A Non-Intrusive Neutron Method for Poison Concentration Monitoring in CANDU Reactors

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Abstract

A neutron reflection method is developed for measuring the concentration of the neutron absorbing gadolinium in the tanks of the liquid poison injection shutdown system of a CANDU reactor. The feasibility of the method is demonstrated experimentally, and Monte Carlo simulations are utilized to design and model the performance of a proposed device.

Résumé

On présente une méthode basée sur la réflection des neutrons pour mesurer la concentration du gadolinium, absorbeur de neutrons, dans les réservoirs du système d'arrêt à injection de poison liquide du réacteur CANDU. On démontre expérimentalement que la méthode est pratique et on utilise des simulations Monté Carlo afin de concevoir un dispositif et d'en modéliser la performance.

Introduction

The liquid poison injection shutdown system, the secondary shutdown system (SDS2), of a CANDU reactor utilizes a gadolinium nitrate, $Gd(No_3)_3 \cdot 6H_2O$, solution to neutron-poison the reactor following a trip. The solution is contained in several (six to eight) tanks that are connected via an open-line to the reactor calandria. The solution is injected, when needed, under pressure into the moderator system. In order for this shutdown system to be effective, the concentration of natural gadolinium in the solution is required to be no less than 4000 mg Gd/Kg solution.

The effectiveness of the SDS2 is presently insured by periodic measurement of the gadolinium concentration. Typically, a sample is taken weekly from one of the tanks and analyzed in the laboratory using atomic absorption. This is supplemented by mass spectroscopy analysis, usually twice a year, in order to ensure the adequacy of the isotopic content of the material used. Natural gadolinium, as shown in Table 1, contains about 30 per cent of the neutron absorbing isotopes, Gd-155 and Gd-157. To avoid the remote possibility of using gadolinium burned-out by a prior reactor irradiation, isotopic analysis is required.

Table 1: The Isotopes and Neutron Cross-Sections of Natural Gadolinium (a)

Isotope	*Abundance (per cent)*	*Thermal Absorption Cross-Section (barns)*
152	0.2	1.1×10^3
154	2.1	90
155	14.8	6.1×10^4
156	20.6	2
157	15.7	2.55×10^5
158	24.8	2.4
160	21.8	0.8
	100.0	4.9×10^4

(a) Lederer & Shirley 1978 [1].

The periodic analysis procedure, in addition to being manual and subject to administrative error, does not provide a continual indication of the effectiveness of the system. It is possible that one or more tanks can be downgraded in gadolinium solution and remain undetected until a sample is withdrawn from the tank a few weeks later. It is desirable, therefore, to have an on-line monitoring device that can continually provide an estimate of the poison concentration in each tank. Such a system would reduce the manual effort required for testing and would improve control room information.

In this paper, we present a new non-intrusive method for on-line monitoring of the neutron-poison solution in each of the tanks of the SDS2. The method is based on neutron reflection and utilizes external neutron sources and detectors. The details of this technique are given later. We begin, however, by

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reviewing some of the methods that can be utilized for gadolinium concentration monitoring.

Measurement Methods

Several methods can be considered for on-line monitoring of the poison concentration in SDS2 tanks. However, in order to be able to choose a suitable method for application, one must define the characteristics of the required device. An on-line monitoring system is desired to be:

1. capable of yielding continual measurement of the solution's neutron absorption ability in each of the SDS2 tanks;
2. non-intrusive to the existing piping system;
3. maintenance free;
4. inexpensive.

An on-line monitoring system is required to provide continual information that can be accessed from the control room. It is also desired that the system provide directly a measure of the amount of neutron-absorbing material available in each tank. This enables the detection of any changes in the isotopic content of the gadolinium. Since the piping network of the SDS2 is designed as a Nuclear Class I system, the monitoring device is preferred to be non-intrusive, to avoid the costly process of modifying the system to accommodate the measuring equipment. The monitoring system also needs to be easy to install, reliable, reasonably maintenance-free, and inexpensive. Some of the techniques that have been previously proposed for poison monitoring are discussed below and assessed against the above characteristics.

