The next figures (concentrations.) show the helium isotope ratios in function of time (Fig. 12). The helium originates from the uranium fission. It can be calculated the xenon isotope composition characteristic for the fission products, the leakages of fuel rods must be considered.

Continuous monitoring of the concentration and isotope ratios of those noble gases in the dissolved gas is a good tool for high sensitivity detection of small leakages of fuel rods.

The sampling and measuring method of noble gases from the primary water of Paks Nuclear Power Plant (Paks NPP) was developed by the Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI). The main components of the dissolved gas are hydrogen and nitrogen. There are a small volume of oxygen and carbon dioxide. In order to measure the noble gas isotopes the other gases have to be separated.

If the water flows down slowly the gas bubbles remain in the upper part of the ampoule, the water cannot drive them to the lower end, and the sample holders can be filled up with the gas. After an ampoule is filled up with gas, the automation drives the primary water to the next ampoule. Fifteen ampoules could be set in the automatic sampling unit.

At the end of the sample collection process the thin upper and lower tube endings of the ampoules are sealed off manually by flame sealing and changed with new sample holders. The gas samples in the sealed 20–25 cm3 ampoules are transferred to the laboratory. A special vacuum system has been developed in the laboratory of ATOMKI for opening the sealed ampoules (Fig. 2). The ampoules can be unwound with breaking down one of the ends. The gas comes through a filter and streams into a quadrupole mass spectrometer where the main components are measured.

The other part of the gas is closed into a steel finger where a special getter material adsorbs all the gas components excepting noble gases at the temperature of 580 °C. After this cleaning procedure a part of the noble gas sample is injected into a VG-5400 type noble gas mass spectrometer. The concentrations and isotope ratios are calculated from peak height measurements. The spectrometer is calibrated with air samples.

INTRODUCTION

During the nuclear fission and other nuclear processes several noble gases are produced in the fuel elements [1]. The monitoring of dissolved noble gases in the primary water can provide relevant information about the killer of heating rods and aid in additional information about some working parameters.

The helium concentrations and Xe/Kr ratios can be used to estimate the content of tritium and alpha emitting isotopes of the primary water.

During the admission of water large amounts of hydrogen and oxygen are formed in the primary circuit [2]. Oxygen originates from the primary water to avoid the corrosion and the formation of oxyhydrogen gas. The presence of argon indicates the air penetration and the required hydrazine amount for the oxygen absorption can be estimated with high accuracy.

The krypton and xenon isotopes, mostly 84Kr and 134,136Xe, are considerable parts of the fission products of 235U. These isotopes are produced inside of the fuel rods in the course of energy producing fission processes. If these noble gas isotopes appear in the primary water in significant concentration and with isotope compositions characteristic for the fission products, the leakages of fuel rods must be considered.

EXPERIMENTAL

Each primary circuit of individual reactor blocks has its own sampling tube. The tubes equipped with valves offer sampling possibilities from the circuits, which are under 40-60 bar pressure. The ATOMKI has developed an automatic sampling unit for collecting gas samples, which can be attached to the sampling systems by a capillary. The sample holders are set up in the sampling unit. These glass ampoules (Fig. 1) are filled in the middle for higher inter volume. The ends of these tubes are stoppering because the last step of a sample collecting procedure is stoppering up.

The sampling period is about eight hours during a reactor shut-down. Fifteen gas samples could be collected automatically into ampoules, which are sealed up without air-penetrating in the end of sampling period.

In the first step of the gas collection process the primary water flows up in the sample holder. The pressure of the water is reduced by the long capillary of the automatic sampling unit and controlled by the valve mounted before the sampling ampoule. The water-flow supplies the air from the ampoule. The next step is to reverse the direction of the flow. During this flowing procedure little gas-bubbles appear in the water because of the degassing effect induced by pressure reduction in the sampling unit.

RESULTS AND DISCUSSION

During the neutron-induced fission of 235U and the spontaneous fission of 239Pu, several noble gas isotopes are formed. The yields of xenon isotopes are shown in Table 1 [1]. It can be seen that the main fission products are the 134Xe and the 136Xe. The fission-product content of xenon in the primary water can be calculated by comparison of the measured and air-born isotope ratios and concentrations.

The next figures (Fig. 3-6) show the noble gas contents dissolved in the primary circuit of the three working reactor blocks on 19th March 2001.

It can be seen that the largest dissolved noble gas concentration is contained in the block-4. It can be also seen that there is xenon surplus in the block-3. The noble gas isotopes ratios (Fig. 7,8) (the black broken lines mean the values of the several noble gas isotopes ratios in the air) indicate that the xenon surplus originates from the uranium fission. It can be calculated the xenon derivatives from uranium and air (Table 2). The helium originates from the uranium fission. It can be seen that the argon content, similarly with xenon and krypton content, increases during the shut-down period (Fig. 11). It means there is a certain air penetration, because the isotope ratios of xenon (Fig. 12) are like in the air (the black broken or thick horizontal lines mean the values of the several noble gas isotope ratios in the air).

The Figure 14 shows the helium isotope ratios in function of time during the shut-down. It can be seen that the isotope ratios of helium (Fig. 14) are like in the air (the black broken or thick horizontal lines mean the values of the several noble gas isotope ratios in the air).

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