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Integral Test on Activation Cross Section of Tag Gas Nuclides Using Fast Neutron Spectrum Fields

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Activation cross sections of xenon and krypton isotopes were evaluated as tag gases to identify capsule rupture in liquid metal cooled fast reactors. The accuracy of the activation cross sections was investigated with several tag gas samples irradiated in the standard neutron field of JOYO and YAYOI reactors. Comparing the measured radioactivities and calculated values using YAYOI neutron fluence and activation cross section processed from the JENDL-3.2 cross section library yielded C/E values of approximately 0.86 to 2.6 for tag gas nuclides. The discrepancy between calculation and measurement appears due to the cross section uncertainty. This study confirmed the present accuracy of tag gas activation cross sections.

KEYWORDS: activation cross section, JENDL-3.2, tag gas, fast reactor, neutron spectrum, JOYO, YAYOI

I. Introduction

Unique blend ratios of stable xenon and krypton gases provide tag gases for the irradiation test capsules in fast reactors. When the capsule breaches, the gases are released into the reactor cover gas space through the sodium coolant. They are collected and analyzed for their isotopic ratios with mass spectrometry to identify the breached capsules. The tag gas nuclides are also activated through (n, γ) reactions during irradiation in the reactor and these activation products can be identified by measuring the gamma-ray spectrum of the argon cover gas.

Gamma-ray spectrometry generally has a higher sensitivity, possibly less than ppm, than mass spectrometry for tag gases diluted in the argon cover gas. Therefore, this method of measuring the activated tag gas nuclides is expected to be an alternative instrumentation technique for identifying failed capsules. The information obtained by this method will enhance the accuracy and reliability of on-line irradiation tests.

II. Irradiation Tests in JOYO and YAYOI

1. In-Core Tag Gas Release Test in JOYO

This method was demonstrated with in-core tag gas release tests carried out in the experimental fast reactor JOYO [1,2]. The tests, illustrated in **Fig. 1**, placed stainless steel capsules containing highly pressurized (> 30 MPa) tag gases in the JOYO Mk-II irradiation core. The tag gas specifications used for this experiment are shown in **Table 1**.

These capsules eventually breached via creep rupture and the krypton and xenon gases were released from the capsule into the reactor cover gas space. The gamma-rays from activated tag gas nuclides were measured with the On-line Gamma-ray Monitor [3] (OLGM), a high purity germanium (Ge) detector, which monitored the JOYO cover gas line as shown in **Fig. 2.**

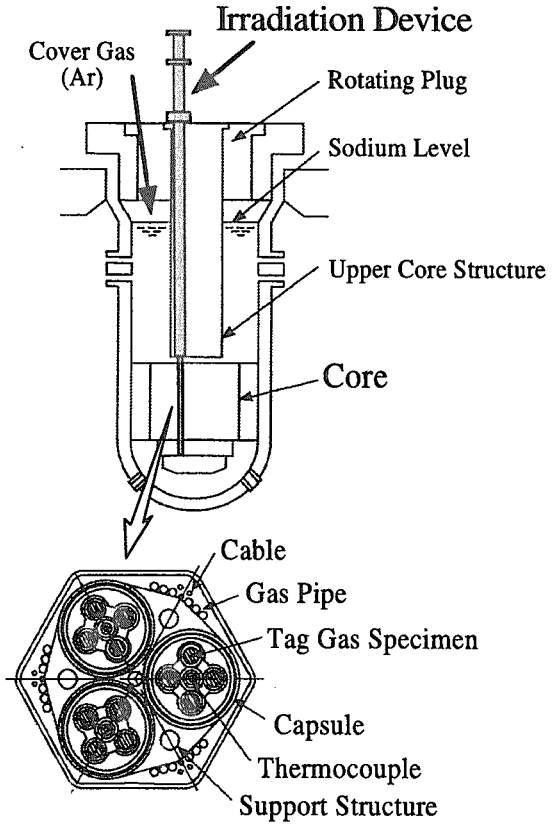

Fig. 1 In-Core Tag Gas Release Test Schematic in JOYO

The OLGM uses a charcoal bed to selectively adsorb xenon and krypton at room temperature and this enhances the detector sensitivity. The OLGM measurements need correction to obtain the absolute radioactivity per unit volume of argon cover gas. The calibration was conducted

