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Monte Carlo calculation of epithermal neutron resonance self-shielding factors in wires of different materials

I.F. Gonçalves, E. Martinho, J. Salgado

Instituto Tecnológico e Nuclear, Estrada Nacional 10, 2686-953 Sacavém, Portugal

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Abstract

Epithermal neutron resonance self-shielding factors in wires of materials used as activation detectors or as targets for radionuclide production have been calculated using the MCNP code. The energy dependent self-shielding factor depends on the ratio scattering/capture cross sections. The self-shielding factors for cobalt and gold have been compared with available values. The self-shielding factor depends on various physical and nuclear parameters. However, an adimensional variable can be adopted that describes the self-shielding factors of different materials by a quasi "universal curve". © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Epithermal neutrons; Resonance self-shielding factor; MCNP code

1. Introduction

Most of the materials used in neutron dosimetry or for radionuclide production for medical purposes exhibit resonances in the epithermal region of the reactor neutron spectrum. This implies that, due to the self-shielding effect, the neutron flux experiences significant perturbation inside the material. Consequently, interpretation of the sample activation requires the application of resonance self-shielding factors. However, the elementary models for resonance self-shielding factor calculations in the epithermal region generally fail. In fact, these models assume that the energy loss in elastic scattering is sufficient for shifting the neutron out of resonance, and multiple scattering is ignored. However, the maximum energy loss in scattering may be comparable to or less than the resonance width and the scattering cross-section may be higher than the capture cross-section. In this situation other approaches need to be used. Lopes (1991) developed a method that includes the effect of multiple scattering inside the sample. Monte Carlo methods can calculate directly the self-shielding factor taking into account the multiple scattering.

The aims of this work are (a) to establish a calculation method for the resonance self-shielding factor in wires useful for multiple purposes, by using the MCNP code (Briesmeister, 1997) and updated cross-sections (ENDF-B6), and (b) to interpret its dependence on several physical and nuclear parameters. To validate the methodology the results are compared with available values obtained by other authors.

2. Methodology of calculation

The resonance self-shielding factor, $G_{res}(R)$, in wires of radius R, is defined as the ratio between the reaction rates per atom in the real sample and in a similar and infinitely diluted sample. Thus:

$$G_{res}(R) = \frac{\int_{E_1}^{E_2} \Phi(E)\sigma_{n\gamma}(E)\,dE}{\int_{E_1}^{E_2} \Phi_0(E)\sigma_{n\gamma}(E)\,dE}, \quad (1)$$

where $\Phi_0(E) \propto E^{-1}$ is the original, non-perturbed, epithermal neutron flux per unit energy interval inside the infinitely diluted sample, $\Phi(E)$ represents the perturbed epithermal neutron flux inside the real sample, $\sigma_{n\gamma}(E)$ denotes the (n, γ) cross-section, and E_1 and E_2 are, respectively, the lower and the upper limits around

E-mail address: ifg@itn.pt (I.F. Gonçalves).

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the resonance energy E_{res}. The total neutron cross-section has been adopted in the calculation of the perturbed neutron flux $\Phi(E)$, which takes into account the neutron scattering in the sample. A fictitious density of $10^{-6}\rho_0$ has been adopted in the simulation of infinite dilution to calculate the non-perturbed reaction rate, ρ_0 representing the density of the real sample.

For a given radius R, a neutron energy dependent resonance self-shielding factor, $G_R(E)$, can be defined as

$$G_R(E) = \frac{RR(E,\rho_0)}{RR(E,10^{-6}\rho_0)}, \quad (2)$$

where $RR(E,\rho_0)$ and $RR(E,10^{-6}\rho_0)$ are the reaction rates for the energy E, corresponding to the densities ρ_0 and $10^{-6}\rho_0$, respectively. It must be noted that E is the energy at which the neutron is absorbed; if the resonance scattering cross section is higher than the absorption cross section, a neutron entering the wire with a energy $E' > E$ can suffer scattering processes before its absorption at the energy E. This definition is different from that given by Lopes and Molina Ávila (1990), where E is the energy of the incident monoenergetic neutron beam.

3. Results and discussion

The resonance self-shielding factor depends on material density, atomic mass, natural abundance, wire radius and in especially on the resonance cross section.

