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Enhancement of thermal neutron self-shielding in materials surrounded by reflectors

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Abstract Materials containing from 41 to 1124 mg chlorine and surrounded by polyethylene containers of various thicknesses, from 0.01 to 5.6 mm, were irradiated in a research reactor neutron spectrum and the ^{38}Cl activity produced was measured as a function of polyethylene reflector thickness. For the material containing the higher amount of chlorine, the ^{38}Cl specific activity decreased with increasing reflector thickness, indicating increased neutron self-shielding. It was found that the amount of neutron self-shielding increased by as much as 52% with increasing reflector thickness. This is explained by neutrons which have exited the material subsequently reflecting back into it and thus increasing the total mean path length in the material. All physical and empirical models currently used to predict neutron self-shielding have ignored this effect and need to be modified. A method is given for measuring the adjustable parameter of a self-shielding model for a particular sample size and combination of neutron reflectors.

Keywords Neutron activation · Neutron self-shielding · Neutron scattering · Neutron reflection

Introduction

Neutron activation of materials in research reactors is widely used for radioisotope production and for neutron activation analysis. For a given radionuclide, the amount of activity produced by the (n,γ) reaction is proportional to the average neutron flux inside the target or sample. The activity produced is also proportional to the thermal neutron activation cross-section as shown in Eq. 1.

$$A = N_0\,\sigma_{th}\,\phi_{th}(G_{th} + G_{ep}Q_0/f)\left(1 - \mathrm{e}^{-\lambda t_i}\right) \quad (1)$$

where N_0 is the number of target nuclei, $Q_0 = I/\sigma_{th}$, with I the resonance integral, σ_{th} the thermal neutron activation cross-section, λ the decay constant, t_i the irradiation time, $f = \varphi_{th}/\varphi_{ep}$, φ_{th} and φ_{ep} are the average unperturbed thermal and epithermal fluxes inside the sample and G_{th} and G_{ep} are the thermal and epithermal self-shielding factors. However, the activity produced is not linear with the number of target nuclei because the neutron self-shielding factors, G_{th} and G_{ep}, decrease as the number of absorbing nuclei increases.

Many methods have been developed to calculate the thermal neutron self-shielding factor for cylindrical samples. They include Monte-Carlo calculations [1–3], and analytical formulas [2–11]. The analytical formula of Chilian et al. [9–11], a sigmoid function fashioned after that of Martinho et al. [2], is the following:

$$G_{th} = \frac{1}{1 + \left(\frac{N_{Av}k_{th}}{r\,(r+h)}\sum_i \frac{m_i\,\sigma_{abs,i}}{M_{at,i}}\right)^{0.964}} \quad (2)$$

with

N_{Av}	Avogadro's number
k_{th}	thermal self-shielding constant, a value of 0.81 was suggested [9–11]
r, h	radius and height of cylinder
m_i	amount of element i (g)
$\sigma_{abs,i}$	thermal neutron absorption cross-section for element i
$M_{at,i}$	atomic mass of element i

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Most of the analytical formulas use the macroscopic thermal neutron absorption cross-section. The Monte-Carlo calculations take into account neutron scattering in the sample, as do explicitly some of the analytical formulas [2, 3, 7, 8]. A recent comparison [3] of three of the analytical formulas revealed that the amount of self-shielding calculated for the same sample could differ by more than 50%. The reason for this was not known [3] but more recent discussions have led to speculate that the reason may be that the empirical formulas were developed under different experimental conditions, with varying influence of neutron reflection from materials surrounding the sample. Previously, the effect of these reflected neutrons was always thought to be small, at most contributing less than 5% to the amount of self-shielding [9, 10], but considering the widely varying amounts of self-shielding recently observed in various laboratories for similar samples [11, 12, D. Bossus private discussion], it was decided to investigate whether reflected neutrons could be the reason.

