VERIFICATION OF U-TARGET CLADDING DETERIORATION WITH DETECTION OF ^{135}Xe

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Abstract

In order to use a depleted uranium spallation target in safe, we introduced a system to check the deterioration of the target cladding by detecting ^{135}Xe, one of fission products. Xenon is sampled gas-chromatographically from the cover gas in the target cooling system and collected in the molecular-sieve trap. The presence of ^{135}Xe in the trap is identified with a high resolution germanium detector. It is found that this method is very effective in verifying the deterioration of the uranium cladding in the early stage.

Introduction

From the strong requirement to a high-flux pulsed neutron source, research facilities have promoted to use a depleted uranium as a spallation neutron production target[1,2]. In KEK, the uranium target has been used as a pulsed neutron source since 1985. The uranium target is covered with the alloy of zirconium (Zircaloy-2), the main purposes of which are to prevent corrosion of the uranium by the circulating water coolant and to avoid the radiogenic isotopes in the target leaking into the coolant. In order to use the uranium target in safe, it is very important to detect the occurrence of the partial and/or complete cladding failures on the surface of the uranium target as early as possible.

^{135}Xe is one of the possible fission products of uranium, of which the fission yield is not only high but also the specific activity is high. Xenon, being a gas, moves readily through very small openings in the cladding into the coolant. Therefore, if the cladding failure takes place, ^{135}Xe is considered to be the most suitable indicator to confirm the evidence of the failures among the possible radioisotopes leaking into the coolant. The gamma-ray spectrometric identification of ^{135}Xe (250 keV) in the coolant, however, is limited with the gamma-rays from other radioactivities produced by the spallation because some of them emit the gamma-rays with the energies close to 250 keV. This makes the detection of ^{135}Xe difficult.

In this paper, we describe a method which is applied to check the deterioration of the uranium target cladding in KEK. We constructed an apparatus with which xenon is separated from the cover gas of the target cooling system and collected in the molecular-sieve trap. The sample collected in the trap is analyzed with a high resolution gamma-ray spectrometer. With this system, we can verify the presence of ^{135}Xe without ambiguity.

Radioactivities in the coolant

In KEK, the uranium target is irradiated with proton beams of 500 MeV for about a week every two weeks. The target assembly is always cooled with water in a closed cooling system, and the water circulates in it through a heat exchanger and a tank filled with the cover gas of helium to remove decomposed gases in the water. Figure 1 shows the gamma-ray spectrum of radioisotopes in the cover gas obtained with a germanium detector. Principal radioisotopes in the cover gas are ^{76}Kr, ^{77}Kr, ^{79}Kr, ^{41}Ar and their daughters. Among the above products, the energies of gamma-rays from ^{76}Kr (252 keV), ^{77}Br (250 keV) are very close to the energy from ^{135}Xe, and ^{135}Xe is not distinguished from them energetically. Furthermore, their half-lives are of the same order of that of ^{135}Xe (9.2 h). A quantitative measurement of ^{135}Xe requires more elaborate analyses after they decayed out.

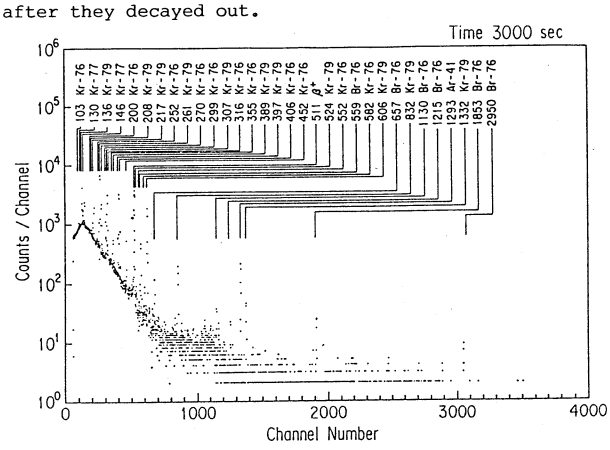

Fig.1 Typical gamma-ray spectrum for the cover gas.

Method

A block diagram of the system is schematically shown in Fig.2. First, about one litter of the cover gas is sampled in a sampling rubber bag from the target cooling system. Then, the gas thus sampled is injected automatically at the rate of 100 ml per one cycle to a gas-chromatograph in the sampling system. A typical chromatograph is shown in Fig.3 which was obtained for a helium-based standard gas. The flow path of a carrier gas from the gas-chromatograph to the xenon trap cooled at liquid nitrogen temperature opens only during the corresponding retention time, and bypasses the trap at the other time. This operation of the sampling system is controlled by the time sequence controller. As shown in Fig.4, one cycle starts on evacuation of the whole system and stops after the effluent of xenon is finished. The cycle is repeated ten times and xenon is collected in the trap. The xenon collected in the trap is analyzed with a germanium detector.

The efficiency of xenon recovery through the sampling system is found to be greater than 99 %. Since this measurement is limited with the sensitivity of the thermal conductivity detector (TCD) of the gas-chromatograph used, the efficiency is expected to be almost 100 %. In the routine measurement, xenon is also added to the sampled cover gas to avoid unexpected tracer effects.

