CRITICALITY DETECTION METHOD
BASED ON FP GAMMA RADIATION MEASUREMENT

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ABSTRACT

A method is proposed to evaluate the extent of subcriticality of an accident-damaged nuclear reactor. In this method the radiation ratio of two FP rare gas nuclides, ^{88}Kr and ^{133}Xe, is measured. From the measured value, the value in the core is estimated by correcting for the time required by the gases to diffuse from the core to the measuring point. A simple expression for neutron multiplication factor has been derived, which uses the corrected ^{88}Kr-to-^{133}Xe radiation ratio and the ^{88}Kr-to-^{133}Xe fission yield ratios from ^{244}Cm and ^{235}U but no information on the amount or distribution of fissile materials, making the proposed method very simple. The method has the advantage that FP rare gases can easily leak from the reactor core through many openings and gaps, reaching germanium counters without reacting with other materials.

Keywords: **criticality safety, FP rare gases, gamma radiation, spontaneous fission, induced fission**

1. INTRODUCTION

In the decommission and disposing of an accident-damaged reactor such as those at Fukushima No.1 Nuclear Power Plant in Japan, it is necessary to confirm that the reactor is subcritical enough to allow workers to approach the site. At the Fukushima site, it is planned to measure neutron flux and xenon gas density; when these quantities rapidly increase or top certain values, an alarm is to be issued. Such methods do not guarantee the reactor's subcriticality, and there is a possibility that evacuation of people is too late.

In an accident-damaged reactor, many structures, such as fuel pins, containment and pressure vessels, are greatly damaged, enabling many kinds of FP gases to leak out of the reactor core. In particular, rare gases, such as Kr and Xe, do not react with other materials, can leak through many gaps and openings, and reach germanium counters far distant from the core.

This paper proposes a method to evaluate the extent of subcriticality of an accident-damaged reactor by measuring the radiation ratio of two kinds of FP rare gases, ^{88}Kr and ^{133}Xe, without detailed information on the amount or distribution of fuel materials. This method can be also applied to criticality monitoring at fuel reprocessing facilities where spent fuels are treated in an unsealed state. It is also thought to be applicable to spent fuel permanent storage facilities where spent fuel pins may lose their shape due to long-term corrosion.

2. CRITICALTY EVALUATION MODEL

For the sake of simplicity, it is assumed that only ^{235}U and ^{244}Cm exist as a fissile material and a spontaneous-fission material, respectively; other such materials can be added to the spontaneous fission neutron source term and induced fission neutron source term, with the basic way of thinking remaining unchanged. The fuel pellets contain minor actinide ^{244}Cm; this nuclide suffers a spontaneous fission and produces about 2.71 secondary neutrons, which in turn induce ^{235}U fissions. The total neutron source N^0 in the core is expressed by

$$N^0(k) = S(^{244}Cm) + S(^{244}Cm)k + S(^{244}Cm)k^2 + S(^{244}Cm)k^3 + \cdots$$

$$= S(^{244}Cm) + \frac{k}{1-k}S(^{244}Cm) = S(^{244}Cm) + S(^{235}U), \quad (1)$$

where $S(^{244}Cm)$ is the neutron source density due to spontaneous ^{244}Cm fission, which can be esteemed as an external source; $S(^{235}U)$ is the neutron source density due to induced ^{235}U fissions; k is the neutron multiplication factor of the reactor; these neutron sources are spatially-averaged ones with importance as a weighting factor.

The No. densities of FP nuclides, ^{133}Xe and ^{88}Kr, satisfy the following balance equations:

$$\frac{dN^0_{^{133}Xe}}{dt} = Y_{^{244}Cm}(^{133}Xe) \times \frac{S(^{244}Cm)}{\nu_{SF}} + Y_{^{235}U}(^{133}Xe)$$

$$\times \frac{S(^{244}Cm)}{\nu_{IF}} \times \frac{k}{1-k} - \lambda_{^{133}Xe} N^0_{^{133}Xe} \quad (2)$$

and

$$\frac{dN^0_{^{88}Kr}}{dt} = Y_{^{244}Cm}(^{88}Kr) \times \frac{S(^{244}Cm)}{\nu_{SF}} + Y_{^{235}U}(^{88}Kr)$$

$$\times \frac{S(^{244}Cm)}{\nu_{IF}} \times \frac{k}{1-k} - \lambda_{^{88}Kr} N^0_{^{88}Kr} \ , \quad (3)$$

where $N^0_{^{133}Xe}$ is the No. density of ^{133}Xe in the reactor core, $\lambda_{^{133}Xe}$ its decay constant, $Y_{^{244}Cm}(^{133}Xe)$ is the fission yield of ^{133}Xe due to ^{244}Cm fission, ν_{SF} and ν_{IF} the numbers of secondary neutrons due to spontaneous ^{244}Cm fission and induced ^{235}U fission, respectively.