Table 1 shows the values of the density, ρ_0, natural abundance, θ, atomic mass, A (Tuli, 2000), resonance energy, E_{res}, scattering, capture and total resonance cross-sections, σ_s, σ_γ, σ_{tot} (ENDF-B6) and the resonance widths, Γ_n, Γ_γ and $\Gamma(\Gamma = \Gamma_n + \Gamma_\gamma)$ (Mughaghab et al., 1981; Mughaghab, 1984), for the studied elements.

In order to define the density corresponding to "infinite dilution" for the calculation of the non-perturbed reaction rate, the dependence of the reaction rate on the density was first studied. Fig. 1 shows the variation of the reaction rate per atom of gold (this material has the highest resonance total cross section of the studied elements and a high density) of a wire with the radius $R = 1$ mm as a function of the density. The reaction rate increases as the density decreases and it saturates for $\rho/\rho_0 \leqslant 10^{-4}$. For this reason, in all calculations the density for infinite dilution was assumed to be $\rho = 10^{-6}\rho_0$.

The effect of self-absorption is clearly demonstrated in Fig. 2, where the perturbed and the non-perturbed neutron fluxes, and the corresponding reaction rates for a gold wire of $R = 1$ mm are depicted. For an infinite diluted sample the neutron flux varies as $1/E$, and the reaction rate shows a well-defined peak. On the other hand, the perturbed flux has a corresponding valley and the reaction rate shows a small broad peak.

Fig. 3 shows the energy dependent self-shielding factor of indium for different values of the wire radius. The minimum of the valley is observed at the resonance energy; for $R = 1$ mm, $G_R(E)$ has its lowest value. As the radius decreases, the effect of shielding diminishes and the curves become narrower. For indium, $\sigma_\gamma > \sigma_s$, the absorption predominates over the scattering and then $G_R(E) < 1$; the curve is practically symmetrical around E_{res}. Similar results were obtained for gold and rhenium, both of which are high neutron absorbers. On the other hand, as can be seen in Fig. 4, for manganese, which is a good scatterer ($\sigma_\gamma < \sigma_s$), the self-shielding factor $G_R(E) > 1$, for $E < E_{res}$. Multiple scattering processes are responsible for this effect. In fact, a neutron entering the wire with energy $E \approx E_{res}$ can suffer one or more scattering interactions before capture, and is absorbed at $E < E_{res}$. The peaks appear at energies $E < E_{res} - \Gamma$. As would be expected the energy at the peak decreases as the radius increases. The effect is more pronounced for manganese, which has a ratio Γ_n/Γ higher than those of the other neutron scatterers, cobalt

Table 1
Some physical and nuclear properties of the studied elements

Element	Nuclide	ρ_0 (g cm^{-3})	θ	A (g)	E_{res} (eV)	σ (E_{res}) (b)			Γ_{res} (eV)		
						$\sigma\gamma$	σs	σ_{tot}	Γ_γ	Γ_n	Γ
Cobalt	Co-59	8.9	1	58.9	132	8.49×10^2	9.52×10^3	1.04×10^4	0.47	5.15	5.62
Copper	Cu-63	8.96	0.692	63.5	579	4.12×10^2	5.05×10^2	9.17×10^2	0.485	0.59	1.075
Gold	Au-197	19.3	1	197.0	4.91	2.74×10^4	3.4×10^3	3.08×10^4	0.124	0.015	0.139
Indium	In-115	7.3	0.957	114.8	1.46	2.80×10^4	1.16×10^3	2.92×10^4	0.072	0.003	0.075
Manganese	Mn-55[a]	7.3	1	54.9	336	46	3.24×10^3	3.29×10^3	0.435	22.0	22.4
	Mn-55[b]				1098	28	1.41×10^3	1.44×10^3	na[c]	15.4	na[c]
Rhenium	Re-185	21	0.374	186.2	2.16	2.33×10^4	1.22×10^3	2.45×10^4	0.055	0.003	0.058

[a] First resonance.
[b] Second resonance.
[c] Not available.

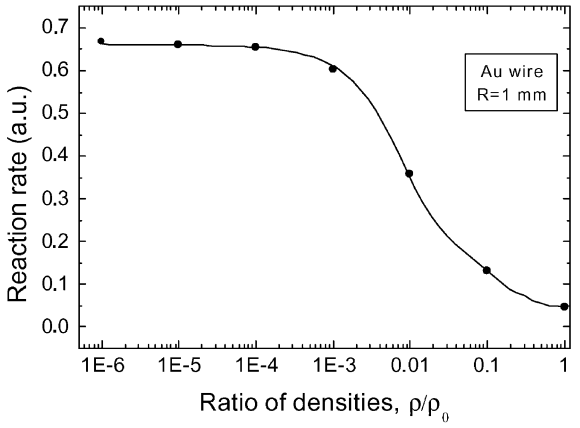

Fig. 1. Reaction rate versus the ratio "diluted density"/real density for a gold wire ($R=$ 1 mm).

