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Development of Gadolinium (neutron poison) monitoring system for fuel reprocessing facilities: Computational model and validation with experiments

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ABSTRACT

Gadolinium, a strong neutron absorber is a good choice of material for use as soluble neutron poison for nuclear criticality control in nuclear fuel cycle facilities. During operation, it's availability has to be ensured in order to maintain the sub-critical state of the system. Towards this, periodic monitoring of presence of Gadolinium, a monitoring system was developed. The technique used is based on thermal neutron attenuation characteristics of the medium considered under study. As the design involves, choice of neutron source, moderator assembly and neutron detector sensitivity, Monte Carlo simulation was employed to optimize the design parameters. The details of simulation and experiments carried out to study the feasibility of the method is described. Results of measurements and comparison with computational model developed are also presented in detail. The results indicate that the proposed system can be successfully deployed for remote online monitoring of neutron poison in Fuel Reprocessing.

1. Introduction

Nuclear facility handling fissile materials are required to be managed such a way as to ensure criticality safety during normal as well as mal operating operations. It depends on many parameters that include mass of fissile materials, concentration, geometry, volume, enrichments of fuel and density. In addition, presence of moderators, absorbers and reflectors also affects the sub-criticality of the system. Such parameters can be controlled by engineered and/or administrative measures during operation of plant. Materials with high neutron absorption cross sections, known as ‘neutron poisons’ are also used for criticality control. Neutron poisons are most effective thermal neutron absorbers, the most commonly used are boron, cadmium and Gadolinium either in soluble or insoluble form.

Of these, Gadolinium (^{157}Gd) is a strong neutron absorber ($\sigma_{th} = 2.54 \times 10^5$ b) and effectively used in fuel reprocessing plant where high concentration of fissile solutions is being handled by addition of neutron absorber. (Lloyd et al., 1972). However for effective criticality control, the concentration of natural Gadolinium in the solution is required about 1.5 kg/m^3 in handling Plutonium rich fissile solution at nuclear fuel cycle facility.

The effectiveness of criticality control has to be insured by periodic measurement of the Gadolinium. Many offline methods used for this purpose such as atomic absorption, mass spectroscopy techniques (Hussein et al., 1987). The procedure, in addition to being manual and subject to administrative errors, does not provide an indication of the effectiveness of the system in terms of neutron absorption behavior of the system. Further, this procedure is also time consuming. It is desirable, therefore to develop a monitoring device of the poison concentration and such a system would minimize the manual effort and improve the results, in addition to remote as well as avoid handling high active solution.

In this work, a non-intrusive method for monitoring of the Gadolinium in the process tank is presented. This method is based on thermal neutron attenuation. The design of Gadolinium monitoring system involves various parameters such as choice of neutron source and strength, moderator assembly and type and sensitivity of neutron detector. Before employing in the field, the feasibility of system has to be assessed by simulating the design and optimizing the parameters. This is accomplished using Monte Carlo code, MCNP (Briesmeister, 2000). Based on the simulation results, experiments were carried out to measure the concentration of Gadolinium present in a cylindrical tank and the results are compared with the simulation.

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Table 1
Different isotopes of Gd and their abundance and thermal neutron capture cross sections.

Gd isotope	Natural Abundance (%)	σ_a (thermal) (barn)
Gd-152	0.20	1050
Gd-154	2.18	85
Gd-155	14.80	60,700
Gd-156	20.47	1.71
Gd-157	15.65	254,000
Gd-158	24.84	2.01
Gd-160	21.86	0.765

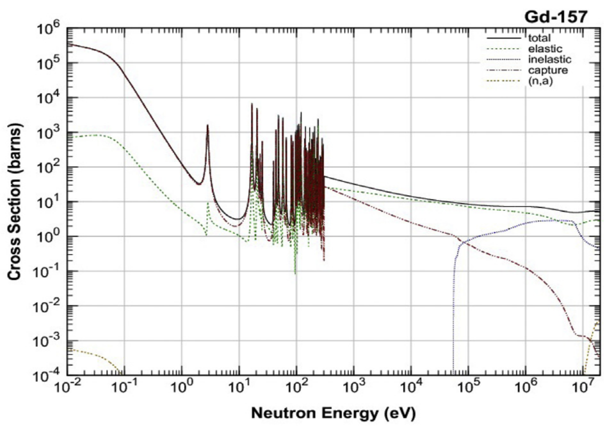

Fig. 1. Neutron absorption cross section data Gd-157.

2. Materials and methods

2.1. Natural Gadolinium

Natural Gadolinium has seven isotopes and the major isotopes are having abundances in the range of 14.8–24.8% for ^{155}Gd - ^{160}Gd (Table 1). The variation of nuclear cross section of ^{157}Gd with energy is plotted in Fig. 1 (Leinweber et al., 2006).