We should recall the definition of the thermal neutron self-shielding factor which characterizes an absorbing sample. In this work we adopt the commonly used definition: the thermal neutron capture reaction rate per atom in an absorbing sample relative to that in a similar but infinitely diluted non-absorbing sample irradiated under similar conditions. We may think of the non-absorbing sample as the flux monitor. An equivalent statement of the definition is: the average thermal neutron flux inside the absorbing sample relative to that inside the flux monitor. Of course, the purpose of the flux monitor is to estimate the neutron flux that prevails inside other samples to be activated. The reaction rate per atom in an absorbing sample may be different from that in the flux monitor for reasons other than self-shielding. For example, the absorbing sample and flux monitor may be in different containers and the container may absorb neutrons or moderate neutrons, changing epithermal neutrons to thermal neutrons. These two effects of the containers should not be confused with the self-shielding in the absorbing sample. Therefore, the absorbing sample and the flux monitor should be irradiated in similar containers. Also, in poorly thermalised neutron spectra and with an absorbing sample containing a large amount of neutron moderator, especially hydrogen, neutron moderation in the sample may increase the thermal neutron flux [13]; this effect is not usually thought of as self-shielding.

In the past, the thermal neutron self-shielding factor has also been defined as the average thermal neutron flux inside the sample relative to the average flux at the surface of the sample or relative to the flux incident on the sample [6]. These definitions divide the problem into two parts: self-shielding inside the sample and flux depression in the vicinity of the sample. However, these definitions are impractical because the only way to measure the flux at the surface of the sample, the unperturbed flux reduced by the flux depression, is to place small monitors at the surface of the sample, which is seldom done except for very large samples [14]. With our definition, the flux monitor is conveniently irradiated before or after the absorbing sample in reactors where the neutron flux is reproducible, or simultaneously at a few mm distance [11] where there is no flux depression.

Experimental

Two thermal neutron absorbing samples, of volume 2.1 cm^3, were prepared by mixing $CaCl_2$ powder with SiO_2 powder. The first sample, called the flux monitor, contained 0.064 g $CaCl_2$ and 2.911 g SiO_2. The second sample contained 1.762 g $CaCl_2$ mixed with 0.979 g SiO_2. It was called the absorbing sample because it contains a large amount of Cl, and Cl has a thermal neutron absorption cross-section of 33.5 b, much greater than those of Ca, Si or O, 0.43 b, 0.17 b and 0.06 b, respectively. Using the self-shielding model of Chilian et al. [9–11], it was estimated that the thermal neutron self-shielding factor would be 0.985 for the flux monitor and 0.732 for the absorbing sample. The two samples were sealed in thin polyethylene bags. For each irradiation, the two samples were placed in 14 mm diameter by 14 mm high aluminum or polyethylene containers. They were irradiated repeatedly on several different days, for 180 s each time, in irradiation sites no. 6 and no. 8 of the Ecole Polytechnique SLOWPOKE reactor, using the pneumatic irradiation systems. Between irradiations the samples were kept in sealed polyethylene containers to avoid the accumulation of H_2O by the $CaCl_2$ powder. The thermal neutron flux in both irradiation sites was approximately 3×10^{11} s^{-1} cm^{-2} and it is known to be reproducible to about 1% from day to day [15]. The neutron spectra are fairly well thermalised, with $f = 48.6$ and 52.7 for sites 6 and 8, respectively [16]; thus, less than 1.8% of the ^{38}Cl activity is produced by epithermal neutrons.

The two samples were irradiated one immediately after the other, both surrounded by the same amount of neutron reflecting material, either 0.4 mm of aluminum or various thicknesses of polyethylene, 0.6, 1.2, 2.5, 3.8, 5.1 or 5.6 mm. For the same thickness, there are far fewer reflected neutrons with aluminum than with polyethylene; the macroscopic thermal neutron scattering cross-sections are 0.09 and 6.8 cm^{-1} for aluminum and polyethylene, respectively. In irradiation site 6 the samples were contained in 26 mm diameter irradiation vials (rabbits) and the 14 mm diameter samples were maintained on the axis of the irradiation vial and 6 mm from the bottom using thin cardboard spacers. The irradiation geometries for four of

the cases are shown in Fig. 1. The aluminum irradiation tube has an outside diameter of 38.1 mm in site 6 and 22.2 mm in site 8. The water reflector is therefore much closer to the sample in site 8.

