted by the possibility that in forming integral spectra, the statistical error of integral intensities could also be obtained, and therefore we could perform fitting with appropriate weighting of the points; also, the goodness-of-fit  $\chi_{red}^2$  could confirm the applicability of Eq. (3). In fitting we made use of the observation already mentioned that Eq. (3) is to be used with differing exponents before and after the age of 100 hours.

The experimental results are presented in Figs 6, 7 and 8. For every 20th channel of the integral spectra we plotted the straight lines fitted by the least squares approach. It is remarkable that the regression lines have the same character for all three fission products: they diverge up to around the 100th hour and converge in the region after the 100th hour. This trend is well observable from the exponent values  $n$  marked in the figures, characterizing the slope.

Fig. 5. Gamma spectrum of uranium fission and activation products. Cooling period: 23 days



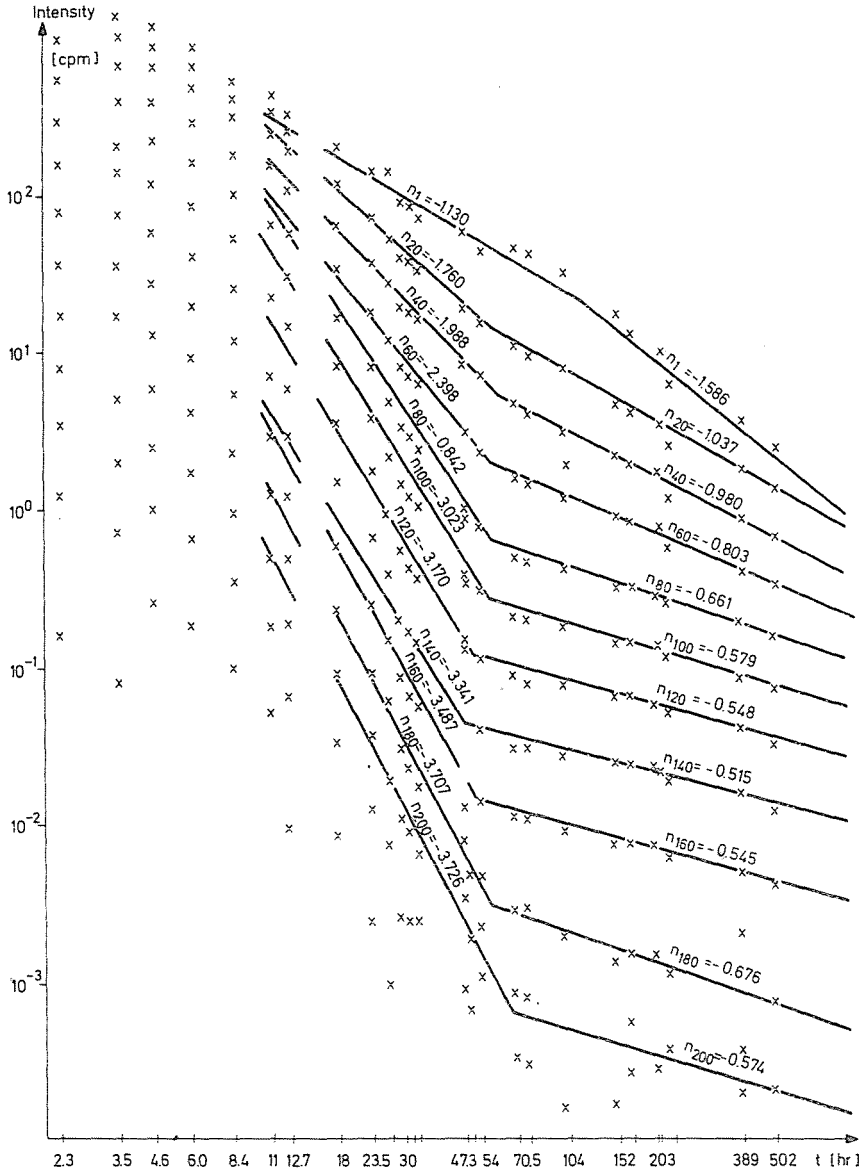


Fig. 6. Integral gamma spectra of  $^{238}\text{U}$  fission products

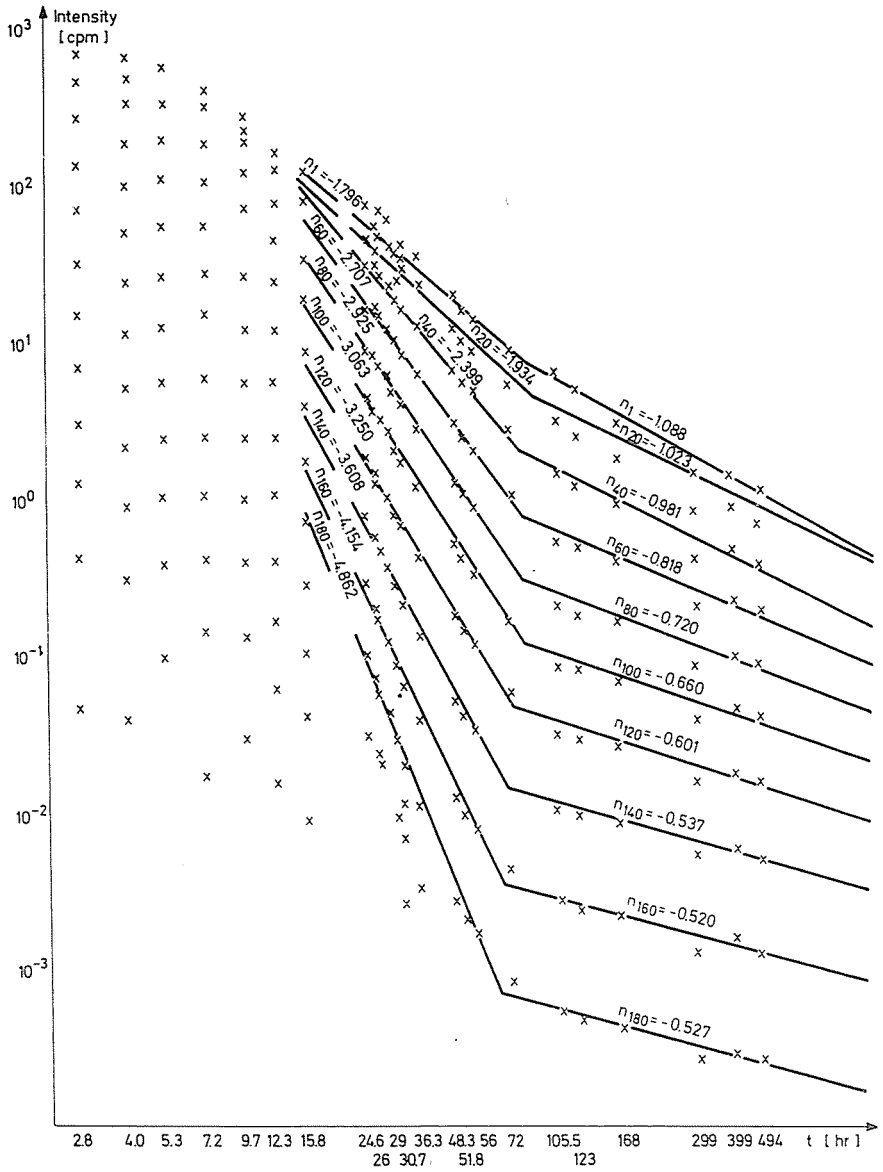


Fig. 7. Integral gamma spectra of  $^{235}\text{U}$  fission products



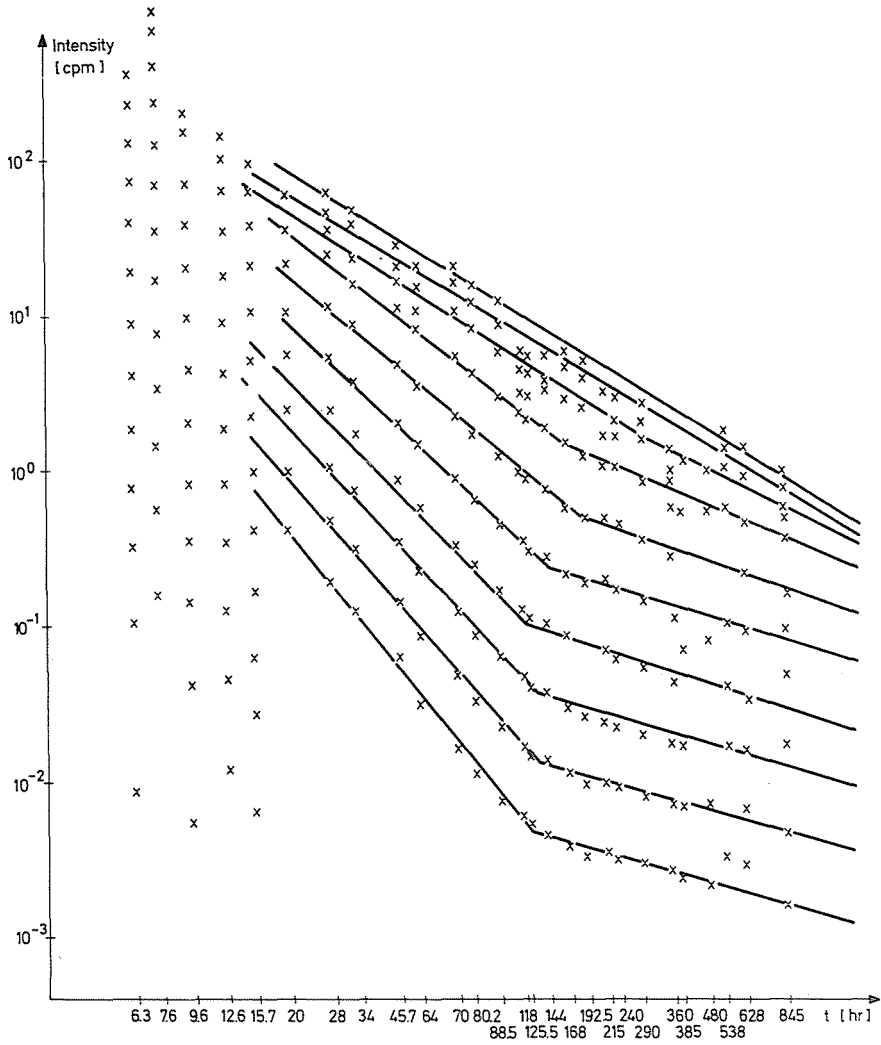


Fig. 8. Integral gamma spectra of  $^{239}\text{U}$  fission products

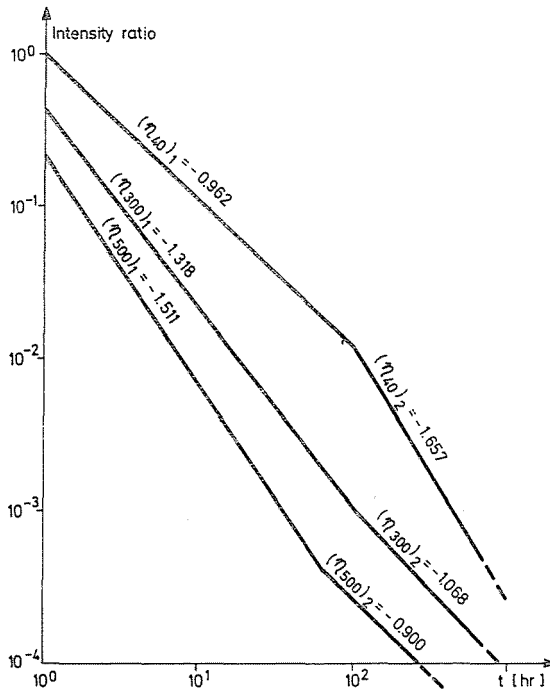


Fig. 9. Intensity ratios vs. time at three discrimination energy levels, for  $^{238}\text{U}$  fission products

The slopes assigned to different discrimination levels and differing in value guarantee the applicability of the quotient function procedure. It is also apparent from the figures that a preferential discrimination value can be selected for continuous observation of dose rate decrease, at which the decrease can be approached by a single function practically during the total time interval of measurement.

In the followings, we performed similar measurements, now with the probe of the designed portable instrument. Among the results obtained, those for the fission products of  $^{238}\text{U}$  are shown in Fig. 9, at the three discrimination energies selected as optimum for instrumental measurement. The curves are normalized in the figure to the initial value of 1 hr.

The quotient functions formed by pairs from the three channels are presented in Fig. 10. The break points in the quotient functions cause a special problem in instrumentation, since two different ages can be assigned to one and the same quotient value. Therefore, if one cannot decide by some other means whether the age to be determined is to the right or to the left of the maximum, repeated measurement is required; thereby, however, the procedure would lose its most important advantage, namely that only one measurement is necessary.