When the FP nuclide No. densities are assumed to be in an equilibrium state, the LHSs of Equations (2) and (3) equal zero. Then the following equations are obtained;

$$\lambda_{^{133}Xe} N^0_{^{133}Xe} = Y_{^{244}Cm}(^{133}Xe) \times \frac{S(^{244}Cm)}{\nu_{SF}} + Y_{^{235}U}(^{133}Xe) \times \frac{S(^{244}Cm)}{\nu_{IF}} \times \frac{k}{1-k} \quad (4)$$

$$\lambda_{^{88}Kr} N^0_{^{88}Kr} = Y_{^{244}Cm}(^{88}Kr) \times \frac{S(^{244}Cm)}{\nu_{SF}} + Y_{^{235}U}(^{88}Kr) \times \frac{S(^{244}Cm)}{\nu_{IF}} \times \frac{k}{1-k} \quad (5)$$

The ratio of equation (5) to equation (4) becomes

$$R_0(^{88}Kr/^{133}Xe) \equiv \frac{\lambda_{^{88}Kr} \times N^0_{^{88}Kr}}{\lambda_{^{133}Xe} \times N^0_{^{133}Xe}}$$

$$= \frac{Y_{^{244}Cm}(^{88}Kr)/\nu_{SF} + Y_{^{235}U}(^{88}Kr)/\nu_{IF} \times \frac{k}{1-k}}{Y_{^{244}Cm}(^{133}Xe)/\nu_{SF} + Y_{^{235}U}(^{133}Xe)/\nu_{IF} \times \frac{k}{1-k}} . \quad (6)$$

Transformation of Equation (6) leads to the following equation:

$$\frac{k}{1-k} = \frac{Y_{^{244}Cm}(^{133}Xe) \times R_0(^{88}Kr/^{133}Xe) - Y_{^{244}Cm}(^{88}Kr)}{Y_{^{235}U}(^{88}Kr) - R_0(^{88}Kr/^{133}Xe) \times Y_{^{235}U}(^{133}Xe)} \times \frac{\nu_{IF}}{\nu_{SF}} . \quad (7)$$

Quantity $R_0(^{88}Kr/^{133}Xe)$ is the ^{88}Kr-to-^{133}Xe radiation ratio in the reactor core; the rare gas atoms comprise those resulting from ^{244}Cm fission and ^{235}U fission, so the ratio depends on the ^{235}U-to-^{244}Cm fission ratio, i.e., as Equation (7) shows, when the reactor approaches criticality, the ratio $R_0(^{88}Kr/^{133}Xe)$ approaches $Y_{^{235}U}(^{88}Kr)/Y_{^{235}U}(^{133}Xe)$ because 235U fission is dominant, and when neutron multiplication factor k approaches zero, the ratio approaches $Y_{^{244}Cm}(^{88}Kr)/Y_{^{244}Cm}(^{133}Xe)$ because ^{244}Cm fission is dominant in this case.

As shown in Figure 1, the ^{88}Kr-to-^{133}Xe yield ratio clearly changes from ^{235}U to ^{244}Cm, which means that, if only the ^{88}Kr-to-^{133}Xe radiation ratio in the reactor core can be measured, the extent of subcriticality of the reactor can be estimated without detailed information on the amount or distribution of fuel materials.

Further transformation of Equation (7) leads to

$$k = \frac{R_0(^{88}Kr/^{133}Xe)\times Y_{^{244}Cm}(^{133}Xe) - Y_{^{244}Cm}(^{88}Kr)}{\left(\frac{\nu_{SF}}{\nu_{IF}}\times Y_{^{235}U}(^{88}Kr) - Y_{^{244}Cm}(^{88}Kr)\right) - R_0(^{88}Kr/^{133}Xe)\times\left(\frac{\nu_{SF}}{\nu_{IF}}\times Y_{^{235}U}(^{133}Xe) - Y_{^{244}Cm}(^{133}Xe)\right)} \quad .(8)$$

Figure 2 shows how neutron multiplication factor k changes with the $R_0(^{88}Kr/^{133}Xe)$; the figure used fission yield data in Table 2; ν -values in Reference (2) were used:

$$\nu_{SF} = 2.712 \qquad \nu_{IF} = 2.466$$

As shown in the figure, the relationship between the two quantities is almost linear, making it easy to confirm the subcriticality of a reactor.

In this method, multiplication factor k can be calculated from $R_0(^{88}Kr/^{133}Xe)$, which is obtained by correcting the measured data $R(^{88}Kr/^{133}Xe)$ as described in the next section.