To measure the ^{38}Cl activity, the samples in their plastic bags were counted for 600 s at a distance of 210 mm from the face of a 33% efficiency germanium semiconductor detector after decay times of about 20 m for the flux monitor and about 180 m for the absorbing sample so that the dead-time would be approximately 10% in both cases. The ^{38}Cl peak areas and specific activities, corrected for dead-time and corrected to zero decay time, were calculated with the EPAA software [17].

Results and discussion

Figure 2 shows the measured specific activities of the flux monitor and the absorbing sample as a function of polyethylene reflector thickness. The relative uncertainty in each measured point is estimated to be 1.3%. This includes the contributions from neutron flux variations with time, 0.5%, variation of irradiation time, 0.2%, radial flux-gradient and uncertainty in the positioning of the sample in the irradiation tube, 1.0%, counting geometry variations 0.3%, dead-time correction, 0.2%, counting statistics, 0.3%. The four measured points for site 8 were multiplied by 1.02 to account for the slightly lower neutron flux in site 8 relative to site 6.

It can be seen in Fig. 2 that the activities of the flux monitor irradiated in site 6 do not vary with reflector thickness, which would seem to indicate that the polyethylene reflectors have no effect on the average thermal neutron flux inside the sample. Actually, it is known that there are three effects: absorption of thermal neutrons,

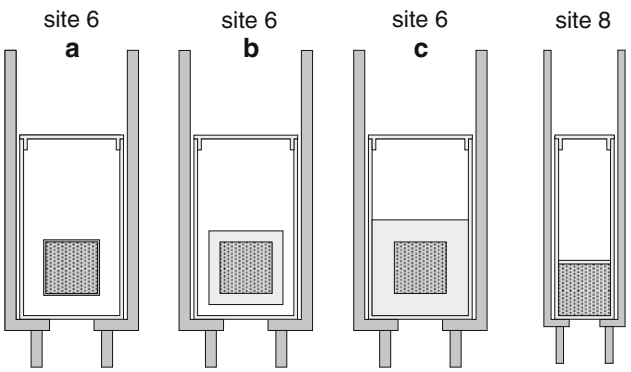

Fig. 1 Some of the irradiation geometries with various neutron reflector thicknesses. 14 mm diameter by 14 mm high sample with **a** 0.4 mm aluminum, **b** 2.5 mm polyethylene, **c** 5.6 mm polyethylene. The aluminum irradiation tubes are surrounded by water reflector

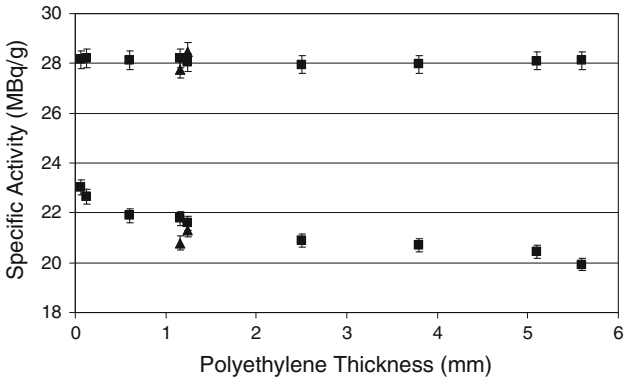

Fig. 2 The measured ^{38}Cl specific activities of the flux monitor *near 28 MBq/g* and the absorbing sample *lower values* as a function of neutron reflector thickness. *Triangles* irradiation site 8. In the case of multiple measurements at the same thickness, the data points are displaced slightly horizontally for clarity

scattering of thermal neutrons towards or away from the sample and moderation of epithermal neutrons to produce more thermal neutrons. In site 1 of the SLOWPOKE reactor, which is less well moderated, $f = 18$, it is known [13] that the third effect dominates and the thermal flux increases with reflector thickness. In site 6, which is better moderated, with $f = 48.6$, the three effects appear to cancel.

