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Technical notes

Potential for the use of a liquid activation circulation material to measure the neutron flux in a high gamma-ray background

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ABSTRACT

The measurement of radiation fields in or near nuclear reactors is important for estimating radiation dose rates and damage, and for developing solutions for shielding problems. Such measurement mush be made in a difficult environment. Many types of detectors have been widely used in reactors to monitor the neutron flux. The activation detector is the most convenient, especially against a high gamma-ray background. In this study, liquid activation material is introduced to obtain a continuous measurement, and a rhodium solution is used to measure the reaction rate. There are three regions in this concept: the activation region, transfer or circulation region, and measurement or detection region. In this study, $Rh_2(SO_4)_3$ with the concentration of 100 g/l is used as the solution. To obtain the maximum reaction rate, an optimum flow rate of 80 cm^3/s was applied. Results showed a maximum counting rate in the equilibrium condition of about 18.6 cps. We conclude that it is possible to use a liquid activation method to measure the neutron flux in a high γ-ray background.

1. Introduction

In March 2011, an accident occurred at the Fukushima Daiichi Nuclear Power Plant in Japan. The accident resulted in core damage and was classified as the 7th or the highest level of nuclear accident [1], according to the International Nuclear Event Scale (INES). The situation of core melt consists mainly of fuels and control rods. According to this condition, fuel debris was formed and distributed, it was assumed, not only within the reactor pressure vessels but also in the primary containment vessels. For the efficient and appropriate removal of the fuel debris, it is necessary to understand its characteristics. It is important to consider the potential for re-criticality accidents during the fuel-removal procedure, and essential to ensure that the fuel remains in a subcritical condition. There are several methods for monitoring sub-criticality and detecting re-criticality including: using gas processing fission product gammas, a detection system with a neutron detector or a monitoring system for a liquid waste treatment and cooling facility.

Many types of neutron detectors can be used to measure the neutron flux, including gas proportional detectors, scintillation neutron detectors, semiconductor neutron detectors, neutron activation detectors, and fast neutron detectors. Each type has specific characteristics, and which is used depends on the intended purposes. In the case of thermal neutron flux, measurement can be conducted using an ionization chamber, fission counter tubes, scintillation detectors, and activation detectors. However, most thermal neutron detectors can only be used in low levels of gamma-ray background. They cannot work effectively under conditions where there is a large contribution of gamma ray. Among thermal neutron detectors, the activation method is the most convenient to use, and it measures thermal neutron flux with a high degree of precision and spatial resolution [2]. Activation detectors are also indispensable for measuring the absolute value of thermal neutron flux.

Actually, it is impossible to measure the neutron flux directly with an activation detector, even though the neutron flux is an important theoretical and experimental concept in reactor physics. The only physical value that can be measured is the reaction rate which is the value of the reaction with the detector composition materials. The gamma ray background level is high in the measurement of nuclear spent fuel [3]. Therefore, high gamma-ray background also appears in Fukushima Daiichi Nuclear Power Plant. In this study, we suggest the use of a neutron activation detector to measure the neutron flux. The advantages of using the activation method include [2]:

- The neutron field in the object of measurement is not disturbed and the resolution for measuring neutron flux is high.

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Table 1
Several candidate stable nuclides for liquid circulation in activation method.

Stable nuclide	Natural abundance (%)	Reaction	$\sigma_{capture}$ (barn)	Resulting nuclide	Half-life (s)
^{7}Li	92.4	(n,γ)	45.4×10^{-3}	^{8}Li	838.75×10^{-3}
^{11}Bi	80.1	(n,γ)	5.075×10^{-3}	^{12}Bi	20.2×10^{-3}
^{15}N	0.364	(n,γ)	24.26×10^{-6}	^{16}N	7.13
^{27}Al	100	(n,γ)	230.3×10^{-3}	^{28}Al	134.7
^{36}S	0.01	(n,γ)	150.5×10^{-3}	^{37}S	303
^{54}Cr	2.365	(n,γ)	404.9×10^{-3}	^{55}Cr	209.82
^{103}Rh	100	(n,γ)	133.2	^{104}Rh	42.3
^{136}Xe	8.857	(n,γ)	130×10^{-3}	^{137}Xe	229.08
^{160}Gd	21.86	(n,γ)	785.5×10^{-3}	^{161}Gd	219.6

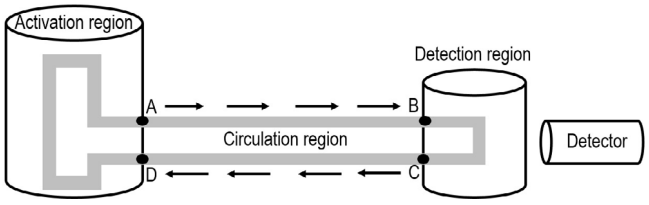

Fig. 1. Schematic design of liquid circulation in activation method.

