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NONDESTRUCTIVE METHOD FOR ASSESSMENT OF NUCLEAR FUEL

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Description of a gamma spectrometric technique which has been developed with the aim to determine an amount of a certain radioactive fission product considering local variations of the linear attenuation coefficient of gamma rays is given. Also a preliminary experiment using a fuel element of TRIGA Mark II reactor in Ljubljana is represented.

1. Introduction

Gamma scanning is the only nondestructive technique for the quantitative measuring of fission or activation products in spent fuel. Data, obtained from such measurements serve for studying the migration of volatile fission products, for characterization the performance of the fuel elements during the irradiation, etc.

On account of the attenuation of gamma rays in matter, the only reliable gamma-spectrometric determination of the desired account is obtained from a measurement of the relevant isotopic density in the fuel. With the methods resembling those used in medical tomography, these problems have been worked on in the field of nuclear technology (see for example Ref. 1 or 2). However, the neglect of local variation of the linear attenuation coefficient of gamma rays in the irradiated fuel remains the main source of systematic error.

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Some years ago we studied how to take into account these effects. It has been decided to combine the (single) emission gamma ray scanning technique with a transmission experiment[3,4]. The method using the additional source of gamma rays was developed sufficiently to enable preliminary measurements to be made. We have also tested the programs for processing of measured data.

2. *The experimental set-up*

For measurement of gamma-ray spectra a high resolution Ge (Li) detector is employed. It views two shielded fuel elements through a narrow collimator (Fig. 1). One of them serves as an additional gamma ray source used in the trans-

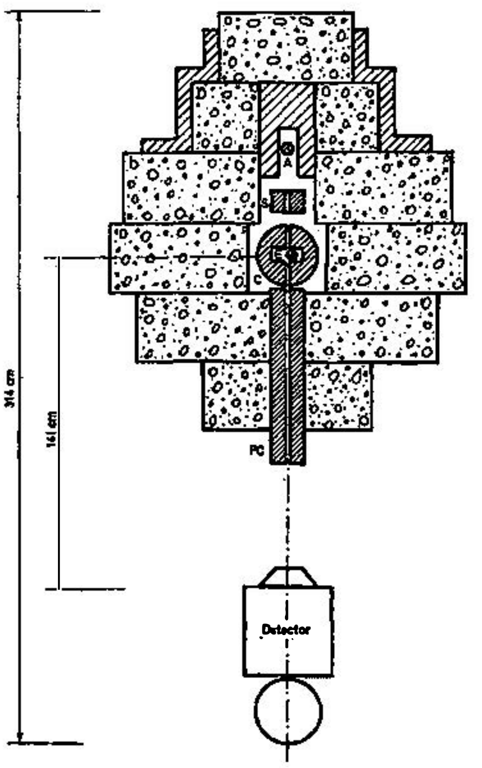

Fig. 1. Horizontal section through the experimental set-up. Shielding is of lead and heavy concrete. The effective width of the gap on the detector side of the collimator C for the 662 keV line is 0.07 cm. Rotation of the lead shutter S by 90° screens the additional fuel element A. The precollimator PC lies between the collimator and detector. E denotes the examined fuel element.

mission part of the experiment. The examined fuel element may be rotated and moved normally to the axis connecting the detecor and additional fixed source (Fig. 2). It hangs on a torsional wire fastened to the vertical axis of the rotator,

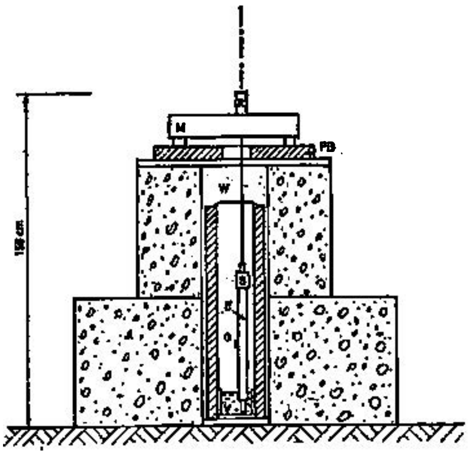

Fig. 2. Vertical section through the experimental set-up. Here M denotes the moving device with rotator R, W the torsional wire, G the collimation gap, S the scraper, E the examined fuel element, PB lead bricks, and V a vessel filled with water.

which is a part of the moving device. The lower end of the examined fuel element dips into a vessel filled with water. In such a way small oscillations appearing when the measured object is moving are damped out.