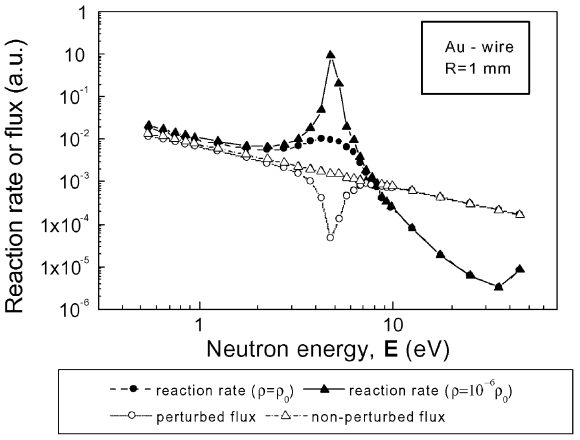

Fig. 2. Dependence of the (n, γ) reaction rate and neutron flux on the neutron energy for a gold wire ($R=$ 1 mm).

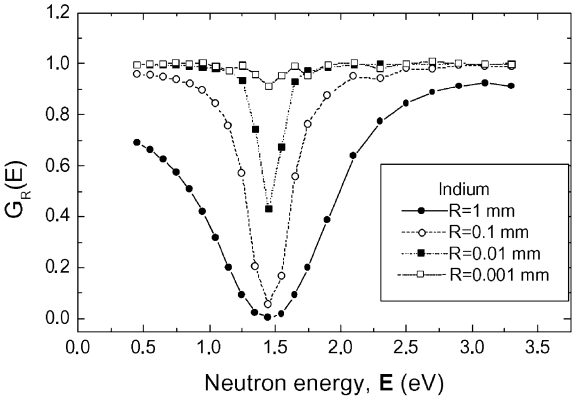

Fig. 3. Variation of the energy dependent resonance self-shielding factor of indium wire of different radii with neutron energy.

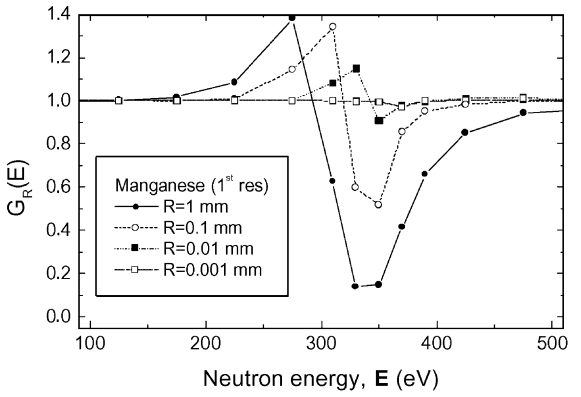

Fig. 4. Variation of the energy dependent resonance self-shielding factor of manganese (first resonance) wire of different radii with neutron energy.

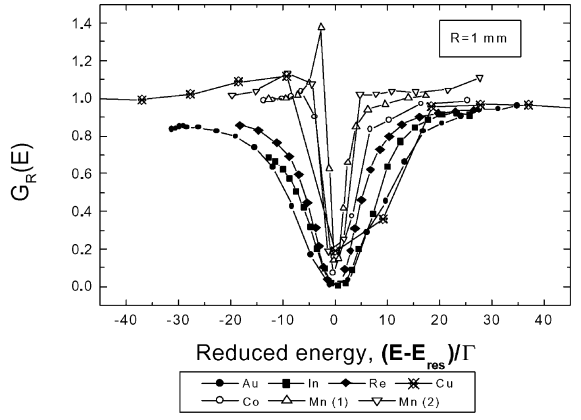

Fig. 5. Variation of the energy dependent resonance self-shielding factors of gold, indium, rhenium, copper, cobalt and manganese wires ($R=$ 1 mm) with the reduced neutron energy.

and copper. Note that if monoenergetic neutrons enter the wire of a high scatterer material with $E > E_{res}$ they can suffer multiple scattering and then can be absorbed when the energy reaches a value $E \approx E_{res}$. $G_R(E)$ is greater than unity for $E > E_{res}$ and a maximum will be observed just above E_{res}. This effect has been observed by Lopes and Molina Ávila (1990).

