

On-line detection of key radionuclides for fuel-rod failure in a pressurized water reactor



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HIGHLIGHTS

- In this work, we introduce a method of on-line fuel rod failure detection using LaBr₃(Ce) detector.
- As there were many kinds of radionuclides in the primary coolant after the fuel rod failure and the resolution of LaBr₃(Ce) detector was not as good as HPGe detector, so it need to select the radionuclides to be monitored in the measurement of fuel rod failure in order to provide reliable and accurate results.
- Nuclides which enter the primary coolant when fuel rod failed was analyzed, and then ¹³⁵Xe and ⁸⁸Kr were eventually chosen as the key radionuclides for on-line monitoring of fuel rod failure.
- The key radionuclides (¹³⁵Xe and ⁸⁸Kr) selected for on-line detection of fuel rod failure of PWR has the advantages of being less vulnerable to interference factors and high confidence.

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ABSTRACT

For early on-line detection of fuel rod failure, the key radionuclides useful in monitoring must leak easily from failing rods. Yield, half-life, and mass share of fission products that enter the primary coolant also need to be considered in on-line analyses. From all the nuclides that enter the primary coolant during fuel-rod failure, ¹³⁵Xe and ⁸⁸Kr were ultimately chosen as crucial for on-line monitoring of fuel-rod failure. A monitoring system for fuel-rod failure detection for pressurized water reactor (PWR) based on the LaBr₃(Ce) detector was assembled and tested. The samples of coolant from the PWR were measured using the system as well as a HPGe γ -ray spectrometer. A comparison showed the method was feasible. Finally, the γ -ray spectra of primary coolant were measured under normal operations and during fuel-rod failure. The two peaks of ¹³⁵Xe (249.8 keV) and ⁸⁸Kr (2392.1 keV) were visible, confirming that the method is capable of monitoring fuel-rod failure on-line.

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1. Introduction

The activity concentration of fission products is the second dominant contributor to prevailing radiation levels in typical PWRs. Activity concentrations of fission products in the primary coolant are easy to obtain from γ emission spectra. These fission products include all isotopes of inert gases (Ivanov, 1998) and volatile iodine. Currently, the high-purity germanium (HPGe) detector is widely used for on-line γ -ray spectrum analyses of primary coolant samples (El-Jaby et al., 2010; Lewis et al., 2007). The HPGe detector can accurately measure the activity concentrations

of the fission products, albeit at low temperature and in the absence of vibration during measurements. Moreover, the current software used with the HPGe spectrometer needs user input when analyzing spectra.

The LaBr₃(Ce) detector is attracting interest for γ -ray spectroscopy (Iltis et al., 2006; Nicolini et al., 2007) because of its excellent energy resolution (2.5–3% at 662 keV) at room temperature, obtained when assembled with a photomultiplier tube (PMT). This make it a major competitor to semiconductor detectors. Because it can overcome the deficiency of the above method, the LaBr₃(Ce) detector has been chosen for on-line monitoring of γ -ray spectra for fuel-rod failure.

There are many kinds of radionuclides in the primary coolant (Awan et al., 2011; Aliberti et al., 2006) as fuel rods begin to fail. Although the energy resolution of a LaBr₃(Ce) detector is not as good as a HPGe detector, selecting those radionuclides that

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provide reliable and accurate results is needed if monitoring of fuel-rod failure with such detectors is to be viable.

2. Methodology

Results from both experiments and operations indicate that the release rate of fission products is proportional to its cumulative amount in the fuel (Iqbal et al., 2007). Furthermore, two rate equations can be developed for some radionuclides in the fuel assembly and coolant. Denoting N_f and N_L as the number of nuclides in the fuel and coolant, respectively, then:

$$\frac{dN_f}{dt} = NF - \lambda N_f - \gamma N_f, \quad (1)$$

$$\frac{dN_L}{dt} = \gamma N_f - \lambda N_L - K_d N_L, \quad (2)$$

where N is the fission rate, F the fission yield, λ the decay constant, γ the escape rate coefficient, and K_d the decrease rate of the radionuclide (including neutron absorption, absorption of ion exchange resin, and leakage).

The activity concentrations of fission products in the primary coolant [Eq. (2)] mainly depend on three factors: 1) the escape rate of fission products from the core, 2) the decay of the radionuclides, and 3) the effects of the purification system. In addition, factors to affect the activity concentrations of key radionuclides in the primary coolant also included the influence of fission products with similar energy and the effect of activation products in the primary coolant.

