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Effects of Reactor Temperature and Sample Mass on the Activation of Biological and Geological Materials with a SLOWPOKE Reactor

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ABSTRACT

Neutron activation analysis with a SLOWPOKE reactor relies on the stability of the neutron flux in the irradiation sites. Flux monitors were irradiated to measure the flux variation with the reactor temperature and with the amount of moderator in the irradiation vial. The thermal flux decreased by 2.7% for a 10°C increase in reactor temperature. The thermal flux increased by up to 8% and the fast flux decreased by up to 13% depending on sample size and hydrogen content.

Index Entries: Neutron activation analysis; nuclear reactor; neutron flux; neutron moderation.

INTRODUCTION

Small research reactors such as SLOWPOKE and MNSR, designed for neutron activation analysis (NAA), will operate for 20 yr or more with the same fuel. Flux monitors are not considered necessary for NAA with these reactors because it has been shown *(1, 2)* that, under favorable conditions, the neutron flux in a given irradiation site is reproducible within about 1%. Sensitivity constants are determined once for each element using standards (or the k_0 method) and used for subsequent analyses over long periods of time *(3)*, assuming a constant neutron flux. To verify the limits of validity of this assumption for a SLOWPOKE reactor, the present study was undertaken to measure the variation of neutron flux with the reactor temperature and with the amount of moderator in the irradiation vial.

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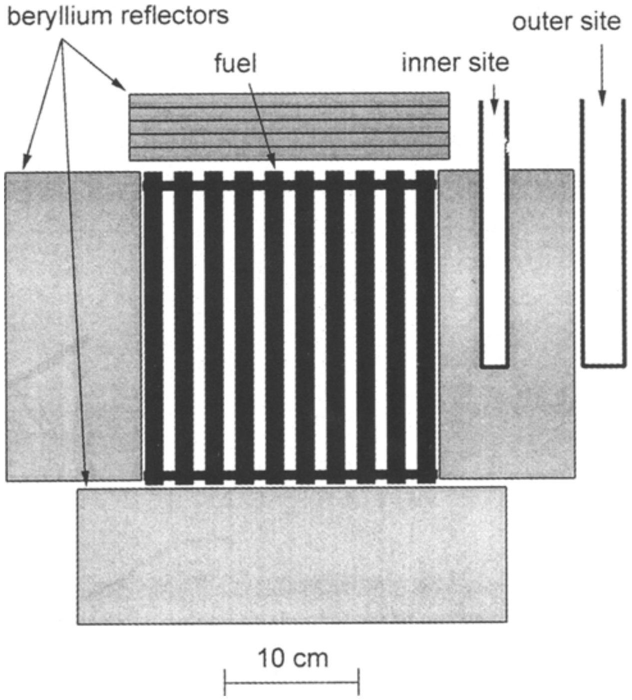

Fig. 1. The SLOWPOKE reactor core with irradiation sites.

MATERIALS AND METHODS

The core of the SLOWPOKE reactor is shown in Fig. 1. There are five inner irradiation sites in the annular beryllium reflector that surrounds the fuel and up to five outer irradiation sites in the surrounding water. The inner sites are at a radius of 15 cm from the axis of the reactor and the outer sites are at a radius of 23 cm. Access to these sites is by pneumatic rabbit systems. In the inner sites, all irradiations are performed in 7-mL polyethylene vials (14 mm diameter × 48 mm), and in the outer sites, 24-mL vials (26 mm diameter × 46 mm).