Owing to the great number of gamma ray spectra measurements the experiment has been partially automated. We worked out a group of congruent routines, which starts and stops the partial measurements, transfers the measured spectra into files, and governes the moving device[5,6]. The spectral data processing is based on ORTEC's analysis software package.

3. *Field of examination and array of pixels*

The reconstruction of an object from its projections can be described as an inverse transformation. If $F(\vec{r})$ represents the value of a physical property, the measured projections are in the most simple case values of the line integral $\int F(\vec{l} + \vec{n}t)\,\mathrm{d}t$ (see Fig. 3). Mathematician Radon was first who solved the basic mathematics of the problem. He proved that the object can be uniquelly reconstructed from an infinite set of projections. In practice, there are always at least two drawbacks: the number of measured projections is limited and their value is of a finite accuracy. Consequently there does not exist a single way which would be able to satisfy all geometrical and precision requirements.

During the measurement the fuel element is moved in the horizontal plane in which lies the detector with the collimation apertures. So we have in essence a two-dimensional image reconstruction from projections. It remains the same even if we make a convolution of such measurements in the vertical direction.

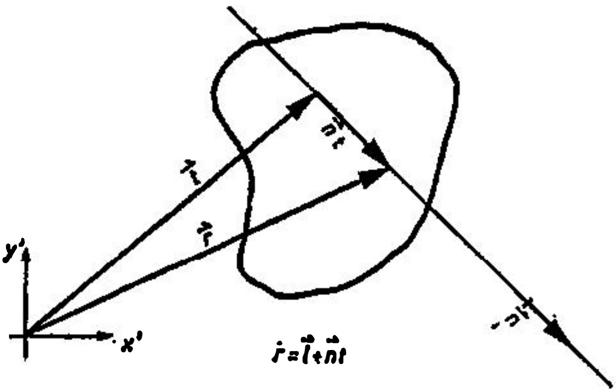

Fig. 3. The line of the integration in the laboratory system x', y'

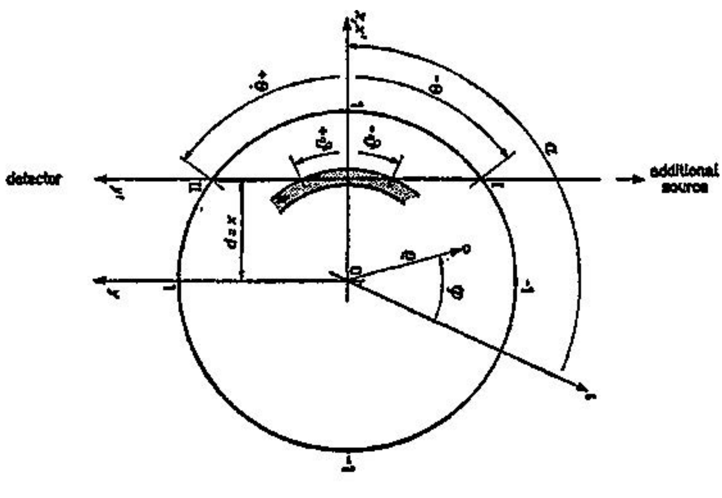

Fig. 4. Field of examination.

We treat the field of examination as a circle of unit radius in which the points are given with polar coordinates ϱ and φ belonging to the body system of the object of examination (Fig. 4). The origins of the coordinate systems x, y and ϱ, φ coincide. The abscissa x represents the distance between the centre and the fixed axis y' connecting the detector with the additional source, and the angle α represents the rotation of the system ϱ, φ with regard to the system x, y.

