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Neutron self-shielding in irradiation channels of small reactors is isotropic

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Abstract In order to predict the self-shielding effect in neutron activation of non-spherical samples in reactor neutron spectra, it is important to know whether the neutron field is sufficiently anisotropic to cause significant variations between horizontal and vertical sample orientation or with orientation relative to the direction towards the reactor core. Metal wires with significant neutron self-shielding were irradiated in several channels of the SLOWPOKE reactor at Ecole Polytechnique Montreal and the TRIGA reactor at the Jožef Stefan Institute. In all cases, the amount of thermal or epithermal self-shielding was found to be identical, within the experimental uncertainty, regardless of the orientation of the wire, indicating that the neutron field is essentially isotropic. Models used to predict neutron self-shielding need to be adjusted accordingly. In our Monte Carlo model, the tube-shaped neutron source was moved back into the moderator and reflecting materials near the sample location were included, which produced an isotropic neutron field at the sample location.

Keywords Neutron activation analysis · Neutron self-shielding · Isotropic · Research reactor

Introduction

Neutron activation analysis is an excellent technique for the chemical analysis of a wide variety of materials over a wide range of concentrations. For materials with high concentrations of elements that strongly absorb thermal and epithermal neutrons, it is necessary to correct the neutron self-shielding effect. To this end, two easy-to-use self-shielding calculation routines, the sigmoid function [1] and MATSSF [2], have recently become available and are still being developed. They are intended for use with the neutron spectrum of any research reactor. However, for the activation of non-spherical NAA samples, the amount of self-shielding may vary depending on the orientation of the sample relative to the direction of the majority of neutrons if the neutron field is non-isotropic. The extreme case is the activation of wires or foils, which are often used for neutron spectrum characterization and neutron fluence monitoring. Using pneumatic irradiation systems, for the irradiation of wires or foil discs with their axis perpendicular to the axis of the rabbit, the orientation of the axis of the wire or foil disc is usually unknown because the rabbit is free to turn on its axis. For this reason, for accurate determination of the amount of self-shielding it is important to know if the neutron field is sufficiently non-isotropic to cause significant differences in the amount of self-shielding in the different cases. In this work, wires with significant thermal or epithermal neutron self-shielding were irradiated in four irradiation channels of two research reactors.

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Experimental

Ecole Polytechnique Montreal SLOWPOKE

At Ecole Polytechnique Montreal, the SLOWPOKE reactor has irradiation channels situated to the side of the fuel, including channel 1 in the annular beryllium moderator/reflector which surrounds the fuel and channel 8 in the water reflector beside the beryllium annulus; a drawing can be found in a previous publication [3]. Channel 1 is near the radial midpoint of the beryllium annulus, where the thermal neutron flux is maximum; the thermal neutron field is therefore thought to be quite isotropic here. For epithermal neutrons in both channels and for thermal neutrons in channel 8, there is a substantial horizontal flux gradient [4], leading to believe that these neutron fields are less isotropic. In fact, the magnitude of the radial flux gradient, up to 1.1% per millimetre, leads us to believe that these neutron fields are probably as anisotropic as those in any small research reactor. Vertical and horizontal wires were irradiated in channels 1 and 8. The vertical wires were necessarily perpendicular to the direction towards the fuel and to the flux gradient.