3. CORRECTION FOR MEASUREMENT POSITION

The above-mentioned radiation ratio $R_0(^{88}Kr/^{133}Xe)$ is the value in the core. It is impossible to directly measure such data in an accident-damaged core. Therefore, a method is described here to estimate the ^{88}Kr-to-^{133}Xe radiation ratio $R_0(^{88}Kr/^{133}Xe)$ using the value $R(^{88}Kr/^{133}Xe)$ measured some distance away from the core.

It takes some time for the FP gases to arrive at a germanium counter located some distance away from the reactor core. The radiation ratios, $R_0(^{88}Kr/^{133}Xe)$ and $R(^{88}Kr/^{133}Xe)$, therefore, have the following relationship.

$$\begin{aligned} R(^{88}Kr/^{133}Xe) &= \frac{\lambda_{^{88}Kr}\times N_{^{88}Kr}}{\lambda_{^{133}Xe}\times N_{^{133}Xe}} \\ &= \frac{\lambda_{^{88}Kr}\times N^0_{^{88}Kr}}{\lambda_{^{133}Xe}\times N^0_{^{133}Xe}}\, exp\left[-\left(\lambda_{^{88}Kr} - \lambda_{^{133}Xe}\right)\tau\right] \\ &= R_0(^{88}Kr/^{133}Xe)\, exp\left[-\left(\lambda_{^{88}Kr} - \lambda_{^{133}Xe}\right)\tau\right], \end{aligned} \quad (9)$$

where τ is the time required by the FP gases leaving the core to reach the germanium counter. Time τ can be obtained by measuring two kinds of FP gases, ^{133}Xe and ^{133m}Xe, which are considered to have the same diffusion speed. In addition, the fission yield ratios of these nuclides from ^{235}U and ^{244}Cm are almost the same.

$$Y_{^{235}U}(^{133}Xe)/Y_{^{244}Cm}(^{133}Xe) = 1.18 \quad (= 6.69/5.66)$$
$$Y_{^{235}U}(^{133m}Xe)/Y_{^{244}Cm}(^{133m}Xe) = 1.12 \quad (= 0.196/0.175)$$

By taking advantage of this, the following expression is obtained:

$$R_0(^{133m}Xe/^{133}Xe) = \lambda_{^{133m}Xe} \times N^0_{^{133m}Xe}/\left[\lambda_{^{133}Xe} \times N^0_{^{133}Xe}\right]$$
$$= \frac{Y_{^{244}Cm}(^{133m}Xe)S(^{244}Cm)+Y_{^{235}U}(^{133m}Xe)S(^{235}U)}{Y_{^{244}Cm}(^{133}Xe)S(^{244}Cm)+Y_{^{235}U}(^{133}Xe)S(^{235}U)}$$
$$= \frac{Y_{^{244}Cm}(^{133m}Xe)[S(^{244}Cm)+1.12S(^{244}Cm)]}{Y_{^{244}Cm}(^{133}Xe)[S(^{244}Cm)+1.18S(^{244}Cm)]}$$
$$\approx Y_{^{244}Cm}(^{133m}Xe)/Y_{^{244}Cm}(^{133}Xe) = 3.09\text{E-}2 \qquad (10)$$

The radiation ratio, $R(^{133m}Xe/^{133}Xe)$, measured by a germanium counter is expressed by

$$R(^{133m}Xe/^{133}Xe) = 3.09\text{E-}2\ exp\left[-\lambda_{^{133m}Xe} + \lambda_{^{133}Xe}\right]\tau. \qquad (11)$$

By using Equation (11), τ can be determined from the measured value, and by using it in Equation (9), the radiation ratio $R_0(^{88}Kr/^{133}Xe)$ can be obtained. Finally, multiplication factor k can be determined by Equation (8).