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Table 1 JOYO Tag Gas Specifications

Tag Gas Volume (cc)		
Xe	Kr	He
1.9	1.9	52~62
Xe Isotope Ratio	Kr Isotope Ratio	
124/129 = 0.038 126/129 = 0.053 128/129 = 0.183 130/129 = 0.139 131/129 = 0.720 132/129 = 0.911 134/129 = 0.353	78/84 = 0.022 80/84 = 0.110 82/84 = 0.260 83/84 = 0.213 86/84 = 0.298	
Total Neutron Fluence = $0.75\sim3.3\times10^{21}$ n/cm^2		
Tag gas was released in a cover gas volume of $6.5\ m^3$ (6.5×10^6 cc)		

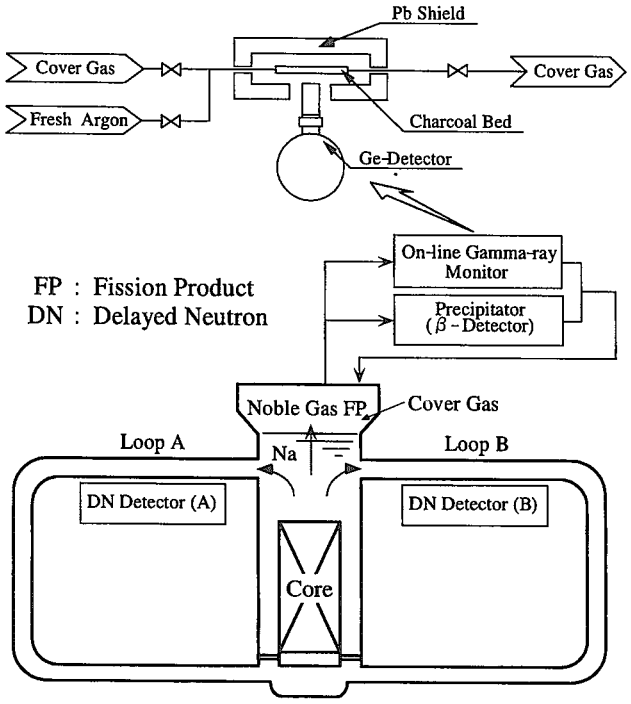

Fig. 2 JOYO On-line Gamma-Ray Monitor

using the sample gas which was taken directly from the reactor cover gas line soon after the capsule breach.

The measured gamma-ray spectrum and the trend of cover gas radioactivities for activation nuclides are shown in **Figs. 3 and 4**. It can be seen that the distinct photo-electron peaks of the activated nuclides ^{125}Xe, ^{127}Xe, ^{129m}Xe, ^{131m}Xe, ^{133}Xe and ^{135}Xe were distinguished with high resolution. These nuclides among the background fission products were clearly detected by the OLGM and this method was found to be applicable.

2. Cross Section Calibration Test in YAYOI

It is important to assure the accuracy of the tag gas activation (n, γ) cross section to apply this technique. Several tag gas calibration samples, which were contained in capsules, were irradiated in the standard neutron field of the fast neutron source reactor YAYOI [4] core center.

The cross sectional view of YAYOI and the position where tag gas capsules were loaded are illustrated in **Fig. 5**. The specifications of tag gas sample used in the experiment are shown in **Table 2**. Four types of tag gas sample were used in five irradiation tests at the center position of YAYOI Glory hole. In each irradiation test, a set of activation foils and wires (U, Np, Fe, Co, Ni, In, Al and Au) were placed near the tag gas capsules in order to evaluate the neutron flux at the irradiation position.