To study thermal neutron self-shielding, the wires used were an alloy of Ni 59.2%, Cr 16.0%, Fe 23.5%, Si 1.3% and Mn 0.104%. The wires were 1 mm diameter and 10 mm long. They were irradiated five at a time on a wooden support, all centred on the same vertical axis with the two vertical wires at top and bottom and the three horizontal wires between them spaced 5 mm apart and at 60 degree angles. Sets of wires were irradiated for 600 s in channels 1 and 8 and then the ^{56}Mn activity of each wire was measured using a gamma-ray spectrometer with a germanium detector. All wires were counted 100 mm from the detector with exactly the same counting geometry. The counting time was 600 s and at least 250,000 counts were acquired in the ^{56}Mn peak at 847 keV. The ^{56}Mn specific activities were calculated by dividing the activity by the mass of Mn in each wire, as determined by weighing. The thermal self-shielding was calculated by the sigmoid function of Chilian et al. [1] to be 2.69%, i.e. $G_{th} = 0.9731$. Since the amount of self-shielding is small, it was necessary to correct accurately the variations in activity due to flux gradients in order to see the variation due to the non-isotropic neutron field. The vertical flux variation was measured by irradiating a 50 mm long Ni–Cr wire in a separate irradiation, cutting it into pieces and measuring the specific activity of each piece. If any variation due to flux gradient were uncorrected, such as that due to a difference in the angle of the rabbit for the monitor wire and that of the rabbit of the five-wire setup, it would be easily seen as a difference in the activities of the two vertical wires irradiated at the top and bottom. For the three horizontal wires of each series it was desired to know the angle between the axis of the wire and the direction towards the reactor core (direction of steepest flux gradient). A strip of Zr foil was made into a circle and placed at the top of the rabbit. After irradiation, the Zr strip was cut into 8 pieces and their ^{97}Zr specific activities measured; the most active piece was the one nearest the reactor core. The activities of the 8 pieces enabled the determination of the orientation of the rabbit with a precision of about 2 degrees.

To study epithermal neutron self-shielding, the wires used were 98.49% Al with 1.51% Au, 1 mm diameter and 10 mm long. They were irradiated five at a time on the same wooden support as was used for the Ni–Cr wires. Sets of wires were irradiated for 600 s in channels 1 and 8 and then the ^{198}Au activity of each wire was measured. The epithermal self-shielding for ^{198}Au in these wires was calculated by the sigmoid function of Chilian et al. [1] to be 11.1%, i.e. $G_{ep} = 0.889$. In channel 1, with $f = 18.0$, 46% of the ^{198}Au activity was produced by epithermal neutrons and in channel 8, with $f = 52.6$, 21%. The vertical flux variation was measured by activating in a separate irradiation a 50 mm long Al–Au wire and cutting it into pieces, and the measured activities of the five horizontal and vertical wires were corrected for the flux variation and specific activities were calculated by dividing by the mass of Au in each wire.

Jožef Stefan Institute TRIGA

Following the results of previous theoretical investigations [2], indium wire was chosen for the experimental investigation of epithermal self-shielding because it was expected to be the most sensitive to any neutron field anisotropy, if present.

The indium wire with purity of 99.999% (Reactor Experiments, Inc., USA) and diameter of 0.762 mm was cut into pieces of 10 mm length. Two experiments were made, with and without polyethylene (PE) containers. In the first experiment, the two aliquots of about 0.04 g were sealed in the centre of PE ampoules (KARTELL, Italy, diameter of 14 mm and height of 50 mm) and fixed to the bottom orthogonally using cellulose paper (one wire lying flat on the bottom and the other wire pointing vertically upwards). The PE ampoules were inserted into the Al-container (diameter 24.5 mm and height 120 mm) for irradiation. In the second experiment the two In wires were inserted directly into the Al-container fixed to the bottom orthogonally as before, with one horizontal and one vertical wire, using only cellulose paper. The sample pairs were then irradiated for 3 min in the central channel (CC) and the carousel facility (CF) at position IC-40 of the TRIGA Mark II reactor with thermal neutron fluxes of 10×10^{12} and 1.1×10^{12} cm^{-2} s^{-1}, respectively [5, 6].

