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Flux variation related uncertainty in neutron activation analysis

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ABSTRACT

The half-life of the product of the nuclear reaction applied to monitor the neutron flux has significant influence on the uncertainties of derived quantities of an investigated sample, such as the reaction rates, isotopic composition. If the flux is supposed to be constant during the irradiation, but its value varies between an upper and a lower limit, a so-called flux variation error occurs in the derived quantities. This error has been quantitatively analysed regarding various irradiation times, decay constants of the monitor foils and the nuclides of interest. Practical formulae have been elaborated, which can be used to handle this type of uncertainty.

1. Introduction

Neutron activation analysis (NAA) [1–3] is a broadly used method for the analysis of material compositions. Of the various nuclear parameters, which are necessary to determine or retrieve from data libraries during the analysis of a sample, the knowledge of the value of the neutron flux is an essential point both in the absolute and the relative method. One of the passive flux monitoring techniques is the so-called multiple foil activation technique [4], where a small piece of material is irradiated, and, subsequently, the neutron flux can be estimated by the measurement of the activity of the reaction products. However, the variation in time of the neutron flux during the irradiation can influence the uncertainty of the measured quantities.

Sima [5] described the effects of the reactor noise on NAA results by applying the Markovian type autocorrelation function of the neutron flux density. This theory is useful to investigate the processes having a time scale similar to that of the flux fluctuations, which usually take place on a short time scale. Flux variations having a longer time scale were investigated by Jacimovic et al. [6], who introduced correction factors of NAA results to consider the linear decrease of the flux. In the current paper, the longer time scale was also under the scope by allowing the flux to alter between predefined upper and lower limits. With the formulae presented here the so-called flux variation error (FVE) can be estimated if the flux is supposed to be constant during the irradiation. The analysis of this area was mostly induced by the investigation of the charged particle activation analysis applied for determining the tritium production rate in the Test Blanket Modules [7] of the ITER Tokamak [8]. A detailed discussion about the theory of that passive diagnostics method and its experimental demonstration are presented in Ref. [9].

2. Uncertainties in the estimation of reaction rates

In NAA, a routine task is to do a quantitative analysis of an unknown sample. Generally, this is carried out in such a way that the sample is irradiated with neutrons together with a so-called monitor foil, then both of them are measured with a detector. The monitor foil is used to determine the neutron flux. It is required that the sample and the monitor foil are exposed to the same neutron spectrum and the same neutron fluence, otherwise corrections need to be applied (self-shielding, flux gradient, etc.). In the following it has been shown that if these basic requirements are fulfilled, at least one factor still remains in this method concerning the time dependency of the flux, which can cause a false measurement.

2.1. Approximations for measuring the neutron flux

Let us start from the differential equation which describes the time evolution of a reaction product

$$dN_1 = -\lambda_1 N_1(t)\,dt + \overline{\sigma}_1 N_{10}\phi(t)\,dt, \tag{1}$$

where

$$\overline{\sigma}_1 = \int \psi(E)\sigma_1(E)\,dE, \tag{2}$$

and it is supposed that $\Phi(E,t) = \psi(E)\phi(t)$ with the constraint $\int \psi(E)\,dE = 1$. λ_1 is the decay constant, N_1 is the number of reaction products, N_{10} is the number of target atoms, $\overline{\sigma}_1$ is the flux weighted cross-section of the reaction, and $\phi(t)$ is the time dependent and $\psi(E)$ is the energy dependent part of the neutron flux ($\Phi(E,t)$). In this case

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the activity of the reaction product can be obtained by the solution of Eq. (1)

$$A_1(t_i) = \lambda_1 \overline{\sigma}_1 N_0 e^{-\lambda_1 t_i} \int_0^{t_i} \phi(t) e^{\lambda_1 t}\, dt + C_0 e^{-\lambda_1 t_i}, \tag{3}$$

where t_i is the irradiation time and set $C_0 = 0$, i.e. $A_1(t_i = 0) = 0$. As a general approach $\phi(t) = \phi_{ref}$ is supposed and formula

$$\tilde{A}_1(t_i) = \overline{\sigma}_1 \phi_{ref} N_{10} (1 - e^{-\lambda_1 t_i}) \tag{4}$$

is used to determine ϕ_{ref} through the measurement of the activity of the reaction product, A_1. In the following, index "1" denotes the monitor foil. In this case, the reaction rate is assigned as $R_1 = \overline{\sigma}_1 \phi_{ref} N_{10}$. Usually, in further calculations ϕ_{ref} is applied, having an uncertainty of $D(\phi_{ref})$, but the error of the derived quantities is reliable if the initial approximation, i.e. $\phi(t) = \phi_{ref}$ was correct. Otherwise, if the value of $\phi(t)$ can vary between ϕ_{min} and ϕ_{max} during the irradiation, then a so-called flux variation error (FVE) is caused, which needs to be considered in the calculation of the error propagation as well.

The change in the flux value can occur due to several reasons. Let us analyze some of them. The simplest case is when the rabbit (sample transit) system changes the sample position undesirably during the irradiation. In any case, if applying a rabbit system, the flux gradient along the transport path should always be considered. It is especially important for the analysis of short-lived nuclides. It also exercises influence on the error, if the foil position and the sample position are not the same and the time dependence of the flux is different at the two places. In case of a neutron generator the flux can rapidly change due to the variation of the deuteron beam, the heat load on the target or the shut-down of the device. So, by irradiating with a neutron generator, the instantaneous flux value can be a bit higher than its average or sometimes can drop to zero level.

