49

Dissolved stable noble gas measurements from primary water of Paks NPP

L. Palcsu[1], T. Pintér[2], M. Molnár[1], M. Mogyorósi[1], Zs. Szántó[1], É. Svingor[1] and I. Futó[1]

[1]Institute of Nuclear Research of the Hungarian Academy of Sciences (INR of the HAS)

[2]Paks Nuclear Power Plant, Hungary (Paks NPP)

S21

Abstract

Monitoring of dissolved noble gases in the primary water of a VVER type NPP can provide relevant information about the kilter of heating rods and detailed additional information about some working parameters. A sampling and measuring method of noble gases from the primary circuit was developed, ~~by the INR of the HAS~~.

The helium concentrations and $^3He/^4He$ ratios can be used to estimate the content of tritium and alpha emitting isotopes of the primary water. By argon content measurements the air penetration and the required hydrazine amount for the oxygen absorption can be estimated with high accuracy. Continuous monitoring of the concentration and isotope ratios of Xe and Kr in the dissolved gas is a good tool for high sensitivity detection of small leakage of fuel elements.

Unit 3.

In case of ~~block 3~~ xenon surplus was detected. Considering the isotope composition results of xenon about 76% of its total amount is uranium fission origin. These results indicate possible leakage of fuel rods.

Introduction

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

The helium concentrations and $^3He/^4He$ ratios can be used to estimate the content of tritium and alpha emitting isotopes of the primary water.

During the radiolysis of water large amounts of hydrogen and oxygen are formed in the primary circuit. Oxygen originates from the air as well. The oxygen has to be extracted from the primary water to avoid the corrosion and the formation of oxyhydrogen gas. The presence of argon indicates the air content of the primary water. By argon content measurements the air penetration and the required hydrazine amount for the oxygen absorption can be estimated with high accuracy.

The krypton and xenon isotopes, mostly $^{134,136}Xe$ and $^{84,86}Kr$, are considerable parts of the fission products of ^{235}U. 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 leakage of fuel rods must be considered. Continuous monitoring of the concentration and isotope ratios of these noble gases in the dissolved gas is a good tool for high sensitivity detection of small leakage of fuel rods.

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

Experimental

Each primary circuit of individual reactor blocks has its own sampling tube. The tubes equipped with valves offer sampling possibility from the circuits, which are under 40-bar pressure. The INR of the 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 flared in the middle for higher inner volume. The ends of these tubes are tapering because the last step of a sample collecting procedure is soldering up.

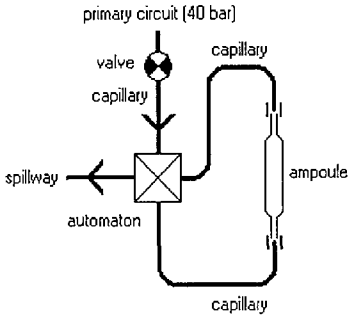


Figure 1.: Sampling unit for gas extraction from the primary circuit

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

In the first step of a 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 ampoules. The water-flow supplants 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. 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 holder 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 by flame sealing manually and changed with new sample holders. The gas samples in the soldered 20-25 cm^3 ampoules are transferred to the laboratory.

A special vacuum system has been developed in the laboratory of INR for opening the sealed ampoules (*Fig. 2.*). The ampoules can be unsealed 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 at the temperature of 500 °C absorbs all the gas components excluding noble gases. 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 by air samples.

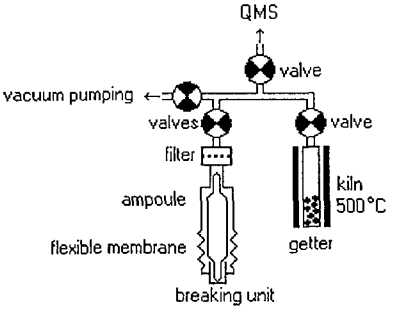


Figure 2.: Vacuum system for analysis of main component and for gas purification

Results and Discussion

During the neutron-indicated fission of ^{235}U and the spontaneous fission of ^{238}U several noble gas isotopes are formed. The yields of xenon isotopes are shown in *Table 1.* It can be seen that the main fission products are the ^{134}Xe and the ^{136}Xe. 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.

Parent	Isobaric yields (%)				
	^{129}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
^{238}U	<0.013	0.48	3.75	5.24	6.3
^{235}U	0.65	2.93	4.38	8.06	6.47

Table 1.: Production of xenon isotopes in fission

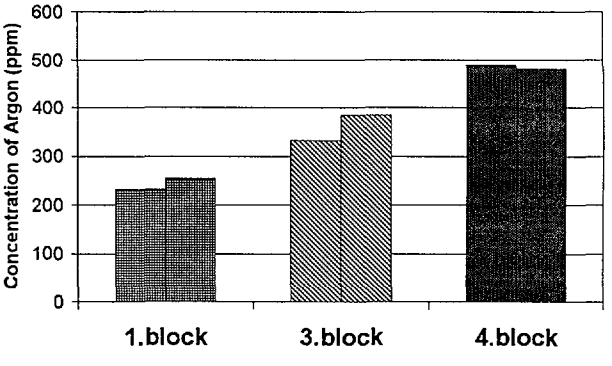


Figure 3a.: Argon in working blocks

The next figures (*Fig. 3a-d*) show the noble gas contents dissolved in the primary circuit of the three working reactor blocks on 15th March 2001.

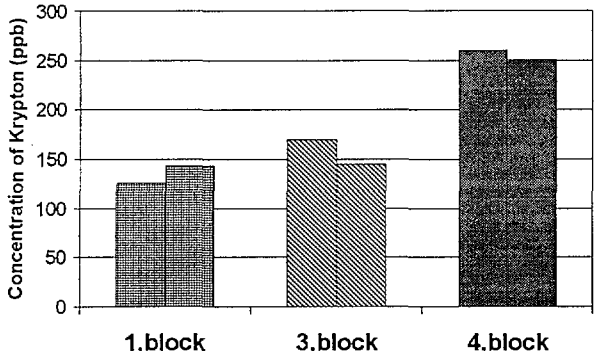


Figure 3b.: Krypton in working blocks

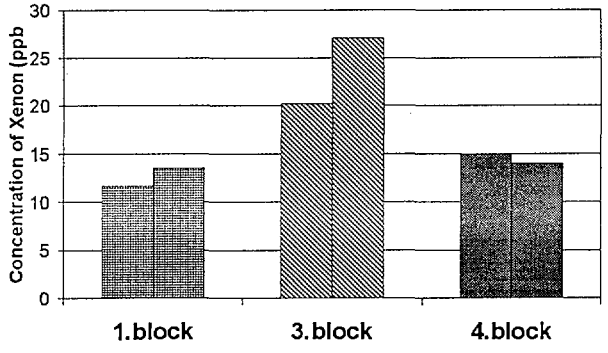


Figure 3c.: Xenon in working blocks