Variation of Neutron Flux with Reactor Temperature

Irradiations were carried out over a period of 6 mo in an inner irradiation site of the Ecole Polytechnique SLOWPOKE reactor at a control console flux setting of $5 \times 10^{11}/\text{cm}^2/\text{s}$, which corresponds to a reactor power of 10 kW. Minimum and maximum temperatures were obtained by operating the reactor in winter with the pool cooling system turned on (domestic cold water circulating through a coil) and in summer with the cooling system turned off. Thermocouples measured the temperature of the water entering and leaving the core. The average of the two values was taken to be the average moderator temperature. Thermal and epithermal neutron fluxes were measured by the bare monitor *(4)* method commonly used in k_0 standardization: Zr foil and Al–Au wire were irradiated and the activities of ^{95}Zr, ^{97}Zr, and ^{198}Au measured with a germanium detector. Three equations involving the three measured activities were solved to obtain ϕ_{th} (the thermal flux), ϕ_e (the epithermal flux), and α (the parameter describing the deviation of the epithermal neutron spectrum from the ideal $1/E$ distribution). The fast neutron flux was measured by the irradiation of Fe wires to produce ^{54}Mn by the ^{54}Fe(n, p) reaction.

During the course of this study, the reactor fuel, enriched to 93% in ^{235}U, which was exhausted after 21 yr of operation, was removed and replaced with 20% enriched fuel. The measurements of neutron flux as a function of reactor temperature were repeated with the newly fueled reactor.

Variation of Neutron Flux with Sample Size

A sample as large as possible is often desired in the NAA of geological and biological material because it is more representative of the material being studied. Also, with this low-flux reactor, a large sample is sometimes needed to improve sensitivity. Flux monitors were irradiated in simulated samples of various sizes in the inner and outer sites to determine the effects of neutron moderation and absorption on the average neutron flux in the sample. The irradiation vials were filled to various levels with water and polyethylene, which are good moderating materials containing hydrogen, with carbon, and with a poor moderator, SiO_2. None of the materials used contained elements with a high-neutron-absorption cross-section. For thermal and fast neutrons, the monitors were 0.6-mm-diameter wires composed of 10% chromium and 90% nickel. For epithermal neutrons, the monitors were 0.5-mm-thick by 2-mm-wide strips of zirconium. In the inner site, the ^{50}Cr(n, γ)^{51}Cr reaction (which has a resonance integral–to–thermal neutron cross-section ratio of only 0.53) is induced 96% by thermal neutrons, whereas the ^{94}Zr(n, γ)^{95}Zr reaction (which has a resonance integral–to–thermal neutron cross-section ratio of 248) is induced 94% by epithermal neutrons. Fast neutrons were measured by the ^{58}Ni(n, p)^{58}Co reaction.

To measure the effect on the neutron flux caused by the moderator, the activity of each monitor was compared to that of a similar monitor irradiated in air. The samples were irradiated sequentially for 30 min each. It is known that the flux in samples without a moderator varies by less than 1% for sequential irradiations at constant temperature. In order to measure the average neutron flux over the volume of the sample, the wires or strips were placed diagonally across the sample. When the flux gradient across the sample is constant, a diagonal wire measures the average flux. When the flux shows a maximum at the center of the sample,

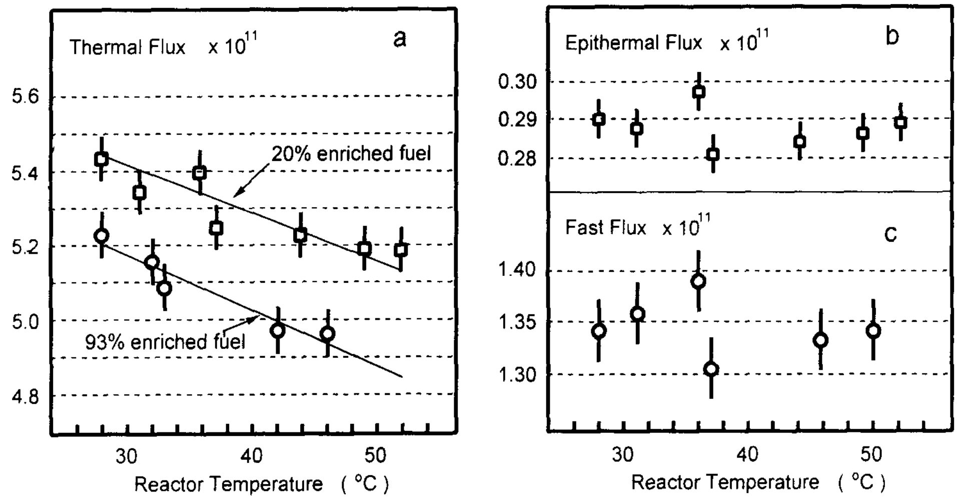

Fig. 2. Variation of neutron flux with reactor temperature.

such as the case where the thermal flux increases inside the sample due to moderation, a diagonal wire will overestimate the average flux. Measurements were performed by irradiating several vertical wires in a sample of water to determine the accuracy of the average flux measured by a diagonal wire; it was found that the diagonal wire overestimated the average flux by only 0.9%.