Fig. 5 shows G_R (E) as a function of a "reduced energy", $E_{red} = (E - E_{res})/\Gamma$ for $R = 1$ mm. Gold, indium and rhenium have higher capture cross sections than the other elements: the corresponding $G_R(E)$ reveal pronounced valleys and the curves are wider. Similar results were obtained for other radii.

Fig. 6 compares the self-shielding factor, G_{res} (R), of gold and cobalt calculated in this work with published values. The values calculated in this work are systematically smaller than those previously published. For cobalt, there is a reasonable agreement; the values of

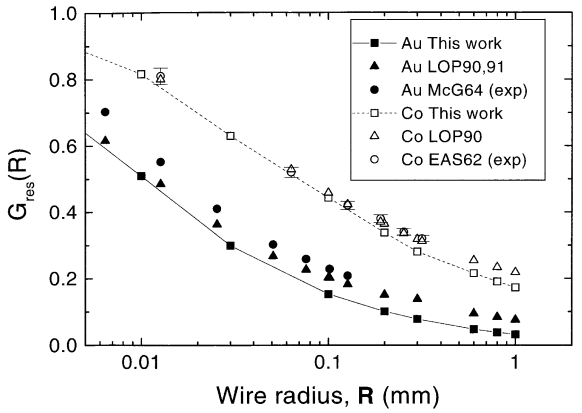

Fig. 6. Comparison of resonance self-shielding factors of gold and cobalt calculated in this work with values (experimental and calculated) published by other authors. LOP90, 91 (Lopes, 1990, 1991), McG64 (McGarry, 1964), EAS62 (Eastwood, 1962).

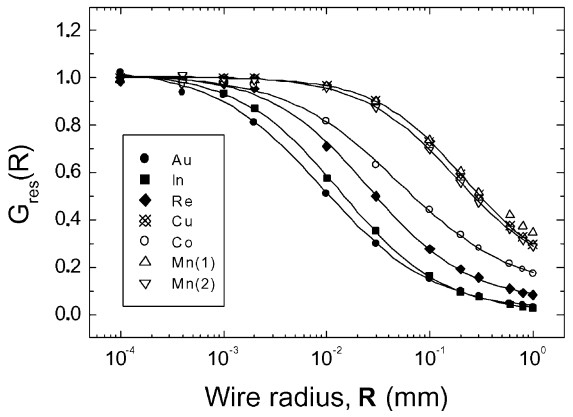

Fig. 7. Dependence of the resonance self-shielding factors of gold, indium, rhenium, copper, cobalt and manganese on the wire radius. The symbol Mn(i) means the ith manganese resonance.

gold reported by McGarry (1964) are significantly higher. The experimental factors were obtained from the ratio of the count rate of cobalt and gold wires relative to the count rate in 0.1% Co–Al alloy wire (Eastwood and Werner, 1962) and 0.272% Au–Ni alloy wire (McGarry, 1964). Eastwood corrected the experimental ratios to isolate the resonance self-shielding factors by using some approximations. These approximations can explain the differences between our results and those of Eastwood. McGarry (1964) considered a 0.272% Au–Ni alloy as an infinitely dilute sample. However, results of the present work (see Fig. 1) show that this is not the case. Applying a correcting factor of 0.78 to the experimental values of McGarry, a good agreement with our results is obtained.

The calculated self-shielding factor, G_{res} (R), as a function of the wire radius is shown in Fig. 7. The behaviour of the curves is similar for all materials. However, the decrease of $G_{res}(R)$ as the radius increases is faster when the resonance cross-section and /or the density is higher (gold, indium and rhenium). Cobalt has an intermediate cross-section and its $G_{res}(R)$ lies among the values of the other materials.

In a tentative attempt to analyse all the results, an adimensional variable, z, was introduced, defined as

$$z = \Sigma_{tot} \times R, \tag{3}$$

Σ_{tot} being given by

$$\Sigma_{tot} = \frac{\rho_0}{A}\,\theta N_A \sigma_{tot}(E_{res}), \tag{4}$$

where N_A is the Avogadro number.

