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ORIGINAL PAPER

Modeling of neutron activation process with Americium Beryllium source. Application to the activation of fluorspar samples

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Abstract This paper shows the mathematical models which represent the phenomena that occur in a neutron activation process. These phenomena are, on the one hand, the neutron flux decreases with the distance between the neutron source and the sample, and on the other hand, the attenuation of the gamma rays originating from the sample activated on its way to the detector position. The development of the mathematical model has been divided into two parts. Firstly, the phenomena are shown separately. Secondly, the phenomena are shown together. Finally, this model is fitted to the neutron activation of a fluorspar sample, and the influence of the two phenomena as defined above can be seen.

Keywords Nuclear activation · Deferred gamma rays · Mathematical model · Fluorspar

1 Introduction

Neutron activation is a process in which an atom emits a characteristic radiation when it is excited by a neutron. This system can be used to determine the presence of certain elements in a sample.

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Neutron activation analysis (NAA) was discovered in 1936 by Hevesy and Levi, who found that samples containing certain rare earth elements became highly radioactive after exposure to a source of neutrons. This observation led to the use of induced radioactivity for the identification of elements.

In the last several decades, this technique has been applied to determine a great variety of elements in many disciplines. These include environmental science as well as, biological, geological, and material science.

The basic elements used in neutron activation are:

- an radioactive source that allows the irradiation of a sample by neutrons and,
- a radiation detector that reads gamma radiation emitted by a sample during the decaying of the radioactive products. This radiation is produced in a given time and is characterized by the energy and time during which it occurs, and is characteristic of each element.

A neutron activation process is characterized by two phenomena that occur:

- the one during radiation which supposes that the neutron flux decreases with the distance between the neutron source and the sample [1].
- with the radiation reading, that implies the attenuation of the gamma rays originated in the decay of ^{16}N traversing the sample to the detector position. This attenuation is exponentially dependent on a characteristic attenuation coefficient of the sample [2].

2 Definition mathematical models

In the procedure used, the base of the container sample is located at a distance "a" of the irradiation position (source), and a distance "b" of the reading position (detector), Fig. 1.

On the one hand, neutrons emerging from the source after they traverse the space "a" (air) arrive at the sample where they do or do not interact with the sample, resulting in the activation of the fluorine atom. Neutron flux is reduced with the distance from the source.

On the other hand, the produced gamma rays traverse the sample to the detector position, and are attenuated as they travel.

2.1 First model

In the first model, we refer specifically to the phenomenon of the reduction of the neutron flux with distance from the source. It is supposed that the activation of a differential element dx of the sample only depends on the distance $(x+a)$ in the direction x to the source, and that the dependency ratio is inversely proportional to the square of the distance of the differential element at the center of the source. Consequently, it is assumed that gamma rays are not attenuated by distance.

In this process, we have observed the following proportionalities (Fig. 2):

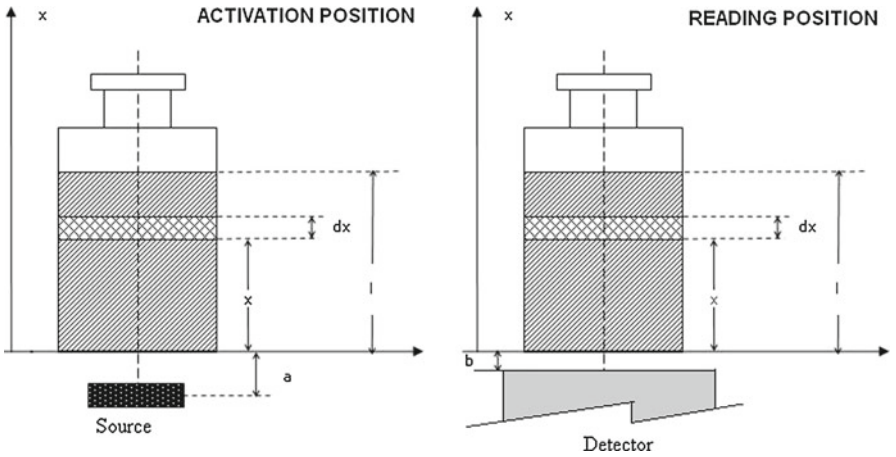

Fig. 1 A.1) Geometry considered in the activation position. A.2) Geometry considered in the reading position