RESULTS AND DISCUSSION

Figure 2a shows the measured thermal neutron flux as a function of reactor temperature for the two types of reactor fuel. In both cases, the flux decreases by about 0.27% per degree Celsius. Figure 2b,c shows the measured epithermal and fast neutron fluxes as a function of reactor temperature for the reactor with 20% enriched fuel. No significant variation was observed for the epithermal and fast fluxes.

The variation in thermal flux with temperature is explained by changes in the properties of the moderator and by the nature of the control system. The neutron flux of a SLOWPOKE reactor is controlled by a "self-powered cadmium flux detector." The current from this detector is amplified and the value of the current is maintained constant within 1% by the reactor control circuitry. Neutrons striking the detector produce gamma-rays by the ^{113}Cd(n, γ) reaction. The gamma-rays eject electrons from the detector element producing an electric current. Thus, the control system actually maintains the ^{113}Cd(n, γ) reaction rate constant. In neutron activation analysis, when we state that a reactor has a constant neutron flux we actually mean that the reaction rate for the activation of the elements of interest is constant. For most elements activated by

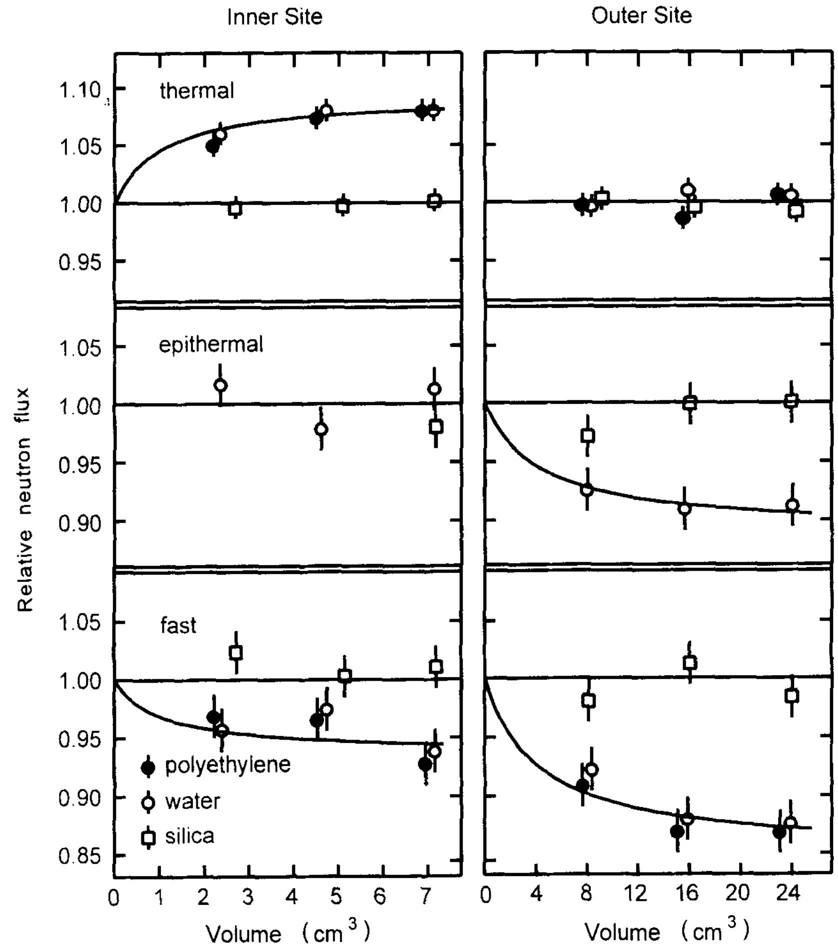

Fig. 3. Variation of thermal, epithermal, and fast neutron fluxes with sample size.