Mass Spectroscopy

Mass spectroscopy provides an accurate means of measuring the isotopic content of an ionized sample. The method is based on the deflection of ions in an electromagnetic field. This provides a direct estimate of the type and amount of isotopes in the sample. The device is delicate, expensive, and requires samples to be withdrawn from each tank.

Atomic Absorption

In this method, a light source of a frequency corresponding to the energy required to excite a gadolinium atom is applied to an atom vapour sample of the solution. The gadolinium present absorbs the incident radiation to a degree proportional to the concentration of gadolinium in the sample. The apparatus used in this method is a delicate one and is difficult to apply on-line. Moreover, the method is intrusive and does not directly measure the neutron absorption ability of the solution.

Electrical Conductivity

The conductivity of a gadolinium nitrate solution varies with its concentration. A conductivity meter can, therefore, be used for gadolinium concentration monitoring. This requires the introduction of a probe into the tank or the associated pipes. However, the conductivity measurement does not provide a direct indication of the neutron absorption ability of the solution. It requires frequent calibration and is easily affected by impurities and the degree of acidity of the solution.

Thermal Neutron Attenuation

This is a direct method for measuring the absorption ability, and consequently the concentration of the neutron-poisoning isotopes. A very strong neutron source would, however, be needed, if the method were to be applied directly to the solution contained in an SDS2 tank. This is due to the large distance neutrons have to travel in a highly absorbing medium. The method can, however, be applied to a small circulating volume of solution drawn from the tank. Since available isotopic neutron sources emit fast neutrons, a thermalization assembly around the source is required. The thermalized neutrons can be utilized to measure the concentration of more than one solution sample located around the neutron source. This method is simple, capable of providing continual measurements, and relatively maintenance free. However, an intrusive sampling system is needed.

Neutron Flux Depression

One may also consider using the flux depression caused by inserting a sample containing gadolinium solution into a neutron medium. The flux depression can be measured directly or by using neutron activation analysis of an internal flux monitor. The magnitude of the flux depression provides a direct measure of the solution neutron absorption ability. However, this is an intrusive method that requires the withdrawal of a solution sample from the tank or the use of a circulating sampling system.

Neutron Activation

Gadolinium produces gamma-active radionuclides under neutron irradiation. The activity of these nuclides can be used, at least in principle, to measure the gadolinium concentration. Activation analysis, however, requires a large neutron flux that can be only obtained inside a nuclear reactor, and therefore cannot be applied on-line.

All the systems discussed above are intrusive in nature, and only the conductivity and neutron attenuation or flux depression methods can be easily applied on-line. The conductivity method does not provide a measure of the neutron absorption ability of the solution, while the neutron attenuation and flux depression methods require a continuous circulating system to be installed on each tank. An alternative method that can overcome the above difficulties is, therefore, needed. A new measurement technique is introduced below.

Neutron Reflection Method

The neutron appears to be the most appropriate probe for monitoring the concentration of poison solutions, since it measures directly the neutron absorption ability of the solution. However, the neutron attenuation, flux-depression or activation techniques, discussed above, require intrusion into the existing piping system. To avoid this problem we propose to utilize neutron scattering for measuring the poison concentration. If an epithermal neutron beam is directed towards a tank, the neutrons will be scattered and slowed-down, some to the thermal energy, by the heavy water contained within the tank. The slowed-down neutrons will then attempt to escape from the tank and can be recorded by a neutron detector located outside the tank. The scattered thermal neutrons will, however, be exposed to absorption by the gadolinium contained in the solution. The neutrons will be also subject to attenuation by the relatively thick (typically 9 mm) stainless steel walls of the tank. This, however, introduces a constant attenuation factor that is independent of the poison concentration. The degree to which thermal neutrons can escape from the tank is, therefore, inversely proportional to the concentration of the gadolinium-absorbing isotopes in the tank. As will be shown later, the maximum neutron scattered flux is obtained at 180 degrees with the incident beam. Neutron reflection is, therefore, proposed for gadolinium concentration monitoring. Figure 1 shows a schematic diagram of the proposed technique. The monitoring system consists mainly of a neutron source and a detector positioned on the side of each tank. The detector is surrounded with a cadmium sleeve, so that only thermal neutrons reflected from the tank are seen by the detector. Radiation shielding surrounds the source and the detector. No shielding is required on the opposite side of the tank, since the reactor wall can be used for this purpose.

