Journal of Radioanalytical and Nuclear Chemistry, Vol. 271, No.2 (2007) 283–287

Corrections for neutron activation analysis with non-1/v nuclides using reactor moderator temperature readings

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(Received April 13, 2006)

Neutron activation analysis using the single comparator method or the k_0 method may be less accurate in the cases of non-1/v nuclides if irradiations are done at varying reactor temperatures. We propose a method to correct this effect, using available reactor moderator temperature readings. It was verified by the irradiation of two non-1/v nuclides, ^{176}Lu and ^{151}Eu. Irradiations were carried out in two irradiation sites at two reactor powers and at estimated neutron temperatures varying from 18 to 46 °C. In all cases the measured activities showed a variation with temperature consistent with published tables of the variation of the Westcott g-factor. The method should be accurate to 1% for most small reactors and eliminates the need to irradiate temperature monitors.

Introduction

The classical relative method of neutron activation analysis requires the irradiation of standards of all elements to be determined with every sample to be analyzed. It is much more convenient to first complete the measurements of the standards and then to irradiate only one flux monitor or comparator with each sample. This single-comparator method assumes that the activation rates of all nuclides will change by the same amount as the neutron flux in the irradiation site changes. If the neutron temperature changes, this assumption will be valid for most nuclides because their thermal neutron activation cross sections vary with neutron energy as $E^{-1/2}$ or as the inverse of the neutron velocity. There are, however, a few non-1/v nuclides for which the assumption does not hold.

To accommodate these non-1/v nuclides, the WESTCOTT formalism[1] has been introduced to describe their activation rates as a function of neutron temperature. This is often used with the k_0 method[2,3] and lutetium is used as a neutron temperature monitor because the ^{176}Lu(n,γ)^{177}Lu reaction deviates the most from 1/v among all easily activated nuclides.[2–4] But it is still inconvenient to irradiate and count the Lu monitor each time there may be a temperature change at the irradiation site. For small research reactors, it is quite likely that the neutron temperature at the irradiation site may be accurately estimated from the temperature of the reactor moderator, which is normally measured continuously. In this work we use a Slowpoke reactor to determine the accuracy of neutron temperature monitoring with available moderator temperature readings.

Experimental

A Slowpoke reactor is shown schematically in Fig. 1. The moderator-cooling water flows through the core by convection and exits through a narrow gap between the annular reflector and the top reflector. It then flows to the water surface. Cooler water descends along the wall of the container and enters the core through the narrow gap between the annular reflector and the bottom reflector. All Slowpoke reactors have a thermocouple, which measures the temperature of the water as it exits the core. The reactor at Ecole Polytechnique also has a thermocouple at the inlet to the core. When the reactor operates at full power, 20 kW, the temperature rise, ΔT, from the core inlet to the core outlet is 24 °C.[5] At half power, 10 kW, the temperature rise is 16 °C. The reactor uses 20% enriched LEU fuel. Several other Slowpoke reactors use 93% enriched HEU fuel. The placement of the core and the irradiation sites is the same, but they run at slightly lower powers and the corresponding ΔT's at full power and half power are 22 °C and 14 °C, respectively.[5]

The neutron temperatures in the inner and outer irradiation sites may be estimated from the temperature of the surrounding moderator. Referring to Fig. 1, the neutron temperature of the outer irradiation site, T_0, should be close to the temperature of the water flowing down past it, which is the temperature of the water entering the core. Thus, relative to the reactor outlet water thermocouple reading, T_R, we have:

$$T_0 = T_R - \Delta T \qquad (1)$$

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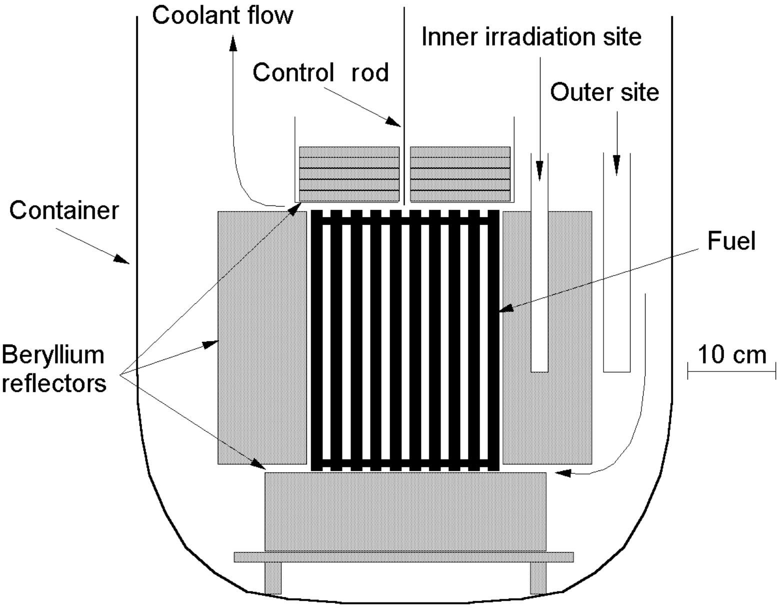