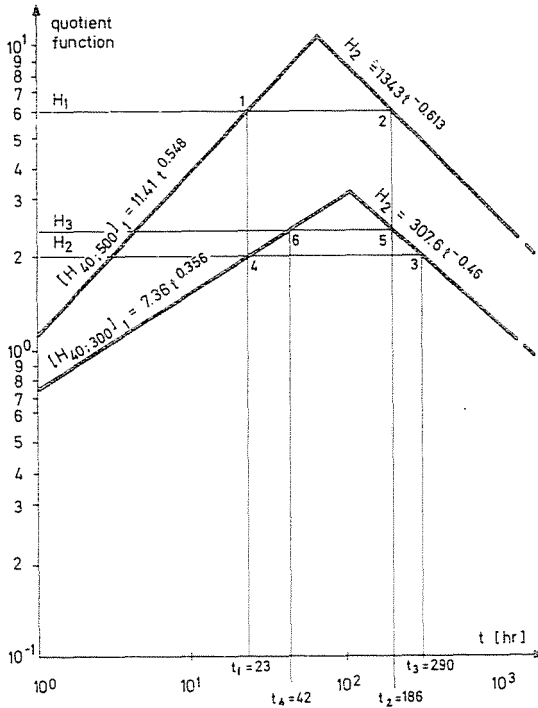


Fig. 10. Quotient function vs. time calculated from the data plotted in Fig. 9

The solution of the problem can be found in the asymmetry of the two pairs of quotient curves. Let us follow this on the example shown here.

Let us assume that the true age of our product is  $t_1$ . This age corresponds, on the curve  $H_{40.500}$ , to the quotient value  $H_1$  while on curve  $H_{40.300}$  it corresponds to the value  $H_2$ . What happens, if we attempt to determine the age of the product by means of the quotients  $H_1$  and  $H_2$  to the right of the maxima?  $H_1$  will correspond to  $t_2$  on the curve of the function  $H_{40.500}$  and to  $t_3$  on the curve of the function  $H_{40.300}$ . These values differ to an extent much larger than experimental error, while  $t_1$  is identical (within experimental error) by both functions. It is hence evident that the true value for the age of the product will be found in the interval below 100 hours.

In the opposite case, when the true age of the product is  $t_2$ , the correct result is obtained in a similar manner. Let us consider points 2 and 5, and points 1 and 6 in Fig. 10. Projecting them to the  $t$ -axis one arrives to the ages  $t_2$  (the correct age) and  $t_1$  and  $t_4$  differing largely. In this manner, the problem of the break in the quotient function is fully eliminated by introducing measurement at a third discrimination level, and age can be determined by three measurements of intensity at one and the same moment of time. It should be

stressed that in the present case, the most favourable quotient functions for age determination (those with the steepest slope) are provided by the quotients of intensities at the channel on the 40 keV discrimination level which measures the standard dose rate, and the 300 and 500 keV channels uncorrected for energy. Thus, the energy dependence of the semiconductor detector, — the electric correction of which being one of the cardinal problems of dose rate measurement — creates favourable conditions in the given case to implement the procedure in question for age determination.

The procedure was also tested with fission products of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . The results were similar to the above.

The two instruments designed to implement the procedures discussed were also built as portable instruments.

The block diagram of the instrument type SZÉM-1 for measuring radioactive contamination and age of the radioactive products is shown in Fig. 11. Its working principle is summarized in the followings. The probe 1 including the energy-selective detector  $De$  and the preamplifier  $E$  serves to transform ionizing radiation of the radioactive products to electric signals. The signals are transmitted to the unit 2 which corrects energy dependence of the detector and allows to pass all signals above one of the selected energy levels (in our case 40 keV). Its output is  $I_{40}$ , that is, the count rate including all pulses, generated by all forms of interaction, that have an energy deposited in the detector higher than 40 keV. The signals of the probe are also transmitted to unit 3; its output is  $I_{300}$  and  $I_{500}$ , resp., that is, the count rates for all pulses exceeding 300 and 500 keV, resp., set on the discriminations  $D_2$  and  $D_3$ . The outputs of 2 and 3 are transmitted to unit  $H$  serving to compute quotients. Age is then computed by the function units  $F_1, F_2, F_3$ .