The activities of the irradiated In wires were measured twice after appropriate cooling times (10 and 24 h) on an absolutely calibrated HPGe detector with 45% relative efficiency. The first measurement was suitable for measurement of ^{116m}In ($T_{1/2} = 54.41$ min, main gamma lines at 416.9, 1097.3 and 1293.5 keV), while the second measurement was appropriate for ^{114m}In ($T_{1/2} = 49.51$ d, main gamma lines at 190.3, 558.4 and 725.2 keV). Additionally, the ^{115m}In ($T_{1/2} = 269.16$ min, gamma line at 336.2 keV) produced by the ^{115}In (n,n′) reaction was measured to check for fast neutron field anisotropy, but within the experimental uncertainty none was observed and the results are not presented. After the second measurement, the vertical wire was cut in two pieces to check for axial flux gradient by measuring again the ^{114m}In gamma lines. It should be mentioned that the second measurement for the vertical wire was performed at the same detector-sample distance (8 cm from the top of the HPGe detector) for the whole wire and the cut-off parts in order to minimize the effects of detector calibration uncertainties.

The HyperLab [7] program was used for net peak area evaluation, while the software package Kayzero for Windows [8] was employed for the specific count rate, effective solid angle and true-coincidence correction calculations.

Results and discussion

Ecole Polytechnique Montreal SLOWPOKE

For thermal neutrons, the measured ^{56}Mn specific activities of the Ni–Cr wires irradiated in channels 1 and 8 are shown in Fig. 1. The ten points on each graph represent two series of five wires. The values for the horizontal wires are plotted at the angle between the axis of the wire and the direction towards the reactor core. For each series of five wires, the specific activities are normalized relative to the mean of the two vertical wires. The precision of the measurements is estimated to be 0.29%. This is the sum of the following contributions, combined in quadrature: weighing 0.13%, homogeneity of Mn in the Ni–Cr wire 0.05%, counting statistics 0.19%, dead-time correction 0.05%, counting geometry reproducibility 0.05%, correction of the flux gradient 0.15%.

The amount of thermal neutron self-shielding was calculated by the sigmoid function [1] to be 2.69% (i.e. $G_{th} = 0.9731$). The changes in specific activity which would be caused by a 10% change in the amount of thermal neutron self-shielding (i.e. 2.42 or 2.96% thermal self-shielding) were also calculated; they are indicated in the figure by the dashed lines. No systematic difference in specific activity between vertical and horizontal wires and no variation with angle between the axis of the wire and the direction towards the reactor core were observed.

For epithermal neutrons, the measured ^{198}Au specific activities of the Al–Au wires are shown in Fig. 2 for three series of wires for channel 1 and two series for channel 8. For each series of five wires, the specific activities are relative to the mean of the two vertical wires. The amount of epithermal neutron self-shielding was calculated by the sigmoid function [1] to be 11.1% (i.e. $G_{ep} = 0.889$). The changes in specific activity which would be caused by a

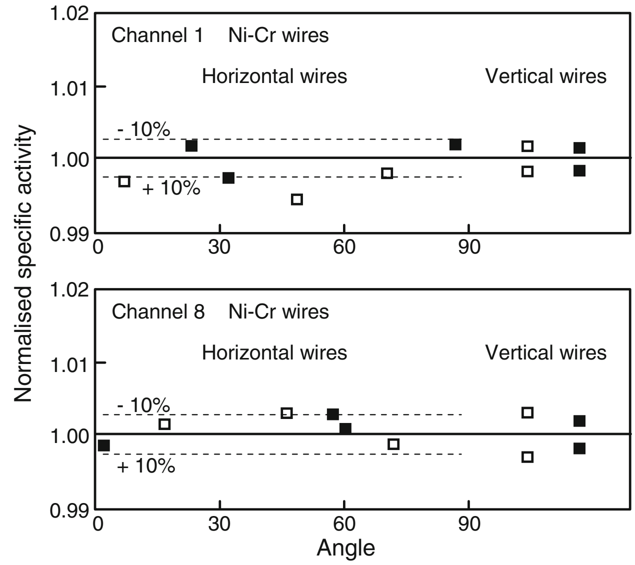

Fig. 1 The measured specific activities of horizontal and vertical Ni–Cr wires irradiated in a SLOWPOKE reactor. *Open squares* the five wires of the first experiment, *closed squares* the five wires of the second experiment. The changes in activity which would be caused by a 10% change in the amount of thermal neutron self-shielding are indicated by the *dashed lines*