Fig. 1. The Slowpoke reactor core showing coolant water flow

The inner irradiation site is located at mid-height of the core, where the core water temperature is T_R–$\Delta T/2$, and at 40% of the distance between the core water and the water near the outer irradiation site, whose temperature is given by Eq. (1). Thus, the neutron temperature in the inner irradiation site, T_I, is estimated by linear interpolation as:

$$T_I = T_R - 0.7\Delta T \qquad (2)$$

To verify whether certain reaction rates vary as expected as a function of these estimated neutron temperatures, Cu, Lu and Eu monitors were irradiated in the inner and outer irradiation sites. The ^{63}Cu(n,γ)^{64}Cu reaction rate is known to vary with neutron temperature as 1/*v*. Copper wires, 10 mm long and of approximate mass 60 mg, were used as 1/*v* flux monitors. To measure some non-1/*v* reaction rates, monitors were prepared from 1000 ppm Specpure Plasma Standards of Lu and Eu, certified accurate to 0.3%, purchased from Alfa Aesar, USA. For both Lu and Eu, 100 μl aliquots were weighed and dried on strips of filter paper, which were rolled into cylinders and inserted in polyethylene vials. The ^{151}Eu(n,γ)^{152}Eu reaction is non-1/*v* but deviates from 1/*v* less than the ^{176}Lu(n,γ)^{177}Lu reaction.[3] These monitors were each irradiated for 10 minutes in the inner or outer irradiation site at reactor full power or half power. At full power the thermal neutron flux was approximately $1 \cdot 10^{12}$ n·cm^{-2}·s^{-1} in the inner site and $0.5 \cdot 10^{12}$ n·cm^{-2}·s^{-1} in the outer site. Between these irradiations the reactor temperature was made to vary as much as possible by running the pool water cooling system with cold water or by shutting it off completely and operating the reactor at full power for prolonged periods. During each irradiation the reactor outlet water thermocouple reading, T_R, was recorded.

The ^{64}Cu ($T_{1/2}$ = 12.7 h), ^{152m}Eu ($T_{1/2}$ = 9.29 h) and ^{177}Lu ($T_{1/2}$ = 164.4 h) activities were measured using their 511 keV, 842 keV and 208 keV gamma-rays, respectively. The monitors were counted 10 cm from a germanium detector. The activities were corrected for decay time and then divided by the mass of the element to give specific activities.

Results and discussion

The relative ^{64}Cu specific activities are shown in Fig. 2 as a function of the neutron temperature of the irradiation site, estimated by Eq. (1) or Eq. (2). In all four situations, two reactor powers and two irradiation sites, the behavior is well described by the lines drawn through the data, which represent a decrease in specific activity of 0.20%/°C. This is caused by a decrease in the neutron flux in the irradiation sites with temperature. This phenomenon is well known[6] and is due to the nature of the neutron detector used to control reactor power and to maintain a constant neutron flux. All Slowpoke reactors use a cadmium neutron detector based on the ^{113}Cd(n,γ)^{114}Cd reaction, which is non-1/*v*. The true neutron flux is that indicated by a 1/*v* nuclide, such as ^{64}Cu. Therefore, the measured ^{64}Cu specific activities were used to determine the effective neutron fluxes as a function of neutron temperature. It should be noted that the fluxes thus determined are relative effective fluxes, not thermal fluxes. This reactor is known to have a thermal to epithermal flux ratio, *f*, of 17.9 in the inner site and 51.1 in the outer site.[7] Considering the ratio of resonance integral to thermal neutron cross section for the ^{63}Cu(n,γ)^{64}Cu reaction ($Q_0 = 1.14$), 7.1% of the ^{64}Cu activity produced in the inner irradiation site is due to epithermal neutrons, as is 2.0% of the ^{64}Cu activity produced in the outer irradiation site.