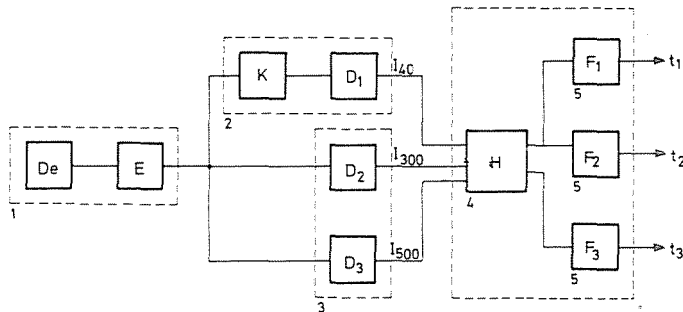


Fig. 11. Block diagram of the instrument SZÉM-1 for age determination  
 $De$  — detector;  $E$  — amplifiers;  $K$  — device to correct energy dependence of the detector;  $D$  — discriminator;  $H$  — quotient computing unit;  $F$  — function computing units

### Instrument data

**Dose rate measurement:** in the range of  $20 \mu\text{Gy/h}$  to  $20 \text{mGy/h}$  ( $\sim 2 \text{mR/h}$  to  $2000 \text{mR/h}$ ) with an error of maximum  $\pm 10\%$ .

**Energy dependence:** in the interval of  $80 \text{keV}$  to  $2 \text{MeV}$  maximum  $\pm 20\%$  relative to  $^{60}\text{Co}$ .

**Age determination:** in the interval of  $30$  to  $600$  hours with an error of maximum  $\pm 30\%$  (at dose rates of  $10 \mu\text{Gy/h}$  to  $20 \text{mGy/h}$ ).

**Power supply:** from  $220 \text{V}$ ,  $50 \text{Hz}$  main network with inbuilt adapter, from  $12 \text{V}$  automobile battery, or from own battery (four batteries  $1.2 \text{V}$ ,  $3 \text{Ah}$  each).

**Temperature of operation:**  $-20 \dots +50 \text{ }^\circ\text{C}$ .

The block diagram of the instrument type SVJ indicating the degree of radiation danger is shown in Fig. 12. The dose rate measuring instrument 1 continuously measures the actual value of dose rate  $P_i$ . This value is transmitted to the comparator circuit 3. The dose rate  $P_1$  is transmitted to the multiplying and storage circuit 2, whose output, the product  $K_1 P_1$ , is also transmitted to 3. The starter 5 electronically starts the counter 6 when the dose rate  $P_1$  appears, and the stopper unit 4 connected with the output of the comparator circuit 3 electronically stops the counter 6 as soon as — with monotonously decreasing values of the dose rate  $P_i$  — the dose rate reaches the value  $K_1 P_1$  ( $K_1 < 1$ ). The dose  $D_{1,2}$  appearing as the output of the counter 6 is then transformed — according to Eq. (14) — by the multiplier circuit 7 into the forecast value of  $D_{1,\infty}$ , that is, the integral dose during the total period of decay.

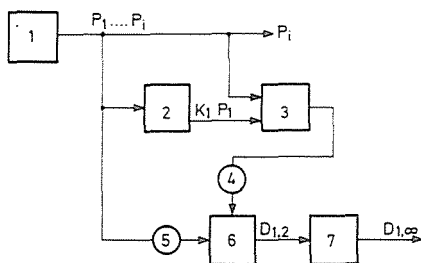


Fig. 12. Block diagram of instrument SVJ indicating the forecast value of the integral dose during total decay time

1 — dose rate meter; 2 — multiplying and storage circuit; 3 — comparator circuit; 4 — stopper unit; 5 — starter unit; 6 — counter; 7 — multiplying circuit

### *Instrument data*

*Dose rate measurement:* in the range of 1 mGy/h to 200 mGy/h ( $\sim 0.115$  R/h to 23 R/h) with an error of maximum  $\pm 10\%$ . Indication: 200 mGy/h to 300 mGy/h (23 R/h to 34.5 R/h). Indication does not change up to 2 Gy/h (230 R/h).

*Energy dependence:* in the 80 keV to 2 MeV range maximum  $\pm 20\%$ .

*Forecast value of integral dose during total decay:* in the range of 10 mGy to 8 Gy ( $\sim 1$  R to 800 R) for fission products of various fissile substances with an error of maximum  $\pm 50\%$ .

*Threshold values for indication:* two stages for dose rate, six stages for dose, set by the manufacturer within the above range as required by the customer.