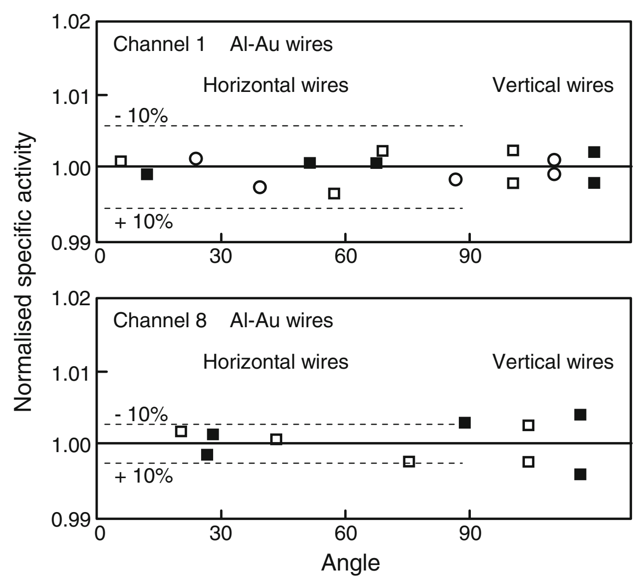

Fig. 2 The measured specific activities of horizontal and vertical Al–Au wires irradiated in a SLOWPOKE reactor. *Open squares* the five wires of the first experiment, *closed squares* the five wires of the second experiment, *open circles* the five wires of the third experiment. The changes in activity which would be caused by a 10% change in the amount of epithermal neutron self-shielding are indicated by the *dashed lines*

10% change in the amount of epithermal neutron self-shielding (i.e. 10.0 or 12.2% epithermal self-shielding) were also calculated; they are indicated in the figure by the dashed lines. In channel 1, 49% of the ^{198}Au activity was

Table 1 Relative differences in measured activities of the horizontally (H) and vertically-positioned (V) wires irradiated in the central channel (CC) of the TRIGA reactor

Nuclide	E_γ (keV)	H/V−1 (%)	unc. (%)	Nuclide	E_γ (keV)	T/B−1 (%)	unc. (%)
Paper only in Al-container							
^{116m}In	416.9	−0.88	0.39	^{114m}In	190.3	0.70	0.64
	1097.3	−1.31	0.45		558.4	1.80	1.75
	1293.5	−1.21	0.33		725.2	3.90	2.02
Average[a]		−1.13	0.22			1.08	0.58
Corrected average[b]		−0.05	0.80				
Polyethylene in Al-container							
^{116m}In	416.9	5.38	4.04	^{114m}In	190.3	−1.60	0.56
	1097.3	2.04	4.35		558.4	−0.40	1.60
	1293.5	0.85	3.77		725.2	0.30	1.89
Average[a]		2.70	2.33			−1.34	0.51
Corrected average[b]		1.35	2.84				

The quoted uncertainties originate from the statistical measurement uncertainties

[a] Variance-weighted average of (H/V−1) over the three measurements

[b] Correction taking into account the axial flux gradient, measured from the activity of ^{114m}In

produced by epithermal neutrons and in the more thermalized channel 8, only 22%. Thus, in channel 8, a 10% change in the amount of epithermal self-shielding (from 11.1 to 12.2%) would cause only a 0.22% change in the total ^{198}Au activity. It is evident in Fig. 2 that there is no systematic difference in specific activity between vertical and horizontal wires and no significant variation with angle between the axis of the wire and the direction towards the reactor core.

