Bulk media analysis using neutrons

Theses

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Introduction

These theses are the summary of my work which I started as an undergraduate student in 1999, continued as a PhD student between September 1999 and August 2002 and finished as a predoctor in June 2003 at the Institute of Experimental Physics, University of Debrecen. My supervisor was prof. dr. Julius Csikai.

The topic of my dissertation is bulk media analysis by neutrons. The dissertation contains our results in the field of the measurement of neutron reflection cross section and flux-perturbation factor and a comparison of hydrogenous and graphite moderator in the use of activation analysis. (The International Atomic Energy Agency started an international research program for bulk hydrogen analysis in 1997 because of its scientific and practical importance.[*])

These measurements were carried out at the Institute of Experimental Physics, University of Debrecen, except the irradiations in the graphite pile and measurements of absolute activity which were executed in cooperation with T. Sanami and T Michikawa at the High Energy Accelerator Organization (KEK), Radiation Science Center in Tsukuba, Japan.

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1. Neutron reflection cross section

Introduction

The concept of neutron reflection cross section (σ_β) was introduced by Csikai and Buczkó in 1999 [1]. It is a microscopic parameter which characterizes backscattering properties of samples. The reflection cross section is based on the concept of albedó (β) which is a macroscopic parameter and was introduced by Amaldi and Fermi in 1936 [2].

The neutron reflection cross section is not a geometry-independent parameter. It depends on constituents of the sample and on the measuring arrangement (Fig. 1). If I and I_0 denote the number of counts in a unit time with and without the sample, we can put the following for $R = (I - I_0)/I_0$:

$$R = C\frac{n\sigma_\beta}{S} = CNd\sigma_\beta \qquad (1)$$

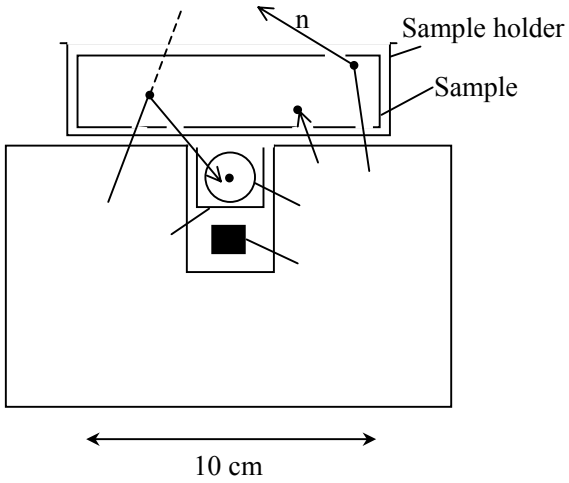
[*] Bulk hydrogen analysis, using neutrons, IAEA/PS/RCM97-1, Vienna, 1997

where $C = 10^{-24}$ cm^2/barn is a factor, n, σ_β (barn), S (cm^2), N (atom/cm^3) and d (cm) are the number of atoms in the sample, the neutron reflection cross section, the surface of the sample looking onto the detector, the number of atoms in a unit volume of the sample and the thickness of the sample, respectively. One should pay attention to choose the right thickness of the sample, especially if it consists of elements having high absorption cross section (Cl, Co, Ag, Hg) or if it consists of some metals (Zn, Fe, Cu) or compounds of these elements, because σ_β is constant only below a certain thickness (~1 cm) of the sample.
The neutron reflection cross section for a molecule is

$$\sigma_{\beta mol} = \sum_i n_i \sigma_{\beta i} \qquad (2)$$

where n_i and $\sigma_{\beta i}$ are the number and the reflection cross section of atoms type i in the molecule.

The neutron reflection cross section of an element is a constant part of the Maxwell-averaged elastic scattering cross section. The ratio in our arrangement shown in Fig. 1 is: $\sigma_\beta / \sigma_{EL} = 0.59$.



