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Use of thermal neutron reflection method for chemical analysis of bulk samples ☆

A. Papp [a,*], J. Csikai [a,b]

[a] Institute of Nuclear Research of the Hungarian Academy of Sciences, (ATOMKI), 4001 Debrecen, Pf. 51, Hungary
[b] Institute of Experimental Physics, University Debrecen (IEP), 4010 Debrecen-10, Pf. 105, Hungary

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ABSTRACT

Microscopic, σ_β, and macroscopic, Σ_β, reflection cross-sections of thermal neutrons averaged over bulk samples as a function of thickness (z) are given. The σ_β values are additive even for bulk samples in the z=0.5–8 cm interval and so the $\sigma_{\beta mol}(z)$ function could be given for hydrogenous substances, including some illicit drugs, explosives and hiding materials of ~1000 cm^3 dimensions. The calculated excess counts agree with the measured $R(z)$ values. For the identification of concealed objects and chemical analysis of bulky samples, different neutron methods need to be used simultaneously.

1. Introduction

The development of fast, non-destructive methods for inspection of cargo containers, trucks and airline baggage as well as for the identification of anti-personnel landmines (APMs) is still required. There are more than 60 million landmines buried in 62 countries. Certain types of APMs contain plastic explosives without or with a low level of metal. Transportation of illicit drugs has shown an increasing trend during the last decade. Various neutron based techniques used for bulk hydrogen analysis and the detection of anti-personnel landmines have been developed under the Co-ordinated Research Programs (CRPs) of the IAEA that started in 1997 and 1999, respectively [1,2]. Some results achieved in this international collaboration were described in a Special Issue of ARI in 2004 [3] and in a survey on the status of the detection of concealed objects [4].

The neutron reflection method has had a leading role among the instrumental analytical techniques not only as moisture gauge and hydrogen analyzer [5] but recently also in the detection and identification of anti-personnel landmines (APMs), illicit drugs and explosives [6]. The atom fractions of the major elements, in particular the C/O, C/H and C/N ratios, are quite different in drugs and explosives as compared to other materials used to hide them. The H content in the plastic APMs is around 24–35%, which suggests detecting these objects via the hydrogen. Investigations have shown that the chemical composition and dimension of the reflectors influence strongly the number of backscattered neutrons which reach saturation at about 8–10 cm thicknesses of the samples using our measuring arrangement [7]. Therefore, it was worthwhile to study the $\sigma_\beta(z)$ and the $\sigma_{\beta mol}(z)$ functions in order to apply them for chemical analysis of bulky samples. Though the reflection cross-section is a semi-exact concept it is assumed that, for a given source–sample–detector arrangement, the σ_β and the $\sigma_{\beta mol}$ values are related to the properties of the samples to be investigated. In addition it was found that the reflection cross-sections are additive [8].

The aims of the present investigations are as follows: 1) to check the proposed [9] analytical expression to describe the increment of the reflected thermal neutrons; 2) to determine the microscopic, $\sigma_{\beta mol}(z)$, and macroscopic, $\Sigma_{\beta mol}(z)$, reflection cross-sections averaged over some bulk elements and compounds as reference data; 3) to estimate the $\sigma_{\beta mol}(z)$ values for some illicit drugs, explosives and hiding materials and also the $R(z)$ values, i.e. the expected excess counts; and 4) to compare the advantages and limitations of the neutron methods used for bulk media assay.

2. Measuring methods

A portable thermal neutron reflection equipment (Bitatron) [7] based on a point like Pu–Be neutron source of 18.5 GBq and a small BF_3 counter placed on the surface of a moderator together with the reflector substance was used for the determination of H content of various samples (motor oils, crude oils, vegetable oils, metals, alloys, dummy landmines, hydrocarbons, organic and

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* Corresponding author.
E-mail address: papppa@atomki.hu (A. Papp).

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inorganic compounds, etc.) of different thicknesses [10]. The optimal arrangement of the moderator–detector–reflector and the cadmium shields rendered the determination of the excess counts of reflected neutrons, $R(z)=[(I(z)-I_0)/I_0]$ measured with ($I(z)$) and without (I_0) a sample of about $\pm 5\%$ precision possible [11].

Different hand-held BF_3 neutron detectors were used [4] for the detection of APMs by the observation of anomaly in the yield of reflected thermal neutrons. The test objects were embedded in dry sand of 1×1 m^2 and a thickness of 0.35 m.

In addition, a neutron-gamma surface gauge has also been used for the detection of the reflected neutrons and gamma-rays originated from sand and air as surrounding materials around ~10 cm diameter and 7 cm thickness test objects.

A Pulse Height Response Spectrometer (PHRS) based on a 5×5 cm^2 cylindrical NE213 scintillator completed with neutron/gamma pulse shape discrimination has been used for the measurements of spectral yields [$Y(E_n)$] of elastically backscattered (EBS) Pu–Be neutrons for a number of samples [12,13]. The knowledge of the spectral yield and the leakage spectra [14] of neutrons versus incident energy is indispensable for the calculation of the reaction rate at a given point of the sample or averaged over the total interrogated region. Typical backscattered spectra of Pu–Be neutrons for 4 cm slabs of graphite, water and 5.6 cm thick liquid nitrogen as compared to the calculated reaction rates [15] are shown in Fig. 1. Details of the leakage spectrum measurement methods are described e.g. in Ref. [16].

The increment of reflected thermal and 1.45 eV resonance neutrons as a function of sample thickness has also been investigated [9].