thermal neutrons, the activation cross-section varies as $1/v$, where v is the neutron velocity. However, the cross-section for the $^{113>}$Cd(n, γ) reaction does not vary as $1/v$; it is fairly independent of neutron velocity up to 0.5 eV neutron energy. Thus, as the neutron temperature (and average neutron velocity) increases, for a constant ^{113}Cd(n, γ) reaction rate, the reaction rate for elements activated by thermal neutrons decreases. It is interesting to note that Diaz Rizo et al. *(5)* observed a substantial increase in the thermal-to-epithermal ratio with increasing reactor temperature for a Triga reactor, the opposite of what was found in this work.

Figure 3 shows the measured variation of neutron flux with sample size for irradiation vials in the inner and outer sites filled to various heights. The water and the silica had a density of 1 g/cm^3 and the polyethylene 0.9 g/cm^3. It can be seen that silica had a negligible effect in all six cases. The samples of carbon (not shown) also had a negligible effect.

The results for polyethylene and water were similar in all cases. The polyethylene had a slightly lower density, which was offset by its slightly higher hydrogen content. The curves drawn through the data illustrate that the effect is zero for zero sample volume and tend toward a maximum effect as the sample increases in length.

To explain the effects caused by polyethylene and water, it should be understood that the flux in the inner sites of a SLOWPOKE reactor is very poorly thermalized: In an inner site with no moderator in the sample vial, the thermal-to-epithermal ratio is 18.4 and the thermal-to-fast neutron flux ratio is 4.0. In the outer sites, the thermal-to-epithermal ratio is 46.5 and the thermal-to-fast ratio is 15.8. For fast neutrons, the hydrogen in the sample reduces the average flux; the effect is greater in the outer site because the sample has a radial thickness about twice that in the inner site. The amount of moderator has no effect on the epithermal neutron flux in the inner site because it produces as many epithermal neutrons as it removes. In the outer site, however, there are relatively fewer fast neutrons to moderate; thus, the removal of epithermal neutrons dominates. For thermal neutrons in the inner site, there are few thermal neutrons for the hydrogen to absorb; thus, the thermalization of epithermal neutrons dominates and the thermal flux increases with sample size. Thompson et al. *(6)* observed a similar effect: The thermal flux increased by 10% at mid-height of a 40-mm-high, 20-mm-diameter sandwich of polyethylene disks. For thermal neutrons in the outer site, there is no net effect because absorption by the hydrogen in the sample equals production by thermalization. Alamin and Spyrou *(7)* observed similar effects in kilogram-sized samples in a poorly thermalized flux; as water was added to the sample, the average thermal flux increased but reached a maximum as absorption began to dominate. For sample size and neutron spectrum similar to those of this work, Matsushita et al. *(8)* found that polyethylene increased the thermal, epithermal, and fast flux gradients across the sample relative to silica, but they did not evaluate the average fluxes in the sample.

CONCLUSIONS

The thermal neutron flux of a SLOWPOKE reactor, which had previously been thought to be reproducible to 1%, was observed to decrease by up to 5% with increasing reactor temperature. However, in normal operation, the temperature varies over a range of only about 10°C, causing less than a 3% variation in thermal flux, which can be ignored in all but the most accurate NAA work.

The measurements of average neutron flux in the sample as a function of sample size suggest that variations will be negligible for geological material such as silicate rocks, unless they contain high concentrations of highly absorbing elements like the rare earths. For biological materials, the thermal flux may be enhanced by as much as 8% and the fast flux reduced by as much as 13%, depending on irradiation site, sample size, and hydrogen content. The curves relating neutron flux to sample volume may be used to correct these effects; the correction should be reduced by the estimated hydrogen concentration relative to that of water. Typical dry biological tissues contain about 7% hydrogen, whereas wet tissues may approach the 11% hydrogen content of water.

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