According to the method developed by Buczkó et al. in 1975 [7], the quantity

$$\eta = \frac{R}{\rho} = \frac{1}{\rho}\frac{I - I_0}{I_0} \qquad (3)$$

(ρ – sample-density) is a linear function of H-content (m%) of the sample.

Results

Measurement of neutron reflection cross section
by activation foil

- We measured the neutron reflection cross section of some elements and compounds by Cu activation foil instead of a BF_3 counter in order for the geometry to be more unambiguous. If A and A_0 denote activity of the foil with and without the sample, eq. (1) is valid for $R = (A - A_0)/A_0$.
- We checked the linear dependence of $\eta - H\,(m\%)$ function using hydrocarbons (CH), CHO compounds and water. Using this result and eq. (2) for hydrogen a value of $\sigma_{\beta,H} = 34.4 \pm 1,2$ barn was obtained.
- We quantified the reflection cross section for carbon, oxygen, aluminum and zinc. These quantities are about two times higher than those measured by a BF_3 counter because of the difference between arrangements and detector-materials.

Relative measurement of neutron reflection cross section
by a BF_3 counter

- We developed a new method to measure reflection cross sections in a relative way. We considered the neutron reflection cross section of CH_2 molecule as a reference. The method is based on our experience that in the case of a polyethylene foil placed in the position ½ d in the sample (that simulates foil distributed uniformly in the sample very well) the $R - d_p$ is a linear function in the range studied, i.e.:

$$R = sd_p + R_T \qquad (4)$$

where s, d_p and R_T are the slope, the thickness of the polyethylene layer and the excess counts originated from the pure sample of thickness δ (Fig. 2).

- Let the thickness of the polyethylene $d_e = |R_T/s|$ be called equivalent thickness. Equivalent thickness is the thickness of the polyethylene foil at which the excess counts originated from the polyethylene in the presence of the sample equals the excess counts originated from the pure sample of the given thickness. This is not the same as the thickness of pure polyethylene having equal excess counts because of the different slope.

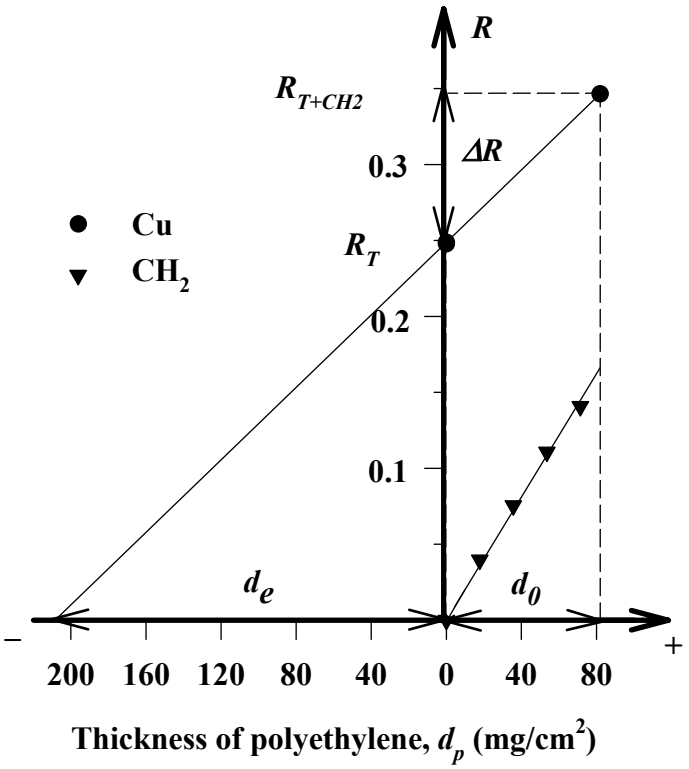


Fig. 2 The concept of equivalent thickness

• The following formula can be deduced from the concept of equivalent thickness:

$$\sigma_{\beta,T} = \frac{\rho_{CH2} d_e}{\rho_T \delta} \frac{M_T}{M_{CH2}} \sigma_{\beta,CH2} \qquad (5)$$

where $\sigma_{\beta,T}$, ρ_T, M_T are the reflection cross section, density and molar mass concerning the sample material, $\sigma_{\beta,CH2}$, ρ_{CH2}, M_{CH2} are the same quantities regarding the polyethylene, δ and d_e are the thickness of the sample and the equivalent thickness.