An array of pixels is formed by dividing the unit circle into K concentric rings with the same width $1/K$ (Fig. 5). These rings are composed from particular area elements (pixels). Areas of all pixels are equal. Inside the single area element the linear attenuation coefficient of gamma rays and the activity are treated as constants.

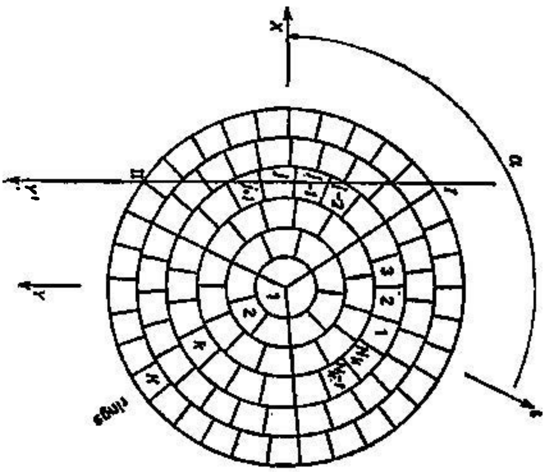

Fig. 5. Array of pixels.

4. Counting rate and activity on the observed cross-section

During the measurement counting rates are being determined for chosen distances p as a function of the rotation α. In the absence of the additional source the only contribution comes from the slice I, II in the field of examination, as it is viewed from the detector. In the following we simply set the efficiency of the detector-collimator system as 1.

Temporarily we assume that the thickness of the slice is negligible in the comparison with the size of a pixel. Thus we take it is a straight line connecting points I and II. An arbitrary point on this line is determined with p, α, φ: $-\Theta(p) \leq \varphi \leq \Theta(p)$. In these circumstances the counting rate is given by:

$$f(p, \alpha) = \int_{\alpha-\Theta}^{\alpha+\Theta} A(p, \alpha, \varphi)\, S(p, \alpha, \varphi, \alpha + \Theta)\, \frac{\partial y(p, \varphi - \alpha)}{\partial \varphi}\, d\varphi$$

where

$A(p, \alpha, \varphi)$ is the activity of the observed gamma line at the point $\frac{p}{\cos(\varphi - \alpha)}, \varphi$

$$S(p, \alpha, \zeta, \xi) = \exp\left[-\int_{\zeta}^{\xi} \mu(p, \alpha, \eta)\, \frac{\partial y(p, \eta - \alpha)}{\partial \eta}\, d\eta\right], \quad \zeta \leq \xi$$

is the attenuation of gamma ray on the straight line between points

$$\frac{p}{\cos(\zeta - \alpha)}, \xi \text{ and } \frac{p}{\cos(\xi - \alpha)}, \xi$$

$\mu(p, a, \eta)$ is the linear attenuation coefficient at the point $\frac{p}{\cos(\eta - a)}, \eta$ and $(\partial y/\partial \varphi)\,\mathrm{d}\varphi$ is the element of the lenght on the y' axis. Subsequently we use the abbreviation

$$K(p, a, \zeta, \xi) = S(p, a, \zeta, \xi) \frac{\partial y(p, \xi - a)}{\partial \zeta}.$$

In the case of a sufficiently fine division of the field of the examination, the simplification of a thin slice breaks down. For this reason we have developed the model which takes into account a real transpassing response $u(\vec{\varrho}$, parameters). This function is proportional to the counting rate when a point source is positioned at $\vec{\varrho}$ in the body system while parameters are kept constant. Because the broadening of the slice in our field of examination does not exced 5%, we treat the transpassing response as a function of only one variable x and parameter p (Fig. 6).

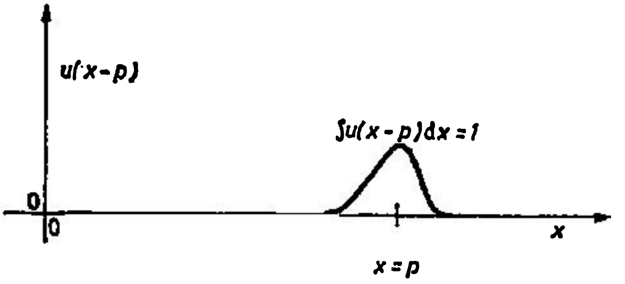

Fig. 6. Transpassing response $u(x - p)$. Thy symbol p represents the distance between the origins of the laboratory system x', y' and of the body system ϱ, φ.