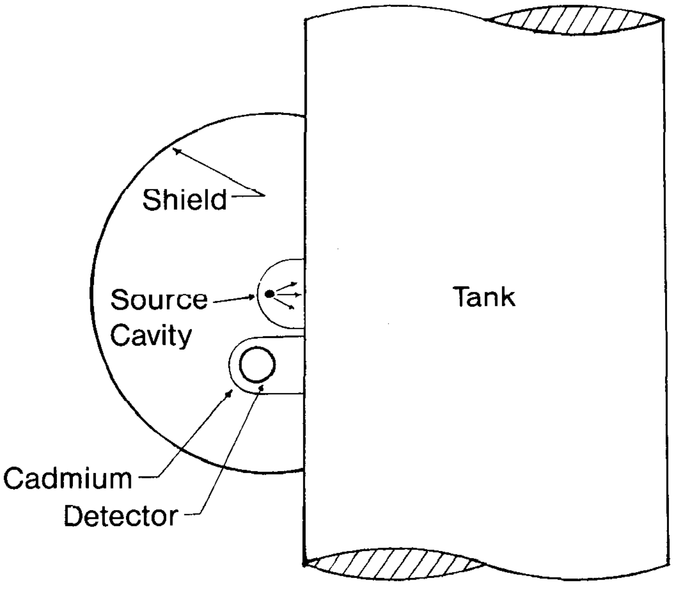

Figure 1 A neutron reflection gadolinium reflector.

The neutron reflection technique, as shown above, is non-intrusive and capable of providing continual measurement of the concentration of the neutron-absorbing isotopes in the solution. The simplicity of the technique makes it a relatively maintenance-free and inexpensive method for poison monitoring. The technique, therefore, meets all the desired characteristics defined earlier. In the following sections, the feasibility of the technique is demonstrated and a conceptual design is presented.

Experiments

In order to prove the feasibility of the neutron reflection technique, a set of experiments was carried out. A sheet-metal cylinder of a diameter equivalent to that of a typical SDS2 tank (236 mm) was used in the experiments. A neutron beam extracted from an americium-beryllium source (average energy of around 5 MeV) was employed. A BF_3 thermal neutron detector and standard counting electronics were utilized. The detector was surrounded with a cadmium sheet so that only neutrons scattered from the tank were detected. Virgin heavy water and gadolinium nitrate salt were supplied by the Point Lepreau Nuclear Generating Station (PLGS) for use in these experiments. Solutions of different concentrations were prepared and their concentrations were verified by atomic absorption analysis performed at PLGS. The gadolinium used was obtained from a shipment whose isotopic content had been previously confirmed by mass spectroscopy.

Results

As a first step, a set of experiments was carried out using light water, which has a slowing-down power much greater than that of heavy water. More thermal neutrons would, therefore, be produced inside a light water tank. Consequently, if one could not measure the gadolinium concentration in light water using neutron reflection, it would not be possible to measure it in heavy water. The results of the light water experiments are shown in Figure 2, on a semilog graph. The figure indicates that the detector response is approximately an exponential function of the gadolinium concentration.

The light water experiments were also used to obtain the optimum detector location. This location is defined as the position at which maximum foreground-to-background count ratio is obtained. The experiments showed that the optimum detector position is located a few centimeters directly beneath the neutron source. The same conclusion was obtained for heavy water.