$$C_m = k_1 * C_p$$

$$C_p = \int_0^l dC_{px}$$

$$dC_{px} = k_2 * dN_{ax}$$

$$dN_{ax} = k_3 * \Phi_{nx} * dN_{fx}$$

$$\Phi_{nx} = \frac{k_4}{(x+a)^2}; \qquad dN_{fx} = k_5 * y * dm$$

$$dm = dV * \rho_m$$

$$dV = S * dx; \qquad \rho_m = \rho_1 * y + \rho_2 * (1 - y)$$

Fig. 2 Expressions used in the proportionalities

- Counts from the sample and reading at the detector C_m are proportional to the counts C_p produced by the sample. The proportionality constant depends on the efficiency of the detector.
- Counts produced in the sample C_p are the integral from $x = 0$ to $x = l$ of the differential counts dC_{px} in an element of base S and height dx.
- The differential of counts produced dC_{px} at the differential element of height dx is proportional to the number of excited atoms in the differential element (dN_{ax}).
- The number of excited atoms dN_{ax} depends on neutron flux and the number of fluorine atoms dN_{fx} at that point.
- The neutron flux Φ_{nx} is inversely proportional to the square of the distance between the point and the source $(x + a)$.
- The number of fluorine atoms in a sample point dN_{fx} is directly proportional to the product of the grade y and the mass dm of differential element.
- The mass dm depends on the volume of differential element dV and the density ρ_m.
- The volume dV is defined by the section of the sample S and by the height of differential element dx.
- The density of the sample ρ_m is in relation to the density of the fluorspar ρ_1, the sterile ρ_2, and the sample grade in per unit.

Substituting these terms:

$$C_m = k_1 * k_2 * k_3 * k_4 * k_5 * \left(P * y + Q * y^2\right) * \int_0^l \frac{dx}{(x+a)^2}$$

where,

$$Q = S * (\rho_1 - \rho_2)$$

And,

$$P = S * \rho_2$$

The parameters Q and P are constants, greater than 0 and have dimensions of mass per unit length.

Making,

$$K = k_1 * k_2 * k_3 * k_4 * k_5$$

and integrating leads to the equation,

$$C_m = K * \left(Q * y^2 + P * y\right) * \frac{l}{l+a}$$

We can express the value of the height of the sample l (in cm) in function of the mass m (in grams), of the cross section of a container sample S(in cm^2), of the grade, of the fluorspar density ρ_1 and of the sterile density ρ_2,

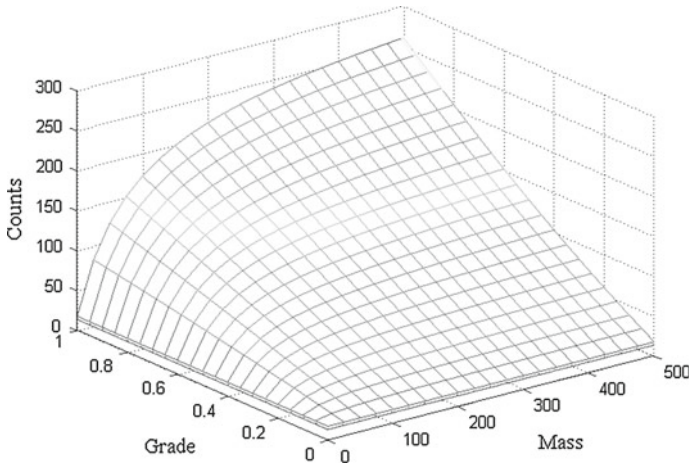

Fig. 3 Representation of the equation in the study carried out with MATLAB

$$l = \frac{m}{(\rho_1 * y + \rho_2 * (1 - y)) * S}$$

Then, in this model the relationship between counts, mass and grade is the following:

$$C_m = \frac{K * m * \left(Q * y^2 + P * y\right)}{a * (m + Q * a + y + P * a)} + F$$

Where, *Cm* is the integral of the counts from the sample in a certain range of channels for this first model,

- P y Q are constants and their values are greater than 0,
- a is the distance between the source and the base of the sample,
- y is fluorspar grade constant in the sample expressed per unit,
- l is the height of the sample.
- F counts from detector background without sample.

This equation is a surface in three dimensions whose *X* and *Y* axes are mass and grade and the *Z* axis is the counts (Fig. 3).

2.2 Second model

In the first model, we refer specifically to the phenomenon suffered by gamma rays produced from an irradiated sample before being detected. In the radiation reading position, the detector is at a distance b from the sample, as shown in Fig. 1. The value b is very small, negligible compared with the height l of the sample. This fact together with the attenuation of gamma rays in the air being lower than in the sample, leads to the observation that attenuation takes place only in the mass of the sample.