• We can quantify the neutron reflection cross section of atoms or molecules forming the given sample relatively after determining equivalent thickness using (5) and the neutron reflection cross section of polyethylene: $\sigma_{\beta,CH2} = 37.6 \pm 1.0$ barn as a reference.

• We quantified the neutron reflection cross sections of many elements in a relative way (as it was discussed above) and in some cases using the absolute method [1]. For the ratio of σ_β and the Maxwell-averaged elastic scattering cross section σ_{EL} we obtained: $\sigma_\beta / \sigma_{EL} = 0.6 \pm 0.02$. Once we know this ratio and σ_β, σ_{EL} can easily be given if it cannot be found in data libraries.

• If we know the reflection cross sections the equivalent thickness $d_{e,X}$ can be calculated, regarding any elements or compounds X as a reference, using the following equation:

$$\rho_X d_{e,X} = \frac{M_X}{M_{CH2}} \frac{\sigma_{\beta,CH2}}{\sigma_{\beta,X}} \rho_{CH2} d_{e,CH2} \qquad (6)$$

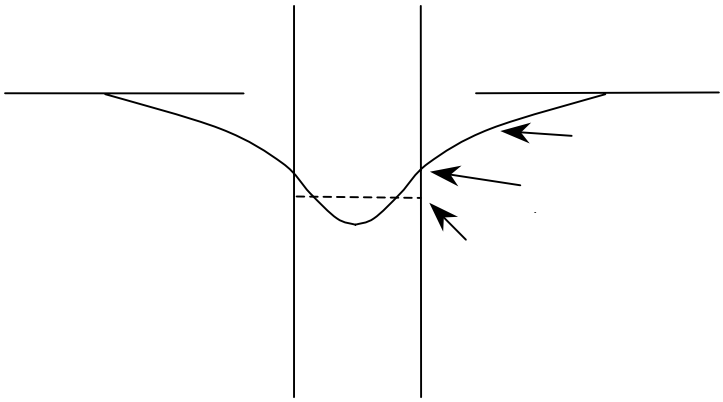
from which $\rho_{H2O} d_{e,H2O} = 1.30 \cdot \rho_{CH2} d_{e,CH2}$ in the case of water,
$\rho_H d_{e,H} = 0.16 \cdot \rho_{CH2} d_{e,CH2}$ in the case of hydrogen.

• Eq. (6) gives the relation between equivalent thicknesses of water and of polyethylene (which is known) if, for example we put water as a reference material (X = H_2O) into the sample of thickness δ. This could be useful mainly if we do not want our sample to have contact with water but we need the calibration line for a water-content analysis. Excess counts regarding the pure sample (R_T) and the equivalent thickness of water as a reference material are sufficient for determining the line (Fig. 2).

2. Flux distribution

Introduction

It is often essential to know the flux distribution in a moderator or bulk sample as exactly as possible to apply neutrons for analysis. This problem can be solved most simply by using detector foils. However, the measuring tools can change the quantity to be measured and modify the results of measurements (Fig. 3), therefore it is necessary to introduce some correction factors.



The foil depletes its neighborhood due to its higher absorption cross section than that of the surrounding material. This is called flux-depression and can be characterized by the flux-depression factor H:

$$H = \phi_s / \phi_0 \tag{7}$$

where ϕ_s and ϕ_0 denote neutron flux on the surface of the foil and unperturbed flux in the absence of the foil, respectively.