The measured ^{177}Lu specific activities were divided by the neutron flux for the irradiation site, reactor power level and temperature, as determined by the ^{64}Cu flux monitors (the lines in Fig. 2). The results are shown in Fig. 3. Since activities are proportional to the product of neutron flux and effective cross-section, the data shown represent the variation in the ^{176}Lu(n,γ)^{177}Lu effective cross section with temperature. For thermal neutron irradiation, the effective cross-section for non-1/*v* reactions in a Maxwell-Boltzmann distribution of neutrons is the product of the cross section at a neutron energy of 0.0253 eV and the WESTCOTT *g*-factor,[1] which varies with neutron temperature. The WESTCOTT *g*-factor has been calculated for this reaction by HOLDEN,[4] and the relative values are shown by the lines in Fig. 3. At 30 °C the variation is +0.40%/°C. For each irradiation site, the line was adjusted to fit the measured data at the lowest temperatures and at reactor half power. As can be seen, the data measured at reactor full power tend to be about 1% lower than those measured at half power. Otherwise, the observed variation of the ^{176}Lu(n,γ)^{177}Lu effective cross section with neutron temperature follows very well that predicted by the calculated *g*-factors. This is confirmation that the relative neutron temperatures estimated by the method proposed here are equivalent to those that would be determined using Lu as a temperature monitor. It should be noted that, like ^{64}Cu, part of the ^{177}Lu activity is due to epithermal neutrons, approximately 9% in the inner irradiation site and 3% in the outer irradiation site. In spite of this epithermal neutron contribution to the effective cross section, the variation with temperature still agrees well with that predicted by the *g*-factors calculated for thermal neutron irradiation.

In the same manner as ^{177}Lu, the ^{152m}Eu specific activities were divided by the measured neutron fluxes, and the results are shown in Fig. 4. They represent the variation in the ^{151}Eu(n,γ)^{152m}Eu effective cross section with neutron temperature. The lines shown are the WESTCOTT *g*-factors calculated by HOLDEN,[4] they vary by –0.11%/°C at 30 °C. For each irradiation site, they were adjusted to fit the measured data at the lowest temperatures at reactor half power. As can be seen, for both irradiation sites, the observed variation of the ^{151}Eu(n,γ)^{152m}Eu effective cross section is similar to the calculated variation of the WESTCOTT *g*-factor.

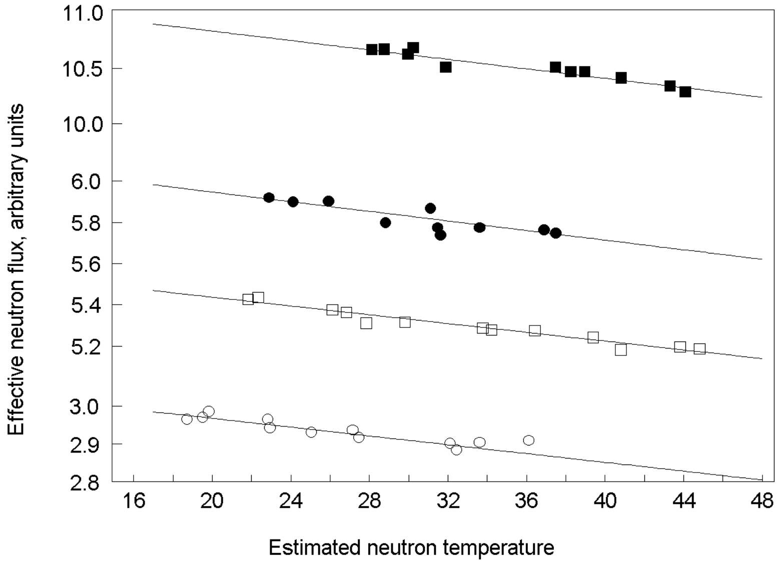

Fig. 2. Effective neutron fluxes as determined from the relative ^{64}Cu specific activities as a function of estimated neutron temperature: ■ inner irradiation site at full reactor power, □ inner irradiation site at half power, ● outer irradiation site at full power, ○ outer irradiation site at half power. The lines represent a variation of -0.20%/°C

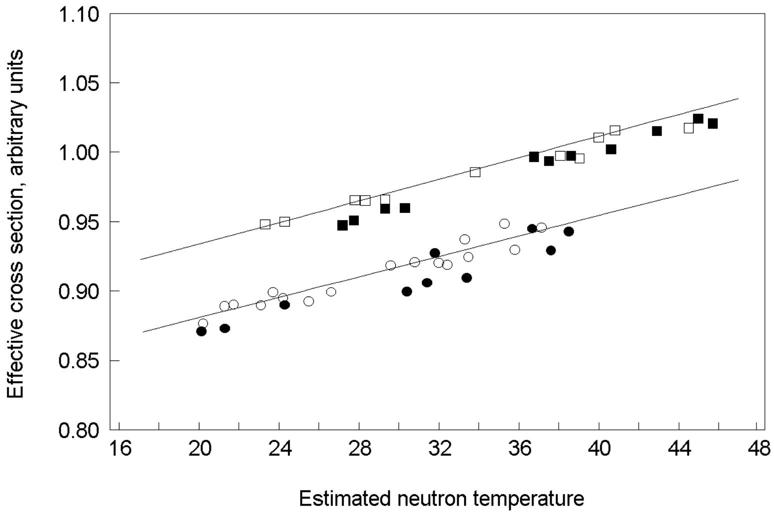

Fig. 3. The measured relative ^{176}Lu(n,γ)^{177}Lu effective cross sections as a function of estimated neutron temperature: ■ inner irradiation site at full reactor power, □ inner irradiation site at half power, ● outer irradiation site at full power, ○ outer irradiation site at half power. The lines are the calculated WESTCOTT *g*-factors