The following theory discusses this flux variation error, depending on the lower and upper extrema of the flux and the investigated decay constants.

2.2. Model for estimating the flux variation error

Consider the problem of determining a reaction rate based on the measurement of the activity of the reaction product where the flux is estimated by a monitor foil, which has been irradiated along with the investigated sample. Denote the decay constant of the monitor foil and of the investigated reaction product by λ_1 and λ_2, respectively. Usually, due to external reasons, during the irradiation the neutron flux value can be constrained between an upper, $\phi_{\max}$, and a lower limit, $\phi_{\min}$. The highest FVE in derived quantities is caused if the flux value takes its maximum at the beginning of the irradiation and then takes its minimum until the end of the irradiation (see Fig. 1a) or, if it takes its minimum at the beginning of the irradiation and then takes its maximum until the end of the irradiation (see Fig. 1b). However, the flux is estimated by applying Eq. (4) (constant flux). This behaviour is induced by the $e^{\lambda t}$ weighting function in Eq. (3). In both cases presented in Fig. 1a, b the time point of the alteration in the flux value can be determined ($t_i - t_-$ and $t_i - t_+$, respectively) with the constraint that such a total flux results in the same activity of the monitor foil as would an equivalent constant flux, ϕ_{ref} do.

First look at the case shown in Fig. 1a. According to Eq. (3) the activity of the monitor foil can be calculated as

$$A_1(t_i) = \lambda_1 \overline{\sigma}_1 N_{10} e^{-\lambda_1 t_i} \left[\int_0^{t_i - t_-} \phi_{max} e^{\lambda_1 t}\, dt + \int_{t_i - t_-}^{t_i} \phi_{min} e^{\lambda_1 t}\, dt \right]. \tag{5}$$

By substituting $A_1(t_i)$ with Eq. (4) one can express t_-, which is

$$t_- = -\frac{1}{\lambda_1} \ln \left[\frac{\phi_{ref}(1 - e^{-\lambda_1 t_i}) - \phi_{\min} + \phi_{\max} e^{-\lambda_1 t_i}}{\phi_{\max} - \phi_{\min}} \right]. \tag{6}$$

Let us transform the fluxes into units of ϕ_{ref}:

$$t_- = -\frac{1}{\lambda_1} \ln \left[\frac{(1 - e^{-\lambda_1 t_i}) - \phi'_{\min} + \phi'_{\max} e^{-\lambda_1 t_i}}{\phi'_{max} - \phi'_{\min}} \right], \tag{7}$$

where $\phi'_{\min} = \phi_{\min}/\phi_{ref}$ and $\phi'_{\max} = \phi_{\max}/\phi_{ref}$. Now $t_i - t_-$ gives the time point where the maximum flux value drops to its minimum. t_+ in case of Fig. 1b can be calculated in the same way

$$t_+ = -\frac{1}{\lambda_1} \ln \left[\frac{(1 - e^{-\lambda_1 t_i}) - \phi'_{\max} + \phi'_{\min} e^{-\lambda_1 t_i}}{\phi'_{\min} - \phi'_{\max}} \right]. \tag{8}$$

For obtaining the investigated reaction rate, R_2, one needs to measure the activity of its product having a different decay constant from λ_1

$$A_2^I(t_i) = \lambda_2 \overline{\sigma}_2 N_{20} e^{-\lambda_2 t_i} \left[\int_0^{t_i - t_-} \phi_{\max} e^{\lambda_2 t}\, dt + \int_{t_i - t_-}^{t_i} \phi_{\min} e^{\lambda_2 t}\, dt \right], \tag{9}$$

where the same equation is used as in Eq. (5) with the flux characteristics shown in Fig. 1a. But the flux is supposed to be constant, so the expected activity and reaction rate according to

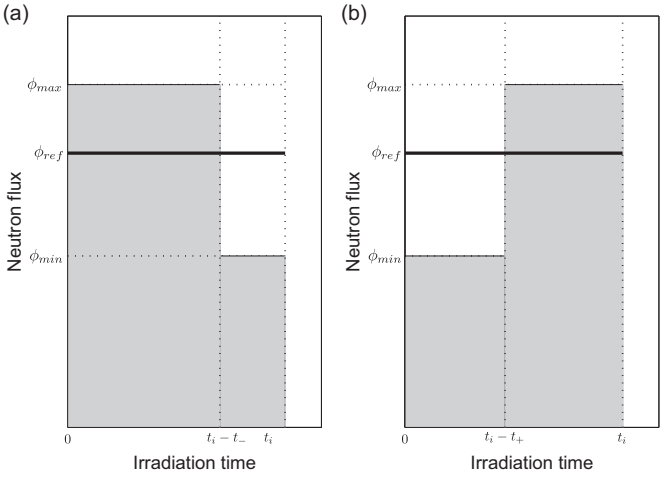

Fig. 1. An illustration of two different flux shapes as a function of time which can be equivalent with a constant, ϕ_{ref}, from the point of view of the activity of the monitor foil at the end of the irradiation. Values are in arbitrary units. The flux limits are $\phi_{\min}$ and $\phi_{\max}$.