In the foil itself the external layers allow fewer neutrons to get into the internal layers so the foil shields itself. This is called self-shielding and can be characterized by the self-shielding factor G:

$$G = \langle \phi \rangle / \phi_s \tag{8}$$

where $\langle \phi \rangle$ is the average flux in the foil:

$$\langle \phi \rangle = \frac{1}{d} \int_0^d \phi(x) dx \tag{9}$$

d and $\phi(x)$ are the thickness of the foil and the flux distribution in the foil, respectively.

The total flux perturbation can be characterized by the flux-perturbation factor F:

$$F = \langle \phi \rangle / \phi_0 \tag{10}$$

which is the product of the self-shielding and the flux-depression factors:

$$F = G \cdot H . \tag{11}$$

As in the case of bulk samples the unperturbed flux against position is not constant, $\langle \phi_0 \rangle$ should be substituted for ϕ_0 in eqs. (7) and (10).

According to Judd [12], $H = 1$ in the range of resonance neutrons if resonances are narrow and are sufficiently separated in energy.

It is not a simple problem to calculate the self-shielding and flux-depression factors but it is not impossible if we know all the parameters in the formulae. Sometimes, however, (especially in activation analysis) the composition of the sample is unknown (since we would like to determine this).

In 1978 Buczkó and Borbély developed a method to determine the average thermal neutron flux in absorbing samples using thin foils [18]. According to

their results thermal neutron flux distribution can be described by the following function:

$$\phi(x) = \left(ax^2 + bx + c\right)^{-1} \tag{12}$$

where x denotes the distance from the surface of the foil, while a, b and c are fitting parameters.

According to Khalil [20], epithermal neutron flux distribution can be described by

$$\phi(x) = \left(a_4x^4 + a_3x^3 + a_2x^2 + a_1x + a_0\right)^{-1} \tag{13}$$

where a_i (i = 0, 1, 2, 3, 4) are fitting parameters.

Results

- We determined the active center of the Pu-Be source which was used in our measurements. It is 1.5 mm below the geometrical center.

Factors F, G and H in thin samples

- We determined the neutron flux distribution and factors F, G and H in thin In and Au foil stacks in the case of thermal and epithermal neutrons, in ε-caprolactam and graphite moderators.
- In our measurements $H \neq 1$ in the case of epithermal neutrons, because the shape of the absorption excitation function of In and Au in epithermal energy range does not fulfill the requirements given in Judd's paper [12].
- According to our results, factors F, G and H can be given in the case of thermal neutrons against stack thickness by the following formulae:

$$F(d) = F_0e^{-fd}, \tag{14}$$

$$G(d) = G_0e^{-gd}, \tag{15}$$

$$H(d) = H_0e^{-hd} \tag{16}$$

where F_0, G_0 and H_0 (=1) belong to thickness $d = 0$, while f, g and h are fitting parameters.

- In the case of epithermal neutrons the exponential relation is valid only in the case of the flux-depression H.

Factors *F*, *G* and *H* in bulk samples

- We determined the neutron flux distribution and factors *F*, *G* and *H* in bulk graphite, ε-caprolactam [$(CH_2)_5CONH$], PVC (C_2H_3Cl), Plexiglas ($C_5H_8O_2$), iron and aluminum samples, in ε-caprolactam and graphite moderators using In, Au and Dy detector foils.
- Fitting the measured points, eq. (12) did not describe flux distribution properly in a number of cases. This occurred only in hydrogenous samples (ε-caprolactam, PVC, Plexiglas), if difference from the surface to the center of the sample was significant (1.3 – 4.4 times). In these cases we used simple quadratic equation for fitting.

Ratio of thermal and epithermal neutrons

- We determined ratios of thermal and epithermal neutron fluxes in ε-caprolactam, PVC and Plexiglas samples using In detector foils.
- The ratio of the averaged thermal and epithermal neutron flux is 6.5 in ε-caprolactam, 7.0 in Plexiglas but 2.7 in PVC because Cl that it contains, absorbs thermal neutrons.