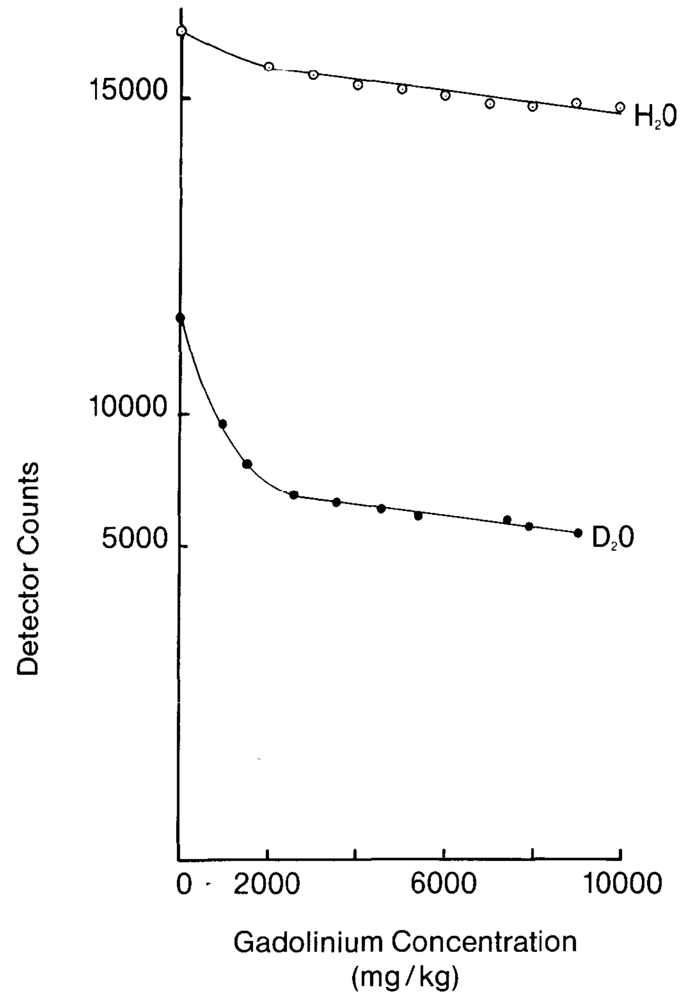

Figure 2 Experimental results.

The experimental results obtained using heavy water solutions are also shown in Figure 2. A nearly exponential relationship exists between the detector count rate and the gadolinium concentration, except at low concentrations. The behaviour of this detector response is explained later, when the physical theory behind the technique is presented.

The experiments presented above demonstrate the feasibility of using neutron reflection for gadolinium concentration monitoring. Although the detector response tends to saturate at high concentration, the logarithm of the signal provides a near-linear relationship that can be used to decide whether the gadolinium concentration has declined. It can be also noted that the reflected signal count rate is always larger than the background signal. This indicates that a useful signal can always be obtained even when the detector count rate is saturated. Given the success of the experiments, it was decided to perform some computer simulations in order to understand further the physics of the problem and to optimize further the performance of the techniques.

Monte Carlo Simulations

The Monte Carlo method is the most suitable approach to simulating the neutron reflection technique, because of the irregularity of the boundary conditions involved. A modified version of the COM [2] program was utilized for this problem. COM is a Center-of-Mass Monte Carlo program that was originally developed to simulate neutron scattering of a two-phase flow in a pipe. The program calculates the response of a thermal point detector located outside the pipe and provides a plot of the distribution of thermal neutrons generated inside the pipe.

The COM program was used to study the effect of the neutron source energy on the detector response. A set of hypothetical mono-energetic sources, and that of a californium-252 neutron source were considered. As it can be seen from Figure 3, the detector response is generally an exponential function of the gadolinium