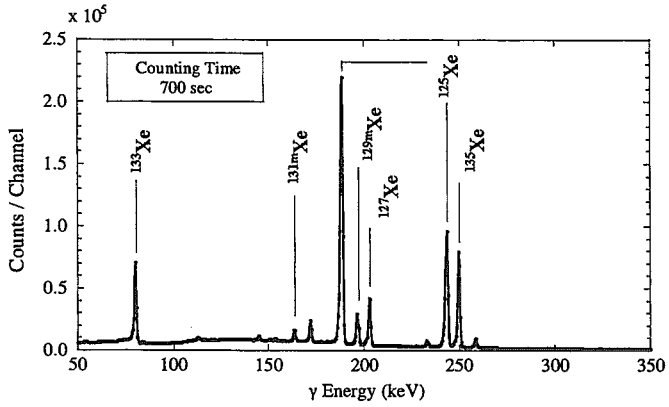

Fig. 3 Gamma-Ray Spectrum of JOYO Cover Gas

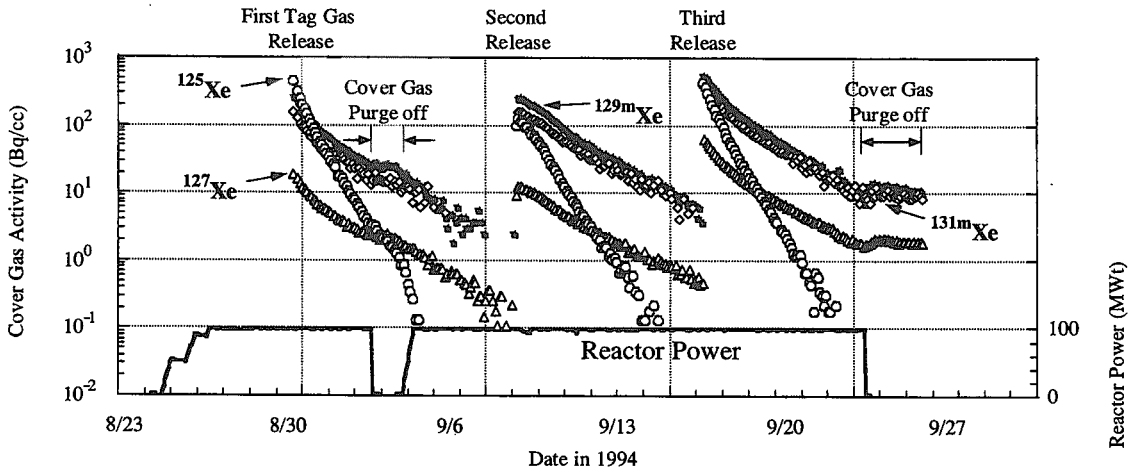

Fig. 4 Measured Tag Gas Activation Products in JOYO

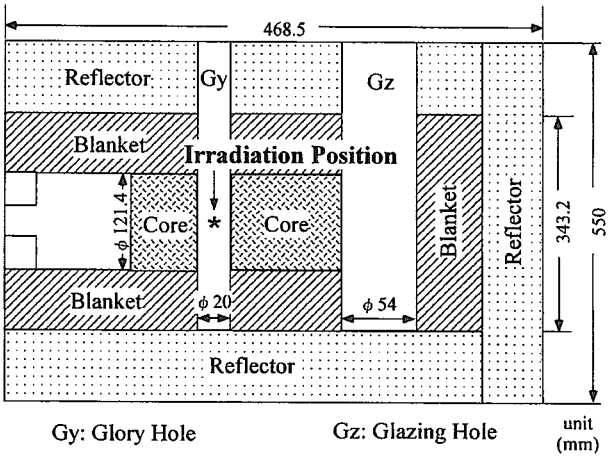

Fig. 5 Cross-sectional View of YAYOI and Tag Gas Irradiation Position

Table 2 YAYOI Tag Gas Specifications

Tag ID	No. 1		No. 2		No. 3		No. 4	
Tag Gas	Xe	Kr	Xe	Kr	Xe	Kr	Xe	Kr
Volume (cc)	1.25	1.25	1.51	0.986	1.06	1.24	1.46	1.03
Xe Isotope Ratio								
124 / 129	0.079		0.059		0.074		0.074	
126 / 129	0.048		0.083		0.104		0.104	
128 / 129	0.185		0.249		0.296		0.296	
130 / 129	0.057		0.132		0.124		0.125	
131 / 129	0.060		0.682		0.637		0.646	
132 / 129	0.025		0.862		0.809		0.816	
134 / 129	0.006		0.329		0.312		0.312	
136 / 129	0.001		0.266		0.266		0.253	
Kr Isotope Ratio								
78 / 80		0.261		0.494		1.49		0.495
82 / 80		1.35		7.56		4.21		7.55
83 / 80		0.217		3.54		3.15		3.60
84 / 80		0.066		16.2		14.1		16.0
86 / 80		0.003		4.90		4.21		4.84