2.1. Interference of purification system

In general, a pressurized water reactor (PWR) is equipped with a purification system (ion exchange system, see Fig. 1) for the primary coolant. When fuel-rod failure occurs in a PWR, the activity concentration of a radionuclide in the primary coolant is

$$C(t) = \frac{MC_0 - \int_0^t C(t)\eta Q dt}{M}, \quad (3)$$

where M is the total mass of the primary coolant, C_0 the initial activity concentration of the radionuclide in the primary coolant, Q the flow rate through the purification system, and η the efficiency of the purification system.

Assuming Q , M , and η are time independent, the solution of Eq. (3) takes the form

$$C(t) = \exp\left(-\frac{\eta Q}{M}t + c\right).$$

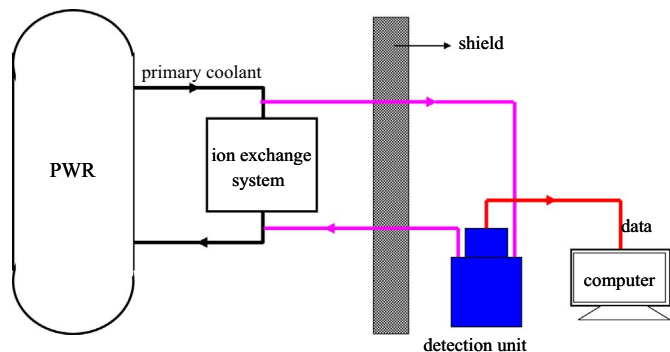


Fig. 1. Schematic showing the different devices of the on-line detection system.

Then $c = \ln C_0$ obtains from $C(0) = C_0$ at $t = 0$. Hence, under continuous purification, the activity concentration of a radionuclide in the primary coolant can be expressed as

$$C(t) = C_0 \exp\left(-\frac{\eta Q}{M}t\right). \quad (4)$$

Compared with other types of fission products, inert gases and halogens are volatile and their relative fractions in releases are large. Hence, they are suitable for on-line detection of failing fuel-rods. The longest time period for monitoring the fuel-rod failure is assumed to be no more than 5 min, and the efficiency of the purification system in removing halogens (such as ^{131}I) with a long half-life must be taken to be its maximum value (90%). Because the activity concentrations of leaking halogens during fuel-rod failure begins to grow from an allowable value, we set M as 100 m^3 , Q as $27 \text{ m}^3/\text{h}$, and hence the activity concentration is $C(t=5 \text{ min}) = 0.9795C_0$ calculated from Eq. (4) with an elapsed time of 5 min. Clearly, the influence of the purification system on the activity concentration of halogens is less than 3% during this period and insignificant. Hence there is no need to consider the effects of purification system when calculating the activity concentrations of key radionuclides.

2.2. Decay of radionuclides

When the PWR is operating, primary coolant flows constantly through the sampling unit. The activity concentrations of radionuclides with short half-lives in the primary coolant change in passing through the sampling unit. However, the activity concentrations of radionuclides with long half-lives in the primary coolant are irrelevant over the temporal and spatial scales involved. Hence, the activity concentration of the radionuclide for each point in the sampling unit is believed to be in a dynamic state of balance.

Let us denote the distance from the entrance of sampling unit (point A), to the point of observation (point B) as l . If the activity concentration of a radionuclide in the primary coolant is $a(l)$, the net count under the full-energy peak corresponding to the radionuclide is

$$C_{\text{count}} = PW\Delta T \int_0^L a(l)\epsilon(l)dl, \quad (5)$$

where P is the emission probability, W the cross-sectional area of the sampling unit, ΔT the measurement time for the radionuclide, L the length of the sampling unit, $\epsilon(l)$ the detection efficiency of γ -rays at point B, and $a(l)$ a function of l for the sampling unit. If the activity concentration of radionuclide in point A is a_0 and the flow rate of coolant is v , the relationship between $a(l)$ and a_0 at point B is

$$a(l) = a_0 \exp\left(-\frac{\ln 2}{T_{1/2}} \cdot \frac{l}{v}\right), \quad (6)$$

where $T_{1/2}$ is the half-life of the radionuclide. There are large differences in half-lives for typical fission products, such as Br and I, of the inert gases. Hence, the half-lives of key radionuclides should be longer than the sampling time (about several seconds). This implies $a(l) \approx a_0$ in Eq. (6), and the value of $a(l)$ can be regarded as constant.

Applying this result in Eq. (5), the activity concentrations of key radionuclides to be monitored in the primary coolant at the entrance point A are

$$a_0 = \frac{C_{\text{count}}}{PW\Delta T \int_0^{l_0} \epsilon(l)dl} \quad (7)$$

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