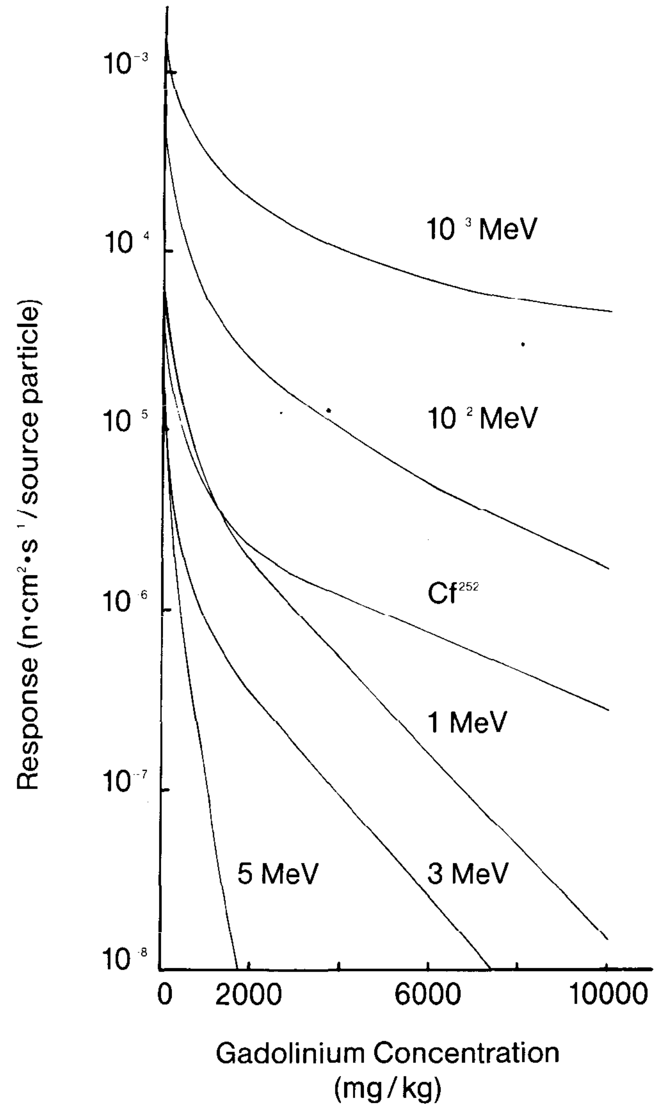

Figure 3 Monte Carlo results.

concentration, with some deviation at low concentrations. This deviation becomes more pronounced at lower energies. A theoretical explanation of this behaviour is given in the next section.

The measurement resolution, as indicated by the slope of the logarithm of the detector responses, increases with the source energy, as Figure 3 shows. The detector response, however, decreases in value as the energy increases. One must therefore choose an appropriate energy at which reasonable resolution and count rates can be obtained.

Mono-energetic isotopic sources are not readily available; therefore, polyenergetic sources must be used. The americium-beryllium source used in the experiments reported above emits neutrons of an average energy of about 5 MeV. This results, as Figure 3 indicates, in a low detector count rate per source neutron. A californium-252 neutron source was, therefore, considered. This is a spontaneous-fission source that emits neutrons of an average energy of about 2 MeV. The californium-252 source appears to provide a good compromise between resolution and count rate.

Empirical Model

For calibration purposes, and to understand better the system's behaviour, a simple empirical model is desired. We present here a model based on the observations obtained from the experimental and Monte Carlo results presented above. The model assumes that the distribution of the thermal neutron cloud generated inside the tank is independent of the gadolinium concentration. This is a reasonable assumption, since gadolinium is not a very effective neutron thermalizer. Gadolinium will, however, absorb the generated thermal neutrons as they diffuse throughout the tank. The amount of neutron absorption is assumed to be governed by the exponential relationship:

$$M_p = I_o \exp[-Kp] + V(p) \qquad [1]$$

where M_p is the detector response for a poison concentration p, I_o is a calibration constant, K is an attenuation parameter that incorporates the microscopic absorption cross section of gadolinium and the geometry of the thermal neutron cloud, and V(p) is a term that accounts for deviation from the exponential function. The exponential nature of the detector response is depicted in the semilog graphs of Figures 2 and 3, obtained from experiments and Monte Carlo simulations, respectively.