Comparison of moderators

- The advantages and limitations of using the graphite pile ($250 \times 190 \times 190$ cm^3) and the cylindrical ε-caprolactam (diameter: 30 cm, height: 30 cm) as moderators are discussed.
- Thermal neutron flux as a function of distance from the source changes less in a graphite moderator than in a hydrogenous one. So it seems to be more practical to use a graphite moderator in the analyses of bulk samples. The change of thermal flux is only 6% in graphite from 10 cm to 20 cm from the source while it is 1/13 in a water moderator.
- The values of the flux-perturbation factor *F*, are closer to 1 in PVC, iron and aluminum samples using either an Al or a Cd sample holder in the case of a graphite than in the case of an ε-caprolactam moderator.
- In the case of thin samples the flux-perturbation factor against stack thickness changes less in an ε-caprolactam moderator than in a graphite moderator.
- The ratio of thermal and epithermal neutrons is high in an ε-caprolactam moderator, i.e. the number of epithermal neutrons is small compared to thermal ones. So it is not practical to use epithermal neutrons in activation analysis.
- As a conclusion we can state that it is more efficient to use thermal neutrons instead of epithermal neutrons in activation analysis, irradiating thin samples in a hydrogenous, while bulk samples in a graphite moderator.

Irodalomjegyzék / References

[1] J. Csikai, Cs. M. Buczkó: The concept of the reflection cross section of thermal neutrons, Applied Radiation and Isotopes, 50, 1999, pp. 487-490

[2] E. Amaldi, E. Fermi: On the absorption and the diffusion of slow neutrons, Physical Review, 50, 1936, pp. 899-928

[3] Csikai Gyula, Daróczy Sándor: Termikus neutronok albédójának vizsgálata, Magyar Fizikai Folyóirat, VII. kötet, 1959, 507-516. oldal

[4] Julius Csikai: Handbook of fast neutron generators, Volume 1, CRC Press Inc, Boca Raton, Florida, 1987, pp. 173-174

[5] JEF Report 14, Table of simple integral neutron cross section data from JEF-2.2, ENDF/B-VI, JENDL-3.2, BROND-2 and CENDL-2, NEA OECD, France, 1994

[6] J. Csikai: Bulk media assay using backscattered neutron spectrometry, Proceedings of the 14th Workshop on Radiation Detectors and Their Uses, S. Sasaki and T. Shibata editors, Tsukuba, Japan, 1-3 February, 2000, pp. 236-246

[7] M. Buczkó, Z. Dezső, J. Csikai: Determination of the bitumen content in asphalt concrete using a neutron reflection method, Journal of Radioanalytical Chemistry, 25, 1975, pp. 179-183

[8] S. M. Al-Jobori, S. Szegedi, Cs. M. Buczkó: Determination of hydrogen content in crude oil by the neutron reflection method, Radiochemical and Radioanalytical Letters 33 (3), 1978, pp. 133-138

[9] A. Sola: Flux perturbation by detector foils, Nucleonics 18 (3), 1960, pp. 78-81, 141

[10] Neutron fluence measurements, Technical reports series No. 107, J. Moteff editor, IAEA, Vienna, 1970, pp. 54-58, 106-111, 126-132

[11] M. W. Thompson: Some effects of the self-absorption of β-rays and neutrons in neutron detecting foils, AERE-RP/R 1549

[12] A. M. Judd: Neutron flux measurements by resonance activation of foils, Nuclear Instruments and Methods, 23, 1963, pp. 29-35

[13] W. Bothe: Zur Methodik der Neutronensonden, Zeitschrift für Physik, 120, 1943, pp. 437-449

[14] C. W. Tittle: Slow-neutron detection by foils I-II, Nucleonics, 8 (6), 1951, pp 5-9 and 9 (1), 1951, pp. 61-67