So in the absence of the additional source, the counting rate is given by the convolution

$$f(p, a) = \int_x \int_{a-\Theta(x)}^{a+\Theta(x)} u(x - p)\, A(x, a, \varphi)\, K(x, a, \varphi, a + \Theta(x))\, \mathrm{d}\varphi\, \mathrm{d}x.$$

From this equation we extract the contributions of the ring with the lowest index k. These lie in the interval $\varphi \in (a - \vartheta(x), a + \vartheta(x))$. Considering the equality $S(x, a, \zeta, \xi) = S(x, a, \zeta, \omega)\, S(x, a, \omega, \xi)$ it follows that

$$\int_x \int_{a-\vartheta(x)}^{a+\vartheta(x)} u(x - p)\, A(x, a, \varphi)\, K(x, a, \varphi, a + \Theta(x))\, \mathrm{d}x\, \mathrm{d}\varphi = f(p, a) -$$

$$- \int_x u(x - p)\, [S(x, a, a - \vartheta(x), a + \Theta(x)) \int_{a-\Theta(x)}^{a-\vartheta(x)} A(x, a, \varphi)\, K(x, a, \varphi, a -$$

$$- \vartheta(x))\, \mathrm{d}\varphi + \int_{a+\vartheta(x)}^{a+\Theta(x)} A(x, a, \lambda)\, K(x, a, \varphi, a + \Theta(x))\, \mathrm{d}\varphi]\ \mathrm{d}x.$$

In the presence of the additional source, the counting rate increases for the transmitted amount. Denoting by P_0 the counting rate for the unscreened additional source while the examined one is being shifted away, we have the counting rate

$$g(p, a) = f(p, a) + P_0 \int_x u(x - p) K(x, a, a - \Theta(x), a + \Theta(x)) \, dx.$$

We write out this relation as

$$h(p, a) = \int_x [u(x - p) K(x, a, a - \Theta(x), a - \vartheta(x)) \cdot K(x, a, a + \vartheta(x), a + \Theta(x)) K(x, a, a - \vartheta(x), a + \vartheta(x))] \, dx.$$

Here, the measuring data are the values g, f, and P_0 in the quotient $h = (g - f)/P_0$.

Unknowns are hidden in the function K.

5. *Discretization*

The number of measured projections is determined in such a way that to each position p_k belongs a certain number of N_k spectra measured with the additional source both screened and unscreened. The i^{th} measurement is done at $a_i = (i - 1) 2\pi/N_k$, where only few neighbouring pixels from k^{th} ring contribute to the counting rate. We avoid here a derivation and only give the result of the discretization:

$$\sum_l q_{il}(k) A_{i+l}(k) = f_i(k) - \sum_j u_j [(S_{ij}^{sp} S_{ij}^{sr} P_{jl}^{zq})(k) - P_{jl}^{sp}(k)] \qquad (*)$$

and

$$h_i(k) = \sum_j u_j S_{ij}^{sp}(k) S_{ij}^{zq}(k) \prod_l e^{-\mu_{i+l}(k) R_{lj}(k)} \qquad (**)$$

where

$$q_{il}(k) = \sum_m u_m S(x_m, a_i, a_i + \vartheta_m, a_i + \Theta_m) R_{lm}(k) \sigma_{m, i+l}(k)$$

$R_{lm}(k)$ is the length of the prime $(\mathrm{I}, \mathrm{II})_m$ at $x = x_m$ within $(i + l)^{th}$ pixel in k^{th} ring (Fig. 7)