The deviation from the exponential function observed at low concentrations can be attributed to the fact that the effective area of the thermal neutron cloud contributing to the detector increases as the gadolinium concentration decreases. That is, for high gadolinium concentration, only those neutrons closest to the tank boundary near the detector contribute significantly to the detector counts, while neutrons near the centre of the tank have a very low probability of reaching the detector. On the other hand, at low gadolinium concentration, thermal neutrons generated near the centre of the tank have a higher chance of reaching the detector. In other words, the thermal cloud appears to the detector as more visible at low gadolinium concentrations. This effect is taken into account in equation (1) by introducing the visibility term V. This term is not only a function of the gadolinium concentration, but it is also a function of the incident neutron energy. This is due to the fact that the concentration of the thermal neutron cloud, and consequently its visibility, change with the incident neutron energy, as Figures 4 and 5 demonstrate.

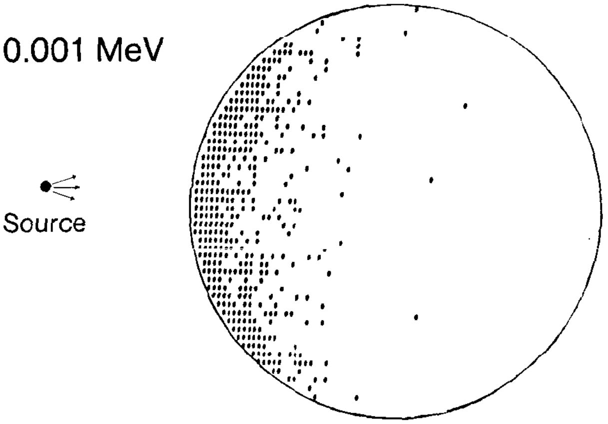

Figure 4 Neutron thermal cloud for a 0.001 Mev source.

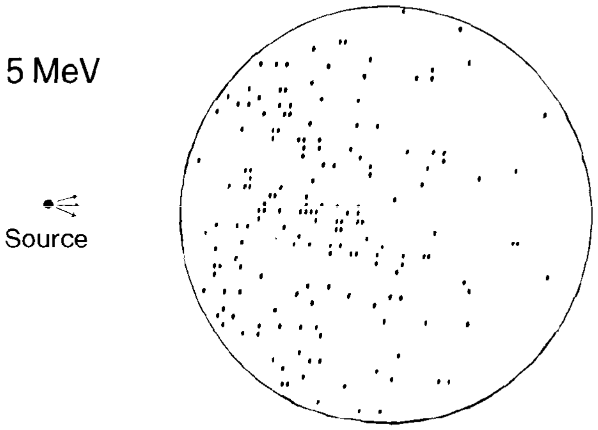

Figure 5 Neutron thermal cloud for a 5 Mev source.

The experimental and Monte Carlo results indicate that the visibility term can be represented by the equation:

$$V(p) = (M_o - I_o) \exp[-\beta p] \qquad [2]$$

where M_o is the detector response at zero poison

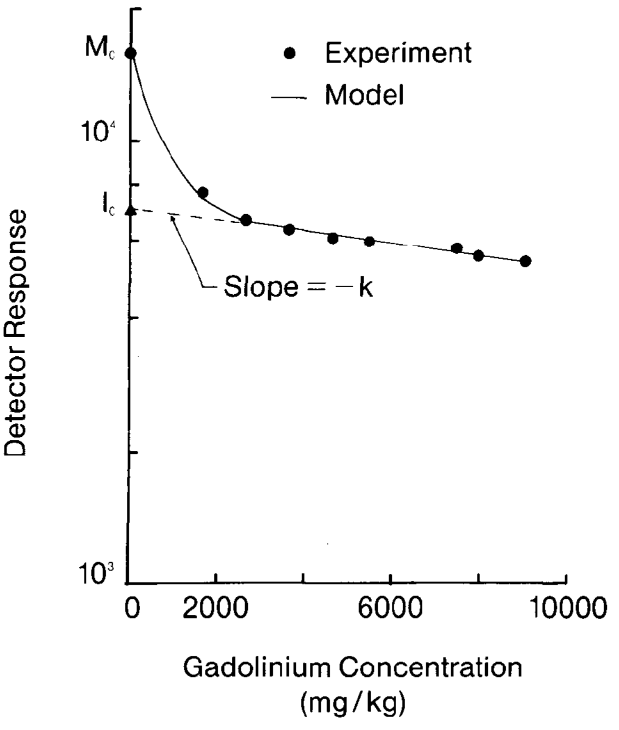

Figure 6 Theoretical model versus americium-beryllium heavy water experiments.