In this process, we have observed the following proportionality, some of them identical to previous case:

- The counts from the sample and reading in the detector C_m have an exponential relation to the counts dC_{px} produced by the sample. The proportionality constant depends on the efficiency and the coefficient of radiation attenuation:

$$C_m = k_1 * \int_0^l e^{-\mu * x} * dC_{px}$$

- As in the previous case, the count differential dC_{px} produced in the differential element of height dx is proportional to the number of excited atoms in the differential element dN_{ax}. The number of excited atoms dN_{ax} depends on neutron flux Φ_n (constant in this hypothesis) and of the number of fluorine atoms dN_{fx} at that point. The number of fluorine atoms at a sample point dN_{fx} is directly proportional to the product of the grade y and of the mass dm of the differential element, and as in the previous case, the mass dm depends on the volume of the differential element dV and on the density ρ_m.

Proceeding as previously, we obtain the equation for this second model, which includes the background, as follows:

$$C_m = \left(K' * Q * y^2 + K' * P * y\right) * \frac{1}{\mu}\left(1 - e^{-\mu * \frac{m}{Q * y + P}}\right) + F$$

The parameters Q and P are the same as in the previous case, while the factor that includes all proportionality constants K' is different.

2.3 Third model

In this third model, it is supposed that the activation of a differential element sample depends on the distance $(x + a)$ in the direction x to the source, and that gamma rays are attenuated on their way to the detector following an exponential $y = ae^{-bx}$.

In this case, the number of counts C in the energy range considered that come to the detector by the neutron activation effect can be expressed as:

$$Solution\ of\ the\ model\ 3 = \alpha\ *\ solution\ of\ the\ model\ 1 + \beta\ *\ solution\ of\ the\ model\ 2$$

α y β being the weight coefficients of each phenomenon individually in the final model, which fulfill the condition of being positive and their sum is equal to 1.

3 Checking the models

In order to determine which phenomena have greater impact on neutron activation, and therefore, the best model that fits to reality, each model is applied to neutron

activation of a fluorspar sample from a concentration plant. Equipment used consists of an Americium Beryllium source of 1 Ci of activity and a gamma ray detector of the type NaI. A prototype has been designed [3].

The reason for using fluorspar samples for testing the models is that the radiation of fluorspar with neutrons from Americium Beryllium source emits a characteristic high energy (6.13 MeV) which comes, according to previous studies [4], from the fluorine present in CaF_2. This radiation comes from ^{16}N originated, only and exclusively, from the nuclear reaction $^{19}F(n, \alpha)^{16}N$. Due to the characteristics of the detector, the energy spectrum of the sample, does not give a single peak at 6.13 MeV, but has a certain width, and has some 'echoes' called 'escape peaks' at 5.11 and 5.62 [5]. For this reason, the counts used in the study are in a range and are not in this exact value (6.13 MeV). The authors know the reactions produced in this mineral using the activation for analyzing fluorine in a fluorspar samples. An activation procedure and a mathematical method that increases the sensitivity were designed [6].

The fluorspar samples with variable fluorite grades [y] from 4 to 97% and variable masses [m] from 50 to 450 g (taken at intervals of 50 to 50) have been used. Samples are found in the same state of humidity and particle size.

3.1 Adjusting the first approach to the experimental data

Equipment was designed specifically to irradiate with neutrons and read the gamma rays emitted from fluorspar samples with the grades and masses specified above. Irradiation and reading times were adjusted according to the reaction sought and the coefficients K, a, Q, P, and F were determined. Nonlinear regression was used to determine the coefficients. The statistical program SPSS [7] was used. In this work only the coefficients obtained from taking the average values of the counts detected in the different tests, with an energy range between 4.5 and 6 are shown.

Algorithms used by statistical programs for nonlinear regressions are iterative processes which require the assignment of initial values for the coefficients. Table 1 shows the initial values for the parameters of the above equation.

Under these initial conditions the program was put into action and the parameters obtained are shown in Table 2.