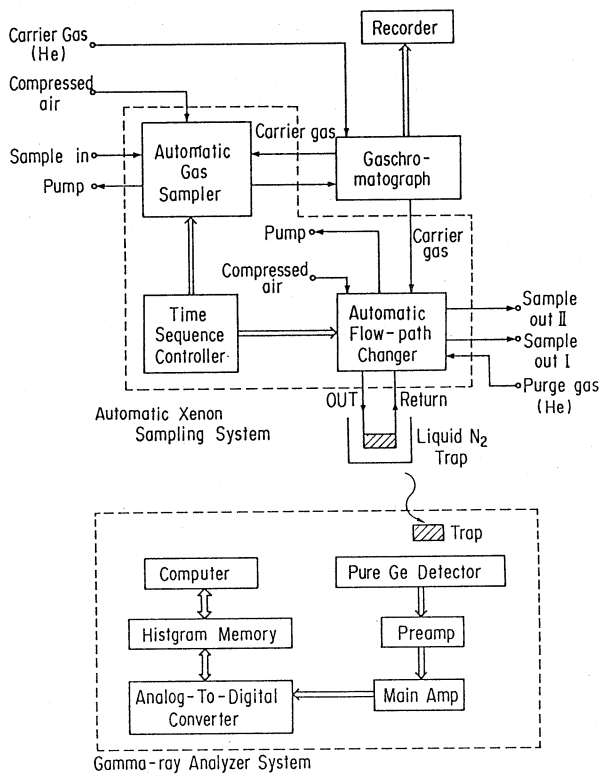

Fig.2 Schematic diagram of the xenon sampling system and the gamma-ray analyzer system.

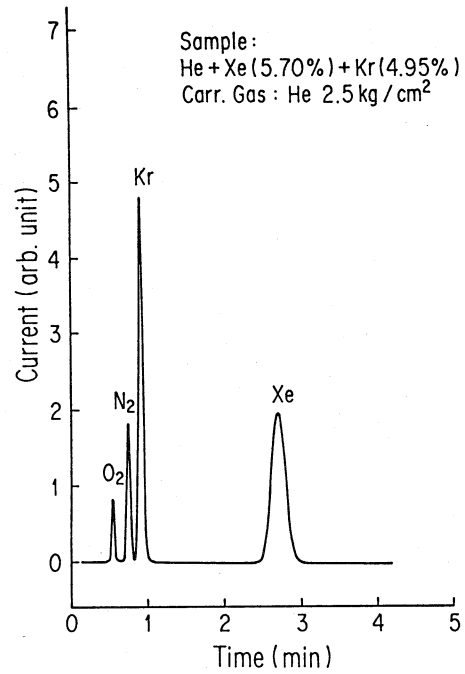

Fig.3 Typical chromatograph of the helium-based standard gas.

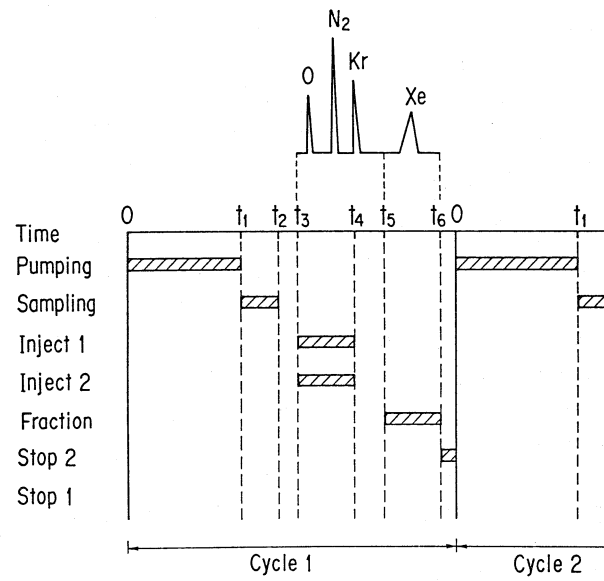

Fig.4 Time sequence of the automatic xenon sampling system.

Discussion

Figure 5 shows a typical gamma-ray spectrum of the cover gas collected in the xenon trap with the above method. Peaks found in the spectrum were the same as those from the trap itself, namely due to the backgrounds. Obviously, the spallation products are completely eliminated and not found in the sample.

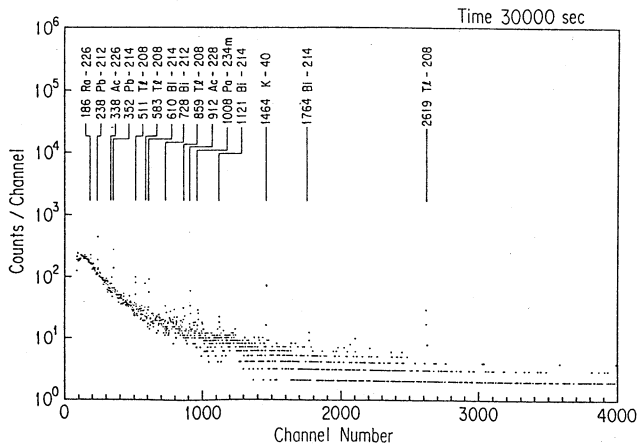

Fig.5 Typical gamma-ray spectrum for the xenon-enriched sample gas.

Background counts are about 10 cph/keV around 250 keV in our low-background germanium detector. The detection limit in this method is estimated to be less than 1 x 10^4 ^{135}Xe atoms per sample of one litter from the level of the backgrounds[3], assuming the total detection efficiency of the germanium detector 20 % and the efficiency of xenon recovery through the sampling system 100 %. So far no ^{135}Xe has been detected in the cover gas of the uranium target in KEK.

Reference

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2. F. Atchison, "A Theoretical Study of a Target Reflector and Moderator Assembly for SNS", RL-81-006 (1981).
3. M. Miyajima, H. Tawara and S. Sasaki, Proc. 4th Workshop on Radiation Detectors and Their Uses, p12, KEK Report 89-5 (1989).