$\sigma_{m,i+l}(k)$ is the element of the self-absorption matrix which corresponds to $R_{lm}(k)$ at i^{th} rotation

$$S_{ij}^{sp}(k) = S(x_j, a_i, a_i + \vartheta_j, a_i + \Theta_j)$$

$$S_{ij}^{sr}(k) = S(x_j, a_i, a_i - \vartheta_j, a_i + \vartheta_j)$$

$$S_{ij}^{zq}(k) = S(x_j, a_i, a_i - \Theta_j, a_i - \vartheta_j)$$

$$P_{jl}^{sp}(k) = \int_{a_i + \vartheta_j}^{a_i + \Theta_j} A(x_j, a_i, \varphi) K(x_j, a_i, \varphi, a_i + \vartheta_j) \, d\varphi$$

$$P_{jl}^{z}(k) = \int_{a_i - \Theta_j}^{a_i - \Theta_j} A(x_j, a_i, \varphi) K(x_j, a_i, \varphi, a_i - \vartheta_j) \, d\varphi.$$

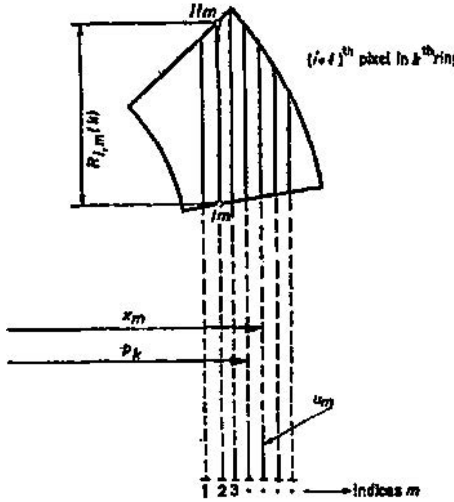

Fig. 7. Illustration of denotations u_j and $R_{l,m}(k)$.

Matrix elements R_{lj} are wholly geometrical quantities, and values u_j are obtainable by a separate measurement. From the nonlinear system of equations (**) we estimate the linear attenuation coefficient of the observed line in the area elements of the ring k. The next step is the determination of the matrices q, σ and S. After that follows the computation of the activity from the linear system of equations (*). The computation begins in the outer ring, since we know the conditions out of the measuring field: there all $A_l(j)$ and $\mu_l(j)$ are equal to zero.

6. *Measurement and results*

In recent years some preliminary measurements have been performed. In this paper we describe the latest of them. The fuel element used was E 3899, which was cooled for about twenty months prior to the measurement.

The diameters of the cylindrical fuel element and of the fuel are 3.8 cm and 3.51 cm, respectively. The diameter of the field of the examination is 4.16 cm. It is divided into 768 pixels. The observed cross section of the fuel element lies almost concentrically with this field. The number of measured spectra for the additional source screened and unscreened was determined so that it equals in both cases to the number of pixels.

From the evaluated gamma-ray spectra the dominant lines of the isotopes ^{137}Cs, ^{134}Cs, ^{95}Zr, ^{106}Ru, and ^{144}Ce were separated. Measuring data were calculated for the day on which the fuel element was taken out of the reactor core.

For the time being we can present the activity distribution of the 662-keV line belonging to the isotope ^{137}Cs. It is shown in Fig. 8 for two orientations of the body system with respect to the laboratory system. The average value of the specific activity in the observed cross-section of the fuel is $1.036\,(1 \pm 0.003) \cdot 10^9\ \mathrm{cm^{-3}\,s^{-1}}$. This corresponds to $2.00\,(1 \pm 0.003) \cdot 10^{18}$ ^{137}Cs atoms per cm^3. The accuracy of the calibration source was not taken into account.