Irradiated at Glory Hole

Total Neutron Fluence = $0.61\sim1.23\times10^{16}$ n/cm^2

After the irradiation, tag gas capsules were taken from the YAYOI irradiation hole. Radioactivities of the tag gas activation nuclides were then determined by measuring the gamma-ray spectrum directly through the capsule using a high purity Ge detector.

III. Comparison with the Calculation

The measured radioactivities were compared with the calculated values using the ORIGEN2 code [5]. The flow chart for calculating radioactivity of tag gas activation nuclides is described in **Fig. 6**. The neutron cross sections of tag gas nuclides were based on the 70 group constants, JFS-3-J3.2 processed from the JENDL-3.2 library [6,7]. These cross sections were collapsed into one group using the neutron spectra of YAYOI and JOYO as shown in **Fig. 7**.

The 100-group neutron spectra for YAYOI and JOYO were determined by means of the foil activation method. The initial guess spectra were adjusted by the NEUPAC J1-Unfolding code package [8] using the measured dosimeter reaction rates and the JENDL-3 dosimetry file [9]. The initial guess spectrum for YAYOI was expressed by a linear combination of two fixed spectrum modes [9] which had been evaluated through the detector inter-comparison studies. The JOYO neutron spectrum was calculated with the DORT two-dimensional discrete ordinate transport code [10] using the JSD-J2/JFT-J2 shielding constant set [11] based on the JENDL-2 library [12,13].