*Power Supply:* from 220 V, 50 Hz main network with inbuilt adapter, or directly from 12 V automobile battery.

*Operating temperature:*  $-20 \dots +50$  °C.

The mentioned instrument is produced by the Gamma Works Budapest.

It is of interest to mention that we also applied the procedure of integral spectrum quotients for the fission product composition of  $^{235}\text{U}$  reported by Nelms and Cooper in 1959 [6]. We confirmed that the procedure is valid for these data. For instance, among the values published in Table 2 of [6], we found that steepness of the quotient function was highest for the integral spectra above the energy values  $E_{\gamma 1} > 1.0217$  MeV and  $E_{\gamma 2} > 2.0435$  MeV. Applying the symbols of Eq. 5, the values of the parameters were  $(H_0)_{1,2} = 4.266$  and  $n_{1,2} = 0.194$ . These values will obviously change with the actual system of measurement applied, and steepness, at best, can be increased, since the above value does not appear promising to achieve satisfactory accuracy in age determination. However, we did not consider the example of interest in this view, but from the view that it confirms the applicability of the procedure using foreign data.

On the other hand, the closely similar result obtained were useful to check that the model substances used in our experimental work were in good agreement with the data from the literature.

Both nuclear measuring procedures developed can be implemented with portable instruments by virtue of their simplicity. However, widespread microprocessor application in Hungary raised the question of further joint development of procedure and instrument. We are therefore continuing research, in order to develop a similarly simple and rapid, but more reliable and more accurate procedure for age determination, making use of the advantages offered by microprocessors.

Summing up: the two new procedures for age determination allow to estimate the external and internal radiation loads caused by fission products more reliably; the simplicity of the procedures allow their application in

portable instruments; rapidness of the procedures reduces time requirement, this being of inestimable significance under extraordinary conditions. Time requirement is further reduced by on-site measurement and evaluation of data.

### References

1. The dosimetry of ionizing radiation in nuclear explosions (in Russian). VKAKHZ, Moscow, 1976.
2. IZRAEL, YU. A.–STUKIN, E. L.: Gamma radiation of radioactive fall-out (in Russian). Atomizdat, Moscow 1967.
3. GUSEV, N. G.: Protection from gamma radiation of fission products (in Russian). Atomizdat, Moscow 1968.
4. PETROV, R. V. et al.: Protection from radioactive precipitates (in Russian). Medgiz, Moscow, 1963.
5. GREGUSHKINA, M. P.: Tables on the composition of fission products of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$  (in Russian). Gosatomizdat, Moscow 1964.
6. NELMS, A. T.–COOPER, J. W.: Health Phys. 5, 427 (1959).
7. Radioactive fall-out from nuclear explosions. (Russian translation from English.) Mir, Moscow, 1968.
8. WAY, K. and WIGNER, E.: Phys. Rev. 72, 1318 (1948).
9. Handbook of civil defense for radiology, biology and chemical protection. Part V. (in Hungarian). National Headquarters of Civil Defense, Budapest, 1978.
10. DOLAN, P. J.: Gamma spectra of uranium-235 fission products at various intervals after fission. AFSWP 524, 1959.
11. MILLER, C. P.: Gamma decay of fission products from the slow neutron fission of  $^{235}\text{U}$ . USNRDL-TR-187, 1957.
12. COOK, C. S.: Health Phys. 6, 42 (1960).
13. SVENSSON, G. K.–LIDÉN, K.: Health Phys. 11, 1033 (1965).
14. SHIROKOV, C. P.–SHEEDOV, S. I.: Radioactive contamination of the environment (in Russian). Atomizdat, Moscow 1962.
15. SILANTEV, A. N.: Atomnaya energiya, 7, 67 (1969).
16. MAMURO, T.–YOSHIKAWA, K.–MAKI, N.: Health Phys. 11, 199m (1968).
17. MAMURO, T. et al.: Health Phys. 12, 757 (1966).
18. MAMURO, T.–MATSUNAMI, T.–FUJITA, A.: Health Phys. 14, 223 (1968).
19. SOLYMOSI, J.: Thesis, Technical University Budapest, 1974.
20. SOLYMOSI, J.–TÖMÖR, J.–GAÁL, L.: Hung. Patent No. 177623.
21. SOLYMOSI, J.: Candidate's thesis, Budapest 1983.

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