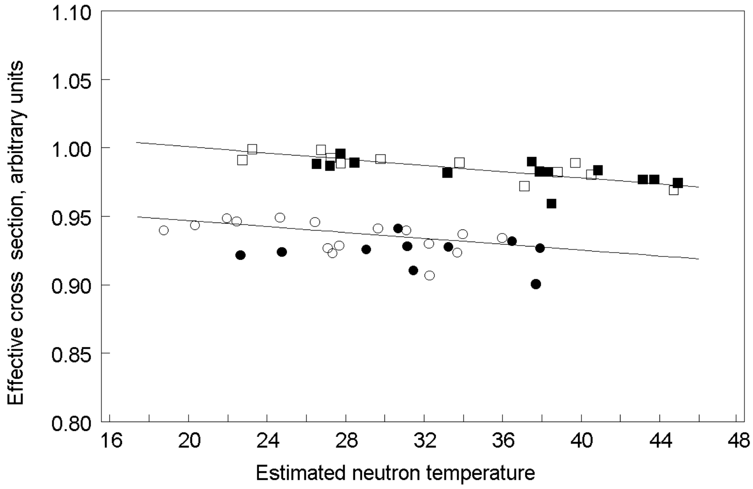

Fig. 4. The measured relative ^{151}Eu(n,γ)^{152m}Eu effective cross sections as a function of estimated neutron temperature: ■ inner irradiation site at full reactor power, □ inner irradiation site at half power, ● outer irradiation site at full power, ○ outer irradiation site at half power. The lines are the calculated WESTCOTT *g*-factors

The above data show that the proposed method for estimating neutron temperature can be used to predict the relative activities of ^{177}Lu and ^{152}Eu, with an accuracy of 2% or better, over a temperature range of more than 20 °C. The proposed method for improving the accuracy of single-comparator NAA for non-1/*v* nuclides is the following: A standard of each element is activated along with a 1/*v* flux monitor, such as Cu, at any neutron temperature, and subsequent irradiations at different temperatures are compared using the same flux monitor for all elements. Temperature corrections for the non-1/*v* cases are done using the calculated variation of *g*-factor with neutron temperature, and the neutron temperature estimated from the reactor thermocouple reading as described above. All other reactions commonly used in neutron activation analysis deviate from 1/*v* less than ^{176}Lu(n,γ)^{177}Lu and ^{151}Eu(n,γ)^{152}Eu. The corrections for them are even smaller and they can be done in the same manner.

With the k_0 method, no element standards are needed, k_0 values are used instead. For 1/*v* nuclides, the k_0 values are proportional to the thermal neutron activation cross section. For the non-1/*v* nuclides, variations with temperature have been accurately accounted for[3] by using a modified WESTCOTT formalism and irradiating a Lu temperature monitor with each batch of samples. With the present method, the temperature monitor needs to be irradiated only once and relative neutron temperatures are deduced for each subsequent irradiation from reactor thermocouple readings.

The proposed method should be reactor-independent. Figures 3 and 4 show that the variation of the ^{177}Lu and ^{152m}Eu activities with reactor power and temperature were predicted with an accuracy of 1% in the inner and outer irradiation sites, even though they have quite different neutron spectra, $f = 17.9$ and $f = 51.1$, respectively, and the relation between neutron temperature and moderator temperature may be different in the two sites.

Conclusions

A simple method has been demonstrated for the correction of NAA data with non-1/*v* nuclides, using available temperature readings rather than the continual irradiation of temperature monitors. For the Slowpoke reactor, where the neutron temperature in the irradiation sites varies over a range of about 20 °C in the extreme case, and only about 10 °C during routine operation, the only effective cross section that varies significantly with temperature is ^{176}Lu(n,γ)^{177}Lu. The ^{151}Eu(n,γ) ^{152}Eu effective cross section was found to vary by only 2% over the extreme temperature range, and the other non-1/*v* reactions vary by even less than that. Thus, for most NAA work, only measurements using ^{177}Lu need to be corrected for temperature changes.

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