As can be seen in Fig. 2, the specific activity of the neutron absorbing sample is always lower than that of the co-irradiated flux monitor, because of neutron self-shielding, and the activity of the absorbing sample decreases from 81.0 to 71.1% of that of the flux monitor as the thickness of the surrounding polyethylene reflector increases from 0 to 5.6 mm. This decreased specific activity of the absorbing sample can only be due to an increase in the amount of neutron self-shielding. The previous Monte-Carlo calculations [1–3] and the commonly used analytical formulas [2–6, 8] take into account neutron scattering in the sample, but not scattering in the materials surrounding the sample since the latter was always considered negligible. Now we see that the effect on the amount of self-shielding can be as much as 52% (from 19.0 to 28.9%). What all these calculations have ignored is the following: when a neutron goes through the sample and comes out, it may reflect off materials surrounding the sample and go back in. It may go through the sample two or three times. Reflections therefore increase the effective path length in the sample and increase the probability of the neutron being absorbed, thus increasing the amount of self-shielding. If this effect increases the amount of self-shielding by 52%, as was seen here, then the average number of times a thermal neutron goes through the sample in the extreme case is at least 1.52 (actually higher because the relation between path length and magnitude of self-shielding is non-linear).

Table 1 Thermal neutron self-shielding factors, G_{th} determined for various polyethylene reflector thicknesses

Irradiation site no.	PE reflector thickness	Ratio specific activities (measured)	G_{th} flux monitor (calculated)	k_{th} model of Chilian et al. (derived)	G_{th} absorbing sample (derived)
6	0 mm	0.810 ± 0.010	0.990	0.549	0.799 ± 0.010
6	0.6 mm	0.780 ± 0.014	0.988	0.669	0.767 ± 0.014
6	1.2 mm	0.773 ± 0.010	0.987	0.695	0.760 ± 0.010
6	2.5 mm	0.750 ± 0.014	0.985	0.797	0.735 ± 0.014
6	3.8 mm	0.739 ± 0.014	0.985	0.846	0.724 ± 0.014
6	5.1 mm	0.728 ± 0.014	0.984	0.900	0.712 ± 0.014
6	5.6 mm	0.711 ± 0.014	0.982	0.981	0.694 ± 0.014
8	1.2 mm	0.752 ± 0.010	0.986	0.786	0.738 ± 0.010

For the neutron absorbing sample irradiated in site 8, the specific activity is 75.2% of that of the flux monitor (mean of two measurements), slightly less than the value of 77.3% for this sample irradiated in site 6 with the same polyethylene reflector thickness. This increase in the amount of self-shielding in site 8 relative to site 6 is thought to be due to the closer proximity of the water reflector in site 8, see Fig. 1, and thus the higher probability of neutrons reflecting back into the sample.

The ratios of measured specific activities were used to determine thermal neutron self-shielding factors. First, the flux monitor was assumed to have $G_{th} = 1$ and the ratio of activities was used as the first trial value of G_{th} for the absorbing sample. Then a small correction for epithermal neutrons was applied using Eq. 1. The corrected value of G_{th} was then used with Eq. 2 to derive a value of k_{th}. This k_{th} was then used with Eq. 2 to calculate an accurate value of G_{th} for the flux monitor. After several iterations of this process the final values were obtained; they are shown in Table 1. It can be seen that the derived values of G_{th} for the absorbing sample are always 1 to 2% lower than the measured ratios of activities. This difference is due to the G_{th} of the flux monitor and the small correction for epithermal neutrons. The thus derived values of k_{th} of the model of Chilian et al. increase from 0.549 to 0.981 as the reflector thickness increases from 0 to 5.6 mm. This 79% increase is interpreted to mean that the average neutron path length in the sample increases by 79% as the amount of neutron reflection increases.

Figure 3 shows the thermal neutron self-shielding factor for a 14 mm diameter by 14 mm high sample calculated by several commonly used analytical self-shielding models: the KAYZERO software package [5], MATSSF [3], the NIST routine [6–8], the Martinho et al. [2] sigmoid function, and the Chilian et al. [9–11] sigmoid function with the suggested parameter $k_{th} = 0.81$, as a function of the amount of chlorine in the sample. Also shown are the value for the flux monitor calculated by the sigmoid function of Chilian et al. with $k_{th} = 0.695$ and some of the values of Table 1 measured in this work for the absorbing sample.