It is more obvious from Table 1 and Fig. 1 that thermal neutron absorption cross section of Gadolinium isotopes vary in the range of 1.05E + 03 to2.54E + 05 b.

2.2. Neutron source – (Am-Be)

Am-Be source which is a fast neutron source emitting neutrons of energy 100 keV to 11 MeV with an average energy of 4.5 MeV is widely used for neutron transmission studies because it is a readily lab source for neutrons and has a very long half life of 433 years (Igwesi and Thomas, 2013). The source strength is 2.56E + 06 n/s per Ci (see Fig. 2).

3. Monte Carlo simulation

Monte Carlo simulation studies were carried out using MCNP code. It is a general purpose, continuous energy, generalized geometry, time dependent, coupled neutron/photon/electron Monte Carlo transport code. Using this code the neutron counts/sec values were estimated from (n, α) reaction. F4 tally was used to estimate the counts. F4 tally calculates the average flux in a cell here inside the detector and gives the results in counts/neutrons. Nuclear Reaction rate (count rate) is calculated by multiplying the calculated fluxes ($\varphi_{MC}(r, \Omega, E)$) with the corresponding microscopic cross sections σ (E) by the relation,

$$r_{MC} = \frac{1}{V}\int_{v} dV \int_{4\pi} d\Omega \int_{0}^{\infty} \varphi_{MC}(r, \Omega, E)\sigma(E)dE$$

where V is the detector volume.

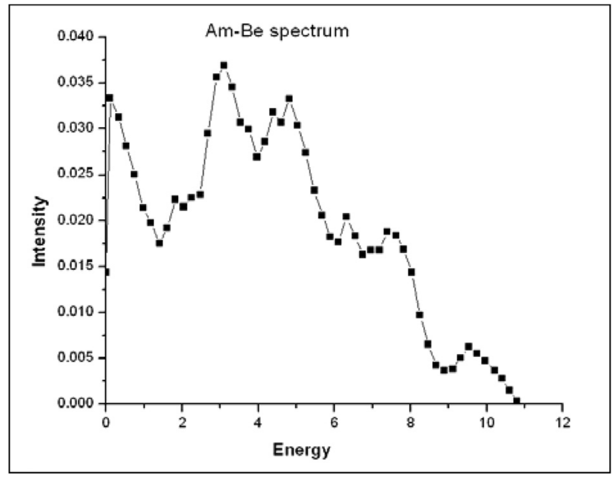

Fig. 2. Am-Be neutron energy spectrum.

3.1. Choice of neutron detector

The detectors used for the present simulation is BF_3 detector, filled with 90% enriched ^{10}B. It detects neutrons by means of (n, α) reaction and has reaction cross section (σ_{th} = 3800 b) for thermal neutron. Hence, these detectors are surrounded by High Density Poly Ethylene (HDPE) to improve the sensitivity.

3.2. Thermal neutron attenuation method

This is a direct method for studying the thermal neutron absorption ability, and it is well known that the neutron absorption is related to concentration of Gadolinium. It requires a strong neutron emitting source with enough thermalizing assembly. The thermalized neutrons can then be utilized to measure the concentration. This method is simple, capable of providing continual measurements, and relatively maintenance free. Fig. 3 shows the schematic diagram explaining the basic principle of the method.

3.3. Modeling in MCNP

The modeled geometry in MCNP is shown in Fig. 4(a) and (b). In this model, the source considered was a 37 GBq Am-Be point source and the cylindrical storage tank of length 26 cm and diameter 16 cm with 6 mm thick SS is modeled for simulation.

The detector assembly consists of a rectangular AISI-304L assembly with high density polyethylene (HDPE) inside. The rectangular assembly is 40.5 cm in length and 43 cm in width with a height of 60 cm. The thickness of the SS used is 0.2 cm. The HDPE block has three holes of 9.75 cm diameter each. A SS cylinder with height of 60 cm and diameter of 9.6 cm is inserted into each hole. The SS cylinders have within it a HDPE block which is machined to obtain a hole of 5.2 cm diameter upto a depth of 48 cm. There is a 0.1 cm thick lining of Cadmium on the outer side of the SS cylinders. The dimensions and characteristics of the detectors are given below:

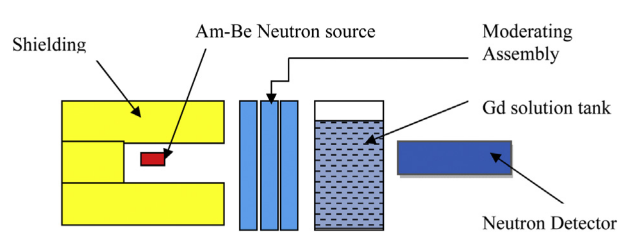

Fig. 3. Schematic representation of the thermal neutron attenuation method.