4. FIGURES

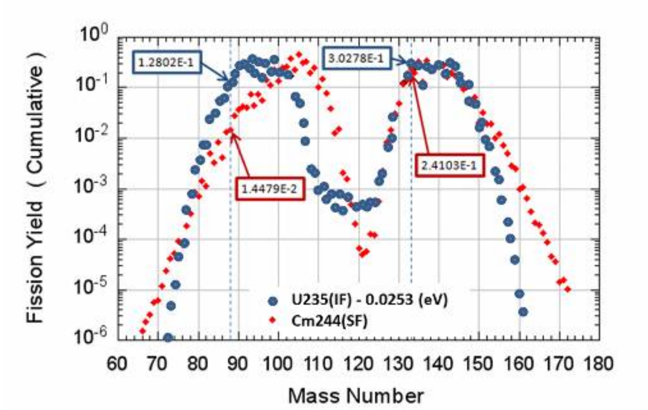

Figure 1. Fission yields data of ^{244}Cm and ^{235}U

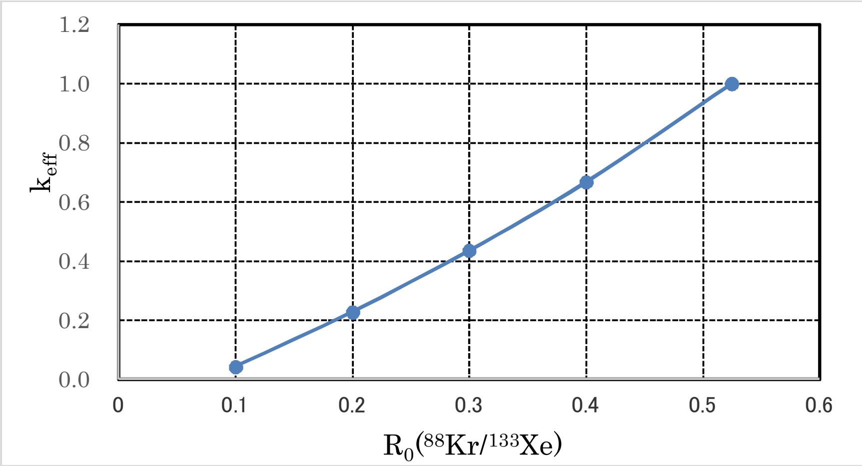

Figure 2. Dependence of Criticality on ^{88}Kr–to-^{133}Xe radiation ratio

5. TABLES

Table 1. Fission yield data of Kr and Xe nuclides from ^{235}U and ^{244}Cm and their half lives[1)]

Nuclide (half-life)	Precurser (half life)	γ ray [MeV] (Release rate, %)	^{235}U fission yield [%]	^{244}Cm fission yield [%]
^{85m}Kr (4.48 h)	^{85}Br (2.9 m)	0.305 (14)	1.40±0.11	0.212±0.034
^{87}Kr (76 m)	^{87}Br (55.6 s)	0.403 (50)	2.60±0.05	0.364±0.164
^{88}Kr (2.84 h)	^{88}Br (16.6 s)	2.392 (35)	3.51±0.08	0.423±0.271
^{131m}Xe (11.8 d)	^{131}I (8.04 d)	0.164 (1.9)	0.0318±0.009	0.0331±0.010
^{133m}Xe (2.19 d)	^{133}I (20.9 h)	0.233 (10)	0.196±0.125	0.175±0.111
^{133}Xe (5.25 d)	^{133}I (20.9 h)	0.081 (37)	6.69±4.28	5.66±3.62
^{135m}Xe (15.7 min)	^{135}I (6.61 h)	0.527 (81)	1.22±0.12	1.76±0.54
^{135}Xe (9.10 h)	^{135}I (6.61 h)	0.250 (90)	6.52±0.05	7.47±0.30
^{138}Xe (14.1 min)	^{138}I (6.5 s)	0.258 (30)	6.29±0.09	6.53±1.04

* γ ray emission probability [%], Y (^{235}U): Cumulative thermal fission yield of ^{235}U, Y (^{244}Cm): Cumulative spontaneous fission yield of ^{244}Cm.

6. CONCLUSIONS

A method has been proposed to confirm the subcriticality of an accident-damaged nuclear reactor. The radiation ratio in the reactor core of two FP rare gas nuclides, ^{88}Kr and ^{133}Xe, which are produced by spontaneous ^{244}Cm fission and induced ^{235}U fission, is estimated from the measured value at a point some distance away from the reactor core; the difference in the radiation ratio between the core and the measuring point results from the time necessary for the FP gases to diffuse from the reactor core to the measuring point. The necessary time is estimated by measuring the ^{133m}Xe -to-^{133}Xe radiation ratio, and can be used to correct the measured ^{88}Kr-to-^{133}Xe radiation ratio. Once the radiation ratio in the core has been determined, neutron multiplication factor of the reactor can be calculated using the fission yield ratios, $Y_{^{235}U}(^{88}Kr)/Y_{^{235}U}(^{133}Xe)$ and $Y_{^{244}Cm}(^{88}Kr)/Y_{^{244}Cm}(^{133}Xe)$, and the estimated ^{88}Kr-to-^{133}Xe radiation ratio.

This method has the advantage that the extent of subcriticality of the reactor can be estimated without knowing detailed information on the amount or distribution of fuel materials, and that rare gas nuclides, ^{88}Kr and ^{133}Xe, do not react with other materials, can easily leak through many gaps and openings, and reach germanium counters. Neutron multiplication factor k can be obtained by a simple expression using the ^{88}Kr-to-^{133}Xe radiation ratio and the above fission yield ratios, making it easy to monitor the extent of subcriticality.

In addition to accident-damaged reactors, this method is applicable to criticality monitoring at fuel reprocessing facilities where spent fuels are treated in an unsealed state. It is also thought to be applicable to spent fuel permanent storage facilities where spent fuel pins may lose their shape due to long-term cladding corrosion.

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