concentration, and β is the visibility coefficient at concentration p.

In order to demonstrate the validity of the above model, let us consider the americium-beryllium heavy water experimental measurements, shown in Figure 2. The slope of the line of the logarithm of the dominant exponential function at high concentration, shown in Figure 6, determines the value of the attenuation parameter K. The intersection of this line with the zero-concentration axis determines the constant I_0 of equation (1). The visibility coefficient β is determined by the slope of the line of the logarithm of equation (2). Figure 6 shows a good agreement between the model and the experimental measurements. Similar results were obtained for the californium-252 Monte Carlo results. This indicates that equation (1) is a valid representation of the detector response. The empirical model developed above can, therefore, be used to produce calibration curves for field-monitoring devices.

Preliminary Design

Based on the above experimental and theoretical evidence, one can now propose a preliminary design of a monitoring system. The device consists of a 2 μg californium-252 neutron source and a BF_3 detector enclosed inside a 200 mm-thick polyethylene shield. The basic configuration of the apparatus remains as shown in Figure 1.

The device should be located as close as physically possible to the tank and positioned so that use can be made of the reactor wall as a radiation shield. Vertical motion of the device can also permit the detection of gadolinium precipitation.

Californium-252 is chosen as the neutron source because, as indicated earlier, it provides a good compromise between resolution and detector count rate. Additional advantages of this source are its low heat generation, high specific activity, and low gamma radiation output. Therefore, no cooling of the device is required, and a minimum gamma ray shielding is needed. A 5 mm-radius sphere of lead surrounding the source is considered to provide sufficient gamma shielding for the proposed source strength.

The main disadvantage of a californium-252 source is its relatively short half-life (2.6 years). In order, however, to obtain the neutron yield of 4.6 million n/s of a 2 microgram (1.07 mCi) californium-252 source, 2.1 Ci of americium-beryllium, or 354 mCi of radium-beryllium would be required. The choice of the californium-252 source, therefore, results in a significant reduction in radioactivity, and consequently in the shielding requirements. A device based on a californium-252 source is, therefore, much more compact in size and of much less weight, compared to a device that uses any alternative isotopic neutron source. The short-half life problem of the californium-252 source is partially compensated for by the fact that the source is commercially widely available at a reasonably low cost. Therefore, the replacement of the source, expected to be required no more than three times during the life-time of the reactor, does not constitute a significant addition to the cost of the device. The effect over time of the change in the source strength can be accommodated for through software, or by providing a dummy reference system to which measured signals can be compared. The latter alternative is possible because of the low cost of the device. A BF_3 thermal neutron detector is proposed in the design for economical reasons. The more efficient, and more expensive He-3 detector is, however, preferred. In order to improve the signal-to-noise ratio, a cadmium sleeve should be used to cover the surface that is not directly exposed to neutrons reflected from the tank. A standard electronics counting system can be used with this device.

Conclusions

This paper has demonstrated the feasibility of a neutron reflection method for gadolinium concentration monitoring in the tanks of the liquid poison injection shutdown system of a CANDU reactor, and has presented a conceptual design of a suitable device. The method is non-intrusive and uses off-the-shelf equipment. The signal obtained from the device provides a direct measure of the neutron absorption ability of the poison solution contained in the tank. The device can also scan the tank to monitor gadolinium precipitation. Further work is being undertaken to construct a prototype system suitable for installation at an operating CANDU reactor.

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Notes and References

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