Fig. 8 shows the resonance self-shielding factor as a function of z. The curve was adjusted to the calculated values of the high neutron absorbers (gold, indium, rhenium and copper). In spite of the great ranges of variation of σ, ρ and A, the proposed variable reduces the curves corresponding to high neutron absorbers to a quasi "universal curve", which enables one to predict the self-shielding factor for a new element. The agreement fails for elements with dominating scattering cross-sections. In fact, the scattering process shifts the neutron energy to a lower value, before capture occurs. Better agreement is obtained with a new adimensional variable, z', defined as

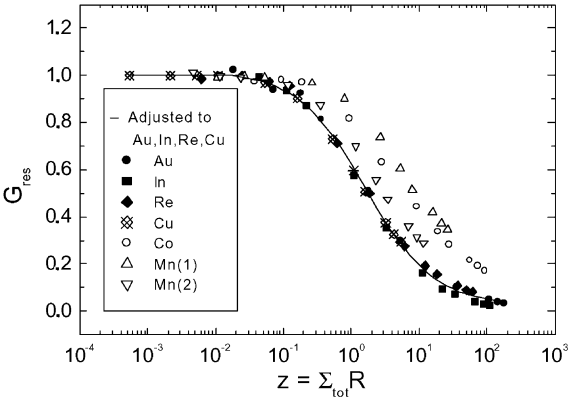

Fig. 8. Dependence of the resonance of self-shielding factors of gold, indium, rhenium, copper, cobalt and manganese on the adimensional variable $z = \Sigma_{tot} \times R$. The symbol Mn($i$) means the ith manganese resonance.

$$z' = z \times \left(\Gamma_\gamma/\Gamma\right)^{1/2}. \tag{5}$$

Fig. 9 represents the self-shielding factor of all the studied elements as a function of z'. It seems that all values can be described by an unique curve. The relative errors in the z' and G_{res} values are estimated to be about

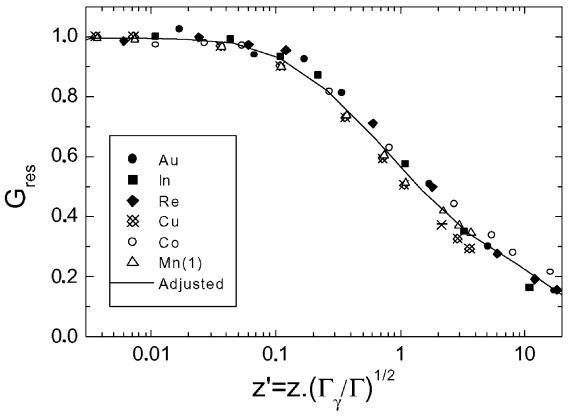

Fig. 9. Dependence of the resonance self-shielding factors of gold, indium, rhenium, copper, cobalt and manganese on the adimensional variable $z' = \Sigma_{tot} \times R \times (\Gamma_\gamma/\Gamma)^{1/2}$. The symbol Mn(1) means the first manganese resonance.

5%. In experimental work, the range of z' is limited to $0.003 < z' < 20$. For $z' < 0.003$, the resonance self-shielding $G_{res} = 1$ and it is not necessary to perform any correction. For $z' > 20$, $G_{res} < 0.15$, the self-absorption in the sample is high and the reaction is very low. Within this range of values, the following equation was adjusted to the calculated points:

$$G_{res}(\log z') = 0.567 - 0.464 \log z' + 0.0318(\log z')^2 + 0.148(\log z')^3 - 0.00849(\log z')^4 - 0.0372(\log z')^5 - 0.00788(\log z')^6 \quad (6)$$

with $r^2 = 0.991$.

4. Conclusions

The energy dependent self-shielding factor and the self-shielding factor for gold, indium, rhenium, copper, cobalt and manganese wires of different radii have been calculated. The energy dependent self-shielding factors of materials with high capture cross-sections have different behaviour than those of materials with high scattering cross-sections. In the first case, $G_R(E) < 1$ for the whole energy interval; on the other hand, in the second case, $G_R(E) > 1$ for $E < E_{res}$ because neutrons entering the wire with energy E_{res} can suffer one or more scattering interactions before capture at a lower energy can occur.

It was shown that an adimensional variable can be introduced, which converts the dependence of the resonance self-shielding factor on physical and nuclear parameters to a quasi "universal curve" valid for all materials. Further work is in progress for foils.

By the time the work was carried out, MCNP version 4C had been released. Some simulations have shown a negligible effect on the results.

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