Table 1 Initial parameters used for nonlinear regression

Density ranges	Parameters	Initial values
$\rho_1 = 1.5 - 1.9\,g/cm^3$		$K = 1$
	$P = S * \rho_2$ between 30 and 50	$P_0 = 45$
	$Q = S * (\rho_1 - \rho_2)$ between 3 and 16	$Q_0 = 9.6$
$\rho_2 = 1.1 - 1.4\,g/cm^3$	α between 0.5 and 8 cm	$\alpha_0 = 2\,cm$
	F between 10 and 25 counts	$F_0 = 13$ counts $= 1$

Table 2 Summary of the model parameters (Model 1)

Parameters. model 1	
Parameters	Energy 4.5–6 MeV
K	9.798
a	1.832
Q	7.515
P	38.978
F	13.598
$R2$	0.994

Table 3 Summary of the model parameters (Model 2)

Parameters. model 2	
Parameters	Energy 4.5–6 MeV
K'	1.992
μ	0.505
Q	10.122
P	43.464
F	13.6
$R2$	0.991

3.2 Adjustment of the second approach to the experimental data

Regression was carried out using the experimental results with a sample group. The procedure was repeated with the energy and value types. The parameters of the resulting equation are reflected in Table 3.

3.3 Adjustment of the third approach to the experimental data

Establishing restrictions of attenuation coefficient μ in the processing of the third model with the program SPSS, we are found that the correlation coefficient in this third model is lower than in previous models, so it follows that the phenomenon of gamma rays attenuation (expressed by β) is negligible.

4 Comparison between models

In the first model a very high attenuation coefficient μ is obtained and the parameters *a, K, P, Q* y *F* are consistent with the real values. However, the attenuation coefficient μ obtained in the second model is about 0.5, which is 10 times higher than expected given the composition of the sample. It therefore follows that the first model is the closest to reality.

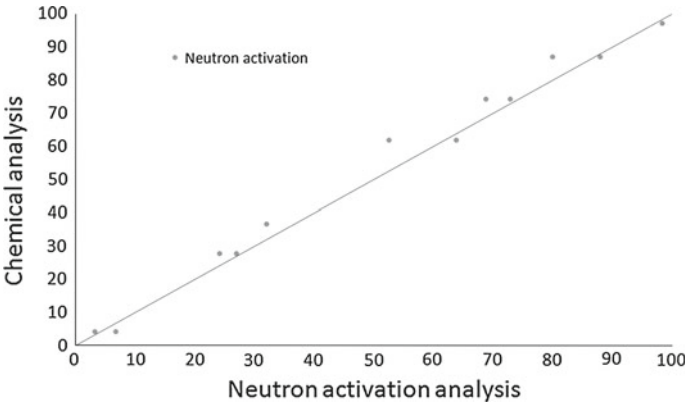

Fig. 4 Comparison between chemical analysis and neutron activation with samples used in the deduction of the model

5 Validity of the first model

Fluorite grades are compared with grades obtained from the first model for activated fluorspar samples with the parameters identified above. These parameters are $K = 9.798$; $\alpha = 1.832$; $Q = 7.515$; $P = 38.978$; $F = 13.598$.

The sample grade of mass known m after activation with a count number C emitted is obtained by:

$$y = \frac{\left[(\text{C-F}) * \text{Q} * a^2 - \text{K} * \text{m} * \text{P}\right] \pm \sqrt{\left[\text{K} * \text{m} * \text{P} - (\text{C-F}) * \text{Q} * a^2\right]^2 + 4 * (\text{K} * \text{m} * \text{Q}) * \left[(\text{C-F}) * \left(\text{P} * a^2 + \text{m} * a\right)\right]}}{2 * \text{K} * \text{m} * \text{Q}}$$

This equation is illustrated in Fig. 3, and the comparison is shown in Fig. 4. Note the linearity of the response and the validity of the model for all range of grades.

6 Conclusions

A neutron activation process from two basic and simultaneous phenomena has been modeled. The phenomena are the reduction of the neutron flux that activates the sample and the attenuation of gamma rays produced during the activation that reach the detector.

The phenomenon of the reduction of the neutron flux is controlled by the inverse of the square of the distance between the sample and the source.

The phenomenon of the attenuation of gamma rays is controlled by an exponential law that depends on the attenuation coefficient μ, characteristic of sample.

The model was assayed by way of irradiation from a fluorspar sample whose fluorine content is reflected by the emission of high- energy gamma radiation (6.13 MeV).

From the correlation of the results with the models and the experiments, it follows that only the effect of reduction of the neutron flux with the distance can be considered as the effect of the other effect is low.

A high correlation coefficient (~ 1) has been obtained from the model. In addition the parameter values *a, K, P, Q* and *F* are consistent with the real values.

After comparison between the values obtained from chemical analysis and those from neutron activation, the model was considered suitable.

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