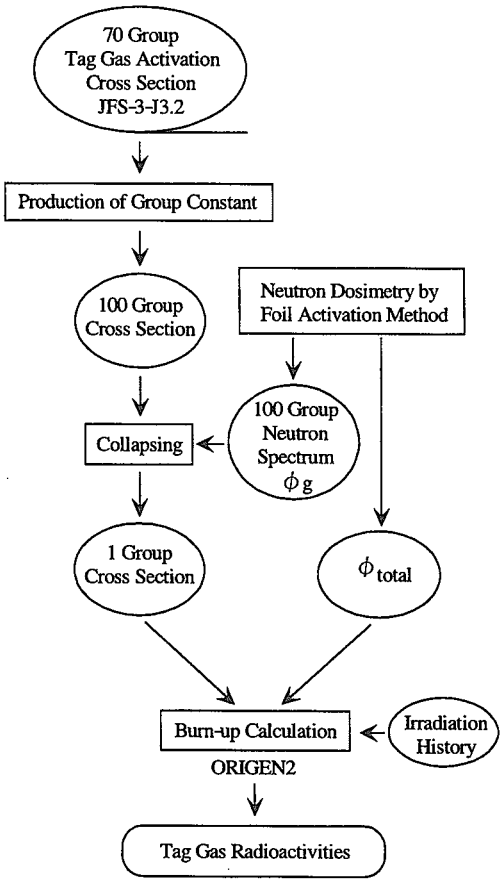

Fig. 6 Flow of Tag Gas Radioactivity Calculation

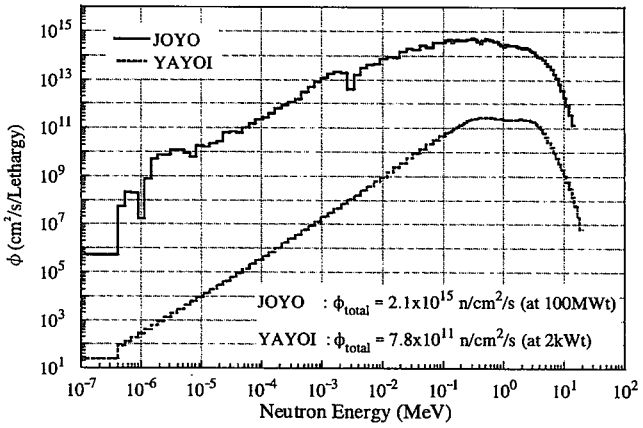

Fig. 7 Neutron Spectrum of JOYO and YAYOI

Comparing the YAYOI calibration tests measured radioactivities (E) and calculated values (C) based on the JENDL-3.2 yielded C/E values for ^{79}Kr, ^{125}Xe, ^{127}Xe, ^{133}Xe and ^{135}Xe that ranged from 0.86 to 2.6 as shown in **Fig. 8**.

Considering YAYOI's small total neutron fluences ($0.63^{\sim}1.27x10^{16}$ n/cm^2), the burn-up of tag gas nuclides and their activation products was negligible. Therefore, the discrepancy between calculation and measurement appears to be due to the uncertainty of these cross sections.

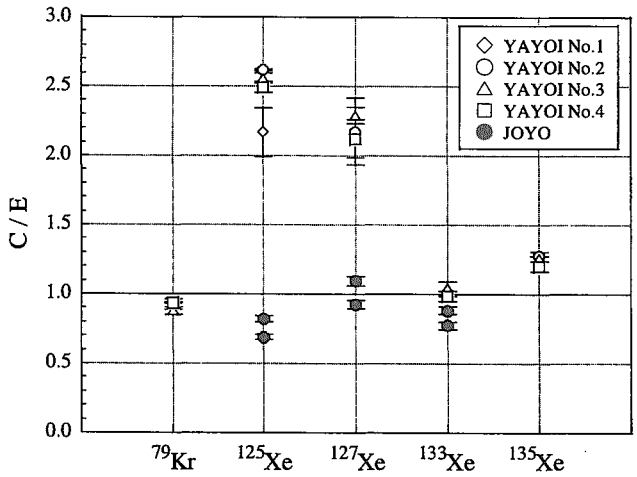

Fig. 8 Calculated to Experimantal Values of Tag Gas Radioactivities

The C/E values for the JOYO tests are similar to the YAYOI results except for ^{125}Xe and ^{127}Xe, which appears to be due to the sensitivity difference to the neutron energy of these nuclides. In **Fig. 9**, the tag gas nuclides have large resonance in the sub-keV energy region for the ^{124}Xe(n, γ) cross section. These results imply that further investigation of these cross section accuracies must be performed.

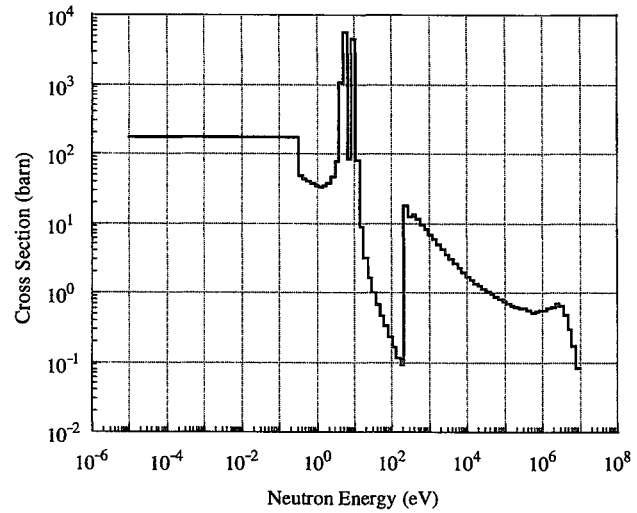

Fig. 9 Cross Section of ^{124}Xe(n,γ)^{125}Xe

IV. Conclusion

Activation cross sections of krypton and xenon were evaluated with irradiation tests in the fast neutron spectrum fields at JOYO and YAYOI. The ratio between calculated values and measured values ranged from about 0.86 ~ 2.6. These differences seem to be due to the activation cross -section error.

The tag gas activation method is applicable in sodium cooled fast reactor and the present accuracies of the tag gas activation cross sections were confirmed. The uncertainty of these cross sections requires further investigation.

Acknowledgements

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