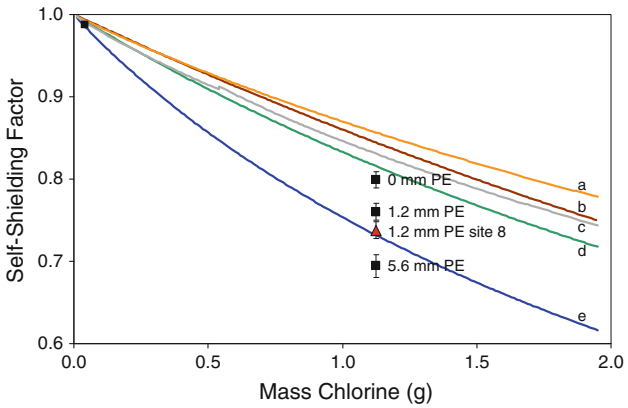

Fig. 3 The thermal neutron self-shielding factor calculated by five commonly used models as a function of the mass of chlorine in a 14 mm diameter by 14 mm high cylinder; *a* KAYZERO, *b* MATSSF, *c* NIST, *d* Martinho et al., *e* Chilian et al. The data point for the flux monitor was calculated and the data points with error bars were measured

It can be seen in Fig. 3 that only the function of Chilian et al. is close to the measured values for the absorbing sample in a polyethylene container. This is because the parameter $k_{th} = 0.81$ [9, 11] was measured for large volume NAA samples in conditions similar to those of the absorbing sample measured in this work. The four other functions were established using theoretical models or Monte-Carlo calculations which ignored reflected neutrons from materials surrounding the sample and they were usually validated using small samples, such as wires, with no polyethylene containers. The model of Martinho et al. [2] comes closer than the other three, possibly because its fitted parameter was determined using a large amount of experimental data and some of them had geometry similar to that of the present work. The amount of self-shielding measured with the absorbing sample of the present work in a typical 1.2 mm thick polyethylene container exceeds the predictions of these four models by 30 to 83%.

If one imagines the case of a very small solid object, such as a wire, irradiated without a polyethylene container,

then the probability of a neutron exiting the object reflecting back in off materials such as the reactor moderator or the pneumatic irradiation system rabbit would be quite small because of the distance and the small size of the object. In this case, models a–d of Fig. 3 would estimate the self-shielding fairly accurately and the sigmoid function of Chilian et al. with $k_{th} = 0.81$ would overestimate the self-shielding, as was pointed out by Trkov et al. [3]. A parameter $k_{th} = 0.5$ would give a better estimation.

It is clear that all the models need to be modified to take into account neutrons reflecting back into the sample from the sample container, the rabbit, the irradiation channel tube and the reflector surrounding the irradiation channel. It is convenient to divide the calculation into two parts: the existing formulas can be used to calculate the self-shielding as a function of sample geometry and chemical composition. The second part is to calculate the effect of reflections outside the sample. With the method of Chilian et al., this simply means determining a new k_{th} for each new sample geometry, container, rabbit and irradiation channel. As was done above for varying thickness of sample container, measurements are carried out with absorbing samples of known composition, surrounded by a given combination of reflectors, and k_{th} is determined by the above iterative procedure if one flux monitor and one absorbing sample are used, or by a least squares fit to the measurements with k_{th} as the adjustable parameter if a larger number of absorbing samples are used.

Conclusions

In a typical fairly large NAA sample enclosed in a typical 1.2 mm thick polyethylene vial, the amount of thermal neutron self-shielding was found to exceed the amounts predicted by four commonly used models by as much as 83%. This enhancement of the self-shielding is explained by the increase in the mean neutron path length in the sample due to a large fraction of neutrons which exit the sample and subsequently reflect back into the sample. All the models need to be modified to take into account reflections from the materials surrounding the sample. A simple procedure has been outlined to measure the k_{th} of the method of Chilian et al. for any irradiation conditions and thus to achieve more accurate self-shielding corrections.

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