[15] K. M. Case, F. de Hoffmann, G. Placzek: Introduction to the theory of neutron diffusion, Los Alamos Scientific Laboratory, U. S. Government Printing Office, Washington, D. C. 1953, p. 153

[16] E. Martinho, I. F. Gonçalves, J. Salgado: Universal curve of epithermal neutron resonance self-shielding factors in foils, wires and spheres, Applied Radiation and Isotopes, 58, 2003, pp. 371-375

[17] Szabó E., Simonits A.: Aktivációs analízis, Műszaki Könyvkiadó, Budapest, 1973, 114-117. oldal

[18] Cs. M. Buczkó, A. Borbély: A simple method for the determination of the average neutron flux for absorbing samples, Journal of Radioanalytical Chemistry, 42, 1978, pp. 393-398

[19] Julius Csikai: Handbook of fast neutron generators, Volume 1, CRC Press Inc, Boca Raton, Florida, 1987, pp. 137-142

[20] G. I. Khalil: Determination of U, Th, Au, Mn and Na in geological samples by nuclear methods, Thesis for candidate degree, Debrecen, 1985, pp. 69-72

[21] Cs. M. Buczkó, M. H. Al-Mundheri, J. Csikai, Z. Dezső: ^{252}Cf neutron irradiation facilities, Nuclear Instruments and Methods, 134, 1976, pp. 101-105

[22] J. Csikai, B. Király, T. Sanami, T. Michikawa: Studies on thermal neutron perturbation factor needed for bulk sample activation analysis, Nuclear Instruments and Methods A, 488, 2002, pp. 634-641

[23] E. Browne, R. B. Firestone: Table of Radioactive Isotopes, V. S. Shirley editor, John Wiley and Sons Inc., New York, 1986

Függelék / Appendix

Az értekezés témakörében megjelent publikációk / Publications concerning the dissertation

1. **B. Király**, J. Csikai: Investigations on thermal neutron reflection by activation method, Applied Radiation and Isotopes, 52, 2000, pp. 93-96

2. J. Csikai, **B. Király**, Cs. M. Buczkó: Targets for bulk hydrogen analysis using thermal neutrons, 20th World Conference of the INTDS, Targets for Particle Beams: Preparation and Use, October 2-6, 2000, Antwerp, Belgium, Lecture, Abstracts: paper 33, Nuclear Instruments and Methods A, 480, 2002, pp. 166-170

3. **B. Király**, J. Csikai, R. Dóczi: Validation of neutron data libraries by differential and integral cross sections, 2000 Symposium on Nuclear Data, November 16-17, 2000, JAERI Tokai, Japan, Abstracts: Poster session P31, Proceedings (JAERI-Conf 2001-006, INDC(JPN)-188/4) pp. 283-288

4. J. Csikai, **B. Király**, T. Sanami, T. Michikawa: Studies on thermal neutron perturbation factor needed for bulk sample activation analysis, Nuclear Instruments and Methods A, 488, 2002, pp. 634-641

5. **B. Király**, T. Sanami, R. Dóczi, J. Csikai: Detection of explosives and illicit drugs using neutrons, 5th International Topical Meeting on Industrial Radiation and Radioisotope Measurement Applications (IRRMA-V), 9-14 June 2002, Bologna, Italy, Lecture, Abstract Book p. 24, accepted for publication in the proceedings (NIM B)

6. **B. Király**, T. Sanami, J. Csikai: Advantages and limitations of thermal and epithermal neutron activation analysis of bulk samples, Applied Radiation and Isotopes, 58, 2003, pp. 691-695

Hivatkozások / Citations

H. R. Vega-Carrillo, C. Torres-Muhech: Low energy neutrons from a (PuBE)-Pu-239 isotopic neutron source inserted in moderating media, Revista Mexicana de Fisica, 48, 2002, pp. 405-412

D. K. Mohapatra, C. P. Reddy: Use of neutron reflection method to assay nuclear materials is solutions, Applied Radiation and Isotopes, 55, 2001, pp. 693-696