Jožef Stefan Institute TRIGA

The results from the measurements at the Jožef Stefan Institute are given in Tables 1 and 2 for both cases: with and without polyethylene capsules. The axial flux gradient is proportional to the ratio of activities between the top and bottom halves (T/B) of the vertical wire, measured from the activity of ^{114m}In, which is the activation product of ^{113}In. Self-shielding for this nuclide is small compared to

Table 2 Relative differences in measured activities of the horizontally (H) and vertically-positioned (V) wires irradiated in the carousel facility at position (IC40) of the TRIGA reactor

Nuclide	E_γ (keV)	H/V−1 (%)	unc. (%)	Nuclide	E_γ (keV)	T/B−1 (%)	unc. (%)
Paper only in Al-container							
^{116m}In	416.9	0.40	0.44	^{114m}In	190.3	−0.20	0.92
	1097.3	0.45	0.39		558.4	4.10	2.84
	1293.5	−0.55	0.33		725.2	10.90	3.27
Average[a]		0.00	0.22			0.92	0.85
Corrected average[b]		0.92	1.06				
Polyethylene in Al-container							
^{116m}In	416.9	0.45	0.39	^{114m}In	190.3	1.20	0.85
	1097.3	0.98	0.41		558.4	−1.10	2.58
	1293.5	1.41	0.35		725.2	−4.70	2.84
Average[a]		0.98	0.22			0.55	0.78
Corrected average[b]		1.54	1.00				

The quoted uncertainties originate from the statistical measurement uncertainties

[a] Variance-weighted average of (H/V−1) over the three measurements

[b] Correction taking into account the axial flux gradient, measured from the activity of ^{114m}In

the self-shielding of ^{115}In, which produces ^{116m}In. The mean distance between the two wire-halves is the same as the distance between the whole horizontal and vertical wires respectively, so the gradient is representative for correcting the ratio of horizontal/vertical (H/V) wire activities. Positive gradient means the horizontal wire activates less, so the gradient must be added to compensate.

Since the horizontal and the vertical wire are the same size and measured at the same position on the detector, their measured specific activities (after correction for the flux gradient) should be the same, except for the differences in the level of self-shielding. In all cases only the ratios of activities are considered, measured with the same gamma-line; therefore the systematic errors cancel out (e.g. detector efficiency, measurement geometry, gamma-emission probability, sample purity, etc.). The uncertainties of the measured ratios of activities are of the order of 1%. According to the original Monte Carlo model with anisotropic neutron field [2], the effective self-shielding factor for ^{115}In in the central channel was 0.309 ± 0.001 for the vertical wire and 0.335 ± 0.001 for the horizontal wire; thus the expected difference in specific activity was about 9%. The experimental results indicate that the difference is much smaller (if present at all).

Why do these results indicate a nearly isotropic neutron field at the sample position when it was originally speculated that it would be anisotropic? It appears that streaming of neutrons down the irradiation channel is less important than the local neutron thermalization effects. A reduced effect of the streaming of neutrons could be explained by the fact that the active core height of the TRIGA reactor is only 38 cm. The dominant effect which gives a nearly isotropic neutron field at the sample position is the scattering of neutrons in the nearby moderator and in the sample container materials.

A refined Monte Carlo model of the TRIGA reactor irradiation facility was tested, in which the surface source was moved to half-way between the fuel and the irradiation channel, thus modelling explicitly the moderator around the irradiation channel. Also, reflecting materials near the sample position were included in the model. The difference in the activities of the two wires calculated with the refined model was less than 1%, which is consistent with the measurements.

Conclusions

It has been shown by experimental measurements that for four irradiation facilities in or near the cores of two small reactors the effects of neutron field anisotropy are practically negligible. These experimental results were confirmed by calculations with a refined computational model that includes some of the material surrounding the irradiation channel. Since the neutron fields in these facilities are thought to be as anisotropic as in any small reactor, it can be concluded that the neutron field anisotropy is not important to self-shielding in the irradiation facilities of all small reactors. However, in larger reactor systems with voided irradiation channels, the users should check for the possible impact of neutron field anisotropy on the level of self-shielding.

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