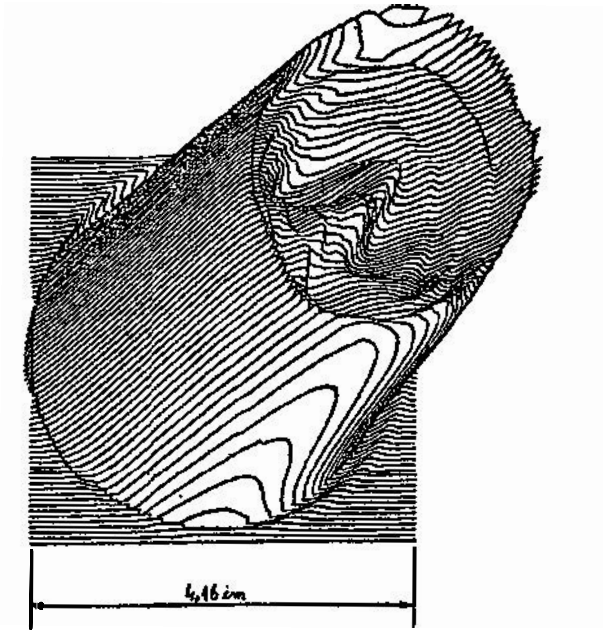

Fig. 8a. The distribution of the isotope ^{137}Cs in arbitrary units.

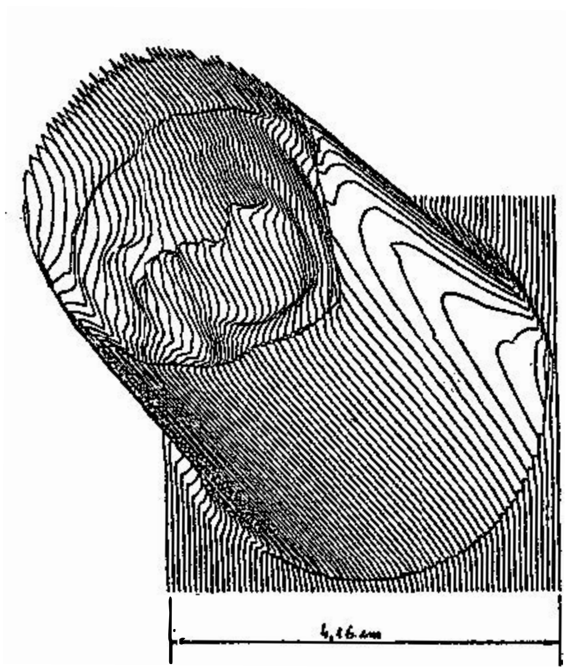

Fig. 8b. The same as in Fig. 8a. Here the body system is rotated for $\pi/2$.

In Fig. 9 we can see the linear attenuation coefficient for the 662-keV line along the axis 3 $\pi/2$, $\pi/2$.

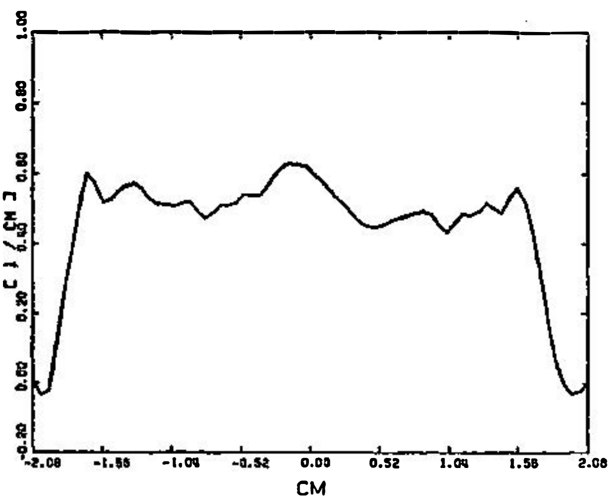

Fig. 9. The linear attenuation coefficient for the 662 keV line along the axis $3\pi/2$, $\pi/2$.

7. *Conclusion*

The described nondestructive technique for the determination of the amount of an active fission or activation product has been applied to some fuel elements of the TRIGA Mark II reactor in Ljubljana. Trial measurements were carried out, and the measured data were successfully processed. Therefore this problem has been proved to be feasible.

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NEPORUŠNA PREISKAVA JEDRSKEGA GORIVA

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Originalno znanstveno delo

Opisujeva gama spektrometrično tehniko za določitev količine radioaktivnih razcepnih produktov, ki omogoča upoštevanje lokalnih sprememb linearnega atenuacijskega koeficienta za žarke gama. Predstavljen je tudi uvodni eksperiment, pri katerem je bil uporabljen eden od gorivnih elementov reaktorja TRIGA Mark II v Ljubljani.