Table 2
Detailed information of ^{103}Rh isotope.

Parameters	Value	Unit
Half-life of ^{104}Rh	42.3	s
Decay constant (λ)	0.0164	s^{-1}
Thermal microscopic capture cross section (σ)	133.2	barn
Branching ratio (γ)	0.014	
Detector efficiency (ε)	25	%
Neutron flux (ϕ)	100	n/cm^2/s
Concentration of ^{103}Rh	100	g/l

- Measurement of the neutrons can be performed in the presence of gamma ray.
- The equipment used for the measurement of induced radioactivity is simple and cost effective.

The disadvantages of an activation detector include:

- The resolution of neutron energy is not high, since the activation reaction is caused by neutrons whose energy lies within a specific energy range.
- Handling and interpretation of experimental data are complicated, since corrections and calibrations are necessary.
- Online measurement is difficult, since it takes considerable amount of time to obtain the results after irradiation.

As mentioned above, the difficulty of online measurement is a drawback of the activation method, especially when gold foil is used. Therefore, we propose a new method for measuring the reaction rate using a liquid circulation activation method. By introducing liquid circulation, we can obtain the continuous or online measurement. The purpose of this study was to show the feasibility of using liquid material in activation method to measure the reaction rate.

2. Material and calculation method

2.1. Concept of liquid circulation method

For many activities, the gold foil activation method is widely utilized to measure thermal neutrons. This method became popular because gold (Au) is 100% composed of ^{197}Au. The decay pattern of an induced nuclide ^{198}Au generated by the activation of ^{197}Au is simple, and the half-life of ^{198}Au is suitable for measuring radioactivity [2].

First, the Au is irradiated in the activation region for a certain period of time. Subsequently, there is a waiting time to reduce the radiation dose before the reaction rate is measured in the detection region. In this study, we propose a new concept by introducing liquid circulation into the activation method to eliminate the wait time before measurement. Fig. 1 presents a schematic of liquid circulation in the activation method. There are three regions: the activation region, circulation region, and measurement region. The liquid solution circulates in a loop from the activation region to the measurement region, as shown in Fig. 1.

2.2. Rhodium characteristics

The material used in the activation method should be a stable nuclide, with a simple decay scheme, a large abundance in nature, and a suitable half-life for measuring radioactivity. Table 1 shows several isotope candidates and also indicates that ^{103}Rh is the most acceptable application in this study. It has 100% natural abundance, a thermal neutron capture cross section of about 133 barn, and half-life of 42.3 s. Table 2 presents information about the decay of ^{103}Rh (n,γ) reaction for thermal neutrons and Fig. 2 shows the decay scheme of ^{103}Rh. It can be seen that when thermal neutrons are captured, ^{104m}Rh (^{104}Rh) nuclei are produced and gamma rays are directly emitted at the same time. ^{104m}Rh nuclei de-excite to ^{104}Rh nuclei with the emission of gamma rays in decay constant λ_1, while ^{104}Rh nuclei from ^{103}Rh (n,γ) reaction and 104mRh de-excitation decay to ^{104}Pd with decay constant λ_2, emitting β and γ rays.

While there are several compounds of rhodium, listed in Table 3, only some of them are in the form of a solution. In this case, $Rh_2(SO_4)_3$ was selected due to its solubility in water and ethanol and its high concentration up to 100-g Rh/liter.

2.3. Detection system in the measurement region

In the activation method using gold wires, the activation region is irradiated and, once the irradiation dose has fully decreased, the ^{198}Au is measured by a High-purity germanium (HPGe) detector. Gamma ray spectrometers utilizing germanium semiconducting detectors are widely used around nuclear facilities. Therefore, we used an HPGe detector in this reference study.

Sensitivity and efficiency are the important variables in a detector. The sensitivity of the detector was evaluated in the thermal neutron flux, where it registered 1 count/s in the 596-keV photo peak for an average neutron flux of 10 n/cm^2/s using a 25% relative efficiency germanium detector; this is equivalent to a low neutron dose rate equivalent of 0.4 μSv/h [4] .

2.4. Calculation method

Fig. 1 shows our schematic design for including liquid circulation in the activation method. There are three regions: the activation region (region I), circulation region (region II), and detection region (region III). It is necessary to recognize the characteristics and decay of the rhodium