The effects of the source–sample–detector geometries on the bulk hydrogen analysis using epithermal neutrons were also studied [17].

A systematic investigation has been carried out on the possible use of the 14 MeV neutrons and the characteristic gamma rays (2.23, 4.44, 10.8 and 6.13 MeV from H, C, N and O, respectively) for the multi-elemental analysis of bulk samples [18].

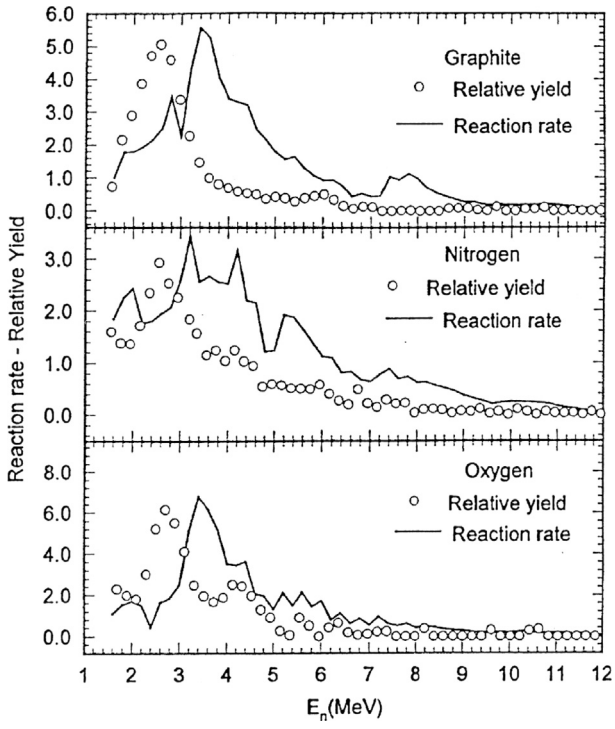

Fig. 1. Measured backscattered spectra (○) of Pu–Be neutrons and the calculated reaction rates (–) for C, N, and O.

Hydrogenous and graphite moderators were applied for the determination of the flux perturbation factor both for thermal and epithermal neutrons [19].

A special irradiation facility for bulk hydrogen analysis based on the activation method using thermal and epithermal neutrons has been tested for cylindrical samples of 8 cm diameter and 10 cm height [20]. Neutrons were produced by four Am–Be sources of 740 GBq total activity. The experimental arrangement is shown in Fig. 2.

Investigations on the possible use of thermal neutron activation analysis of bulky samples of unknown composition were carried out both with hydrogenous and graphite moderators [21].

3. Results

In our previous paper [6] it was shown that the increment of the excess count, $R(z)$, produced by the reflected thermal neutrons as a function of the thickness of the sample (z), can be given by the following expression:

$$R(z)=R_0[1-\exp(-N\sigma_\beta z)]=R_0[1-\exp(-\Sigma_\beta z)] \quad (1)$$

where Σ_β is the "so called" macroscopic reflection cross-section [8] for a given material. The values of Σ_β were deduced from the experimental data obtained for the $R(z)$ function. The analytical form of the $R(z)$ function was determined also from the experimental data taking the average values of the Σ_β parameters related to the increment in the reflected neutrons.

As shown in Fig. 3 the shape and magnitude of the measured excess counts for thermal neutrons can be approximated by Eq. (1) both for elements and compounds. These values were accepted as reference data for the calculation of $R(z)$ functions in the case of unknown samples. The reproducibility of the data points for the Bitatron measurements does not exceed $\pm 10\%$.

On the basis of Eq. (1) it is expected that the $\sigma_\beta(z)$ values should decrease exponentially with the thickness of the reflector materials, i.e.

$$\sigma_\beta(z)=\sigma_\beta(0)\exp(-kz) \quad (2)$$

This expression is recommended for the calculation of the $\sigma_{\beta mol}(z)$ values and the relative excess counts [$R(z)$] for hydrogenous bulk samples. The reference $\sigma_\beta(z)$ functions with their numerical parameters are shown in Fig. 4. The $\sigma_{\beta mol}(z)$ functions are given by [8] the following expression:

$$\sigma_{\beta mol}(z)=\sum_i n_i\sigma_{\beta i}(z)=R/CN_{mol} \quad (3)$$

where N_{mol} is the number of molecules/cm^2 in the target and n_i is the number of atoms of type i with cross-section $\sigma_{\beta i}$ of the molecule.

These data for major elements H, C, N and O of plastic explosives and drugs were determined from the $R(z)$ functions of carbon (C), water (H_2O), paraffin (CH_2) and ammonium nitrate (NH_4NO_3) by using the reflection method. A comparison of the measured and calculated $R(z)$ functions also rendered the determination of $\sigma_\beta(z)$ values for nitrogen possible. Data measured in this experiment for different samples are summarized in Table 1.

The R_0 and Σ_β values deduced from Eq. (1) were accepted for the calculation of the $\sigma_\beta(z)$ functions used for the estimation of the $\sigma_{\beta mol}(z)$ data for samples of unknown compositions. The R_0, Σ_β, $\sigma_\beta(0)$ and k values are given in Table 1. Using these R_0, Σ_β data in Eq. (3), the measured $R(z)$ functions could be reproduced for the 2–6 cm thicknesses of the samples. The data are summarized in Table 1: the measured and calculated yields of reflected neutrons were found to be within 10%. This result obtained for the first time has a great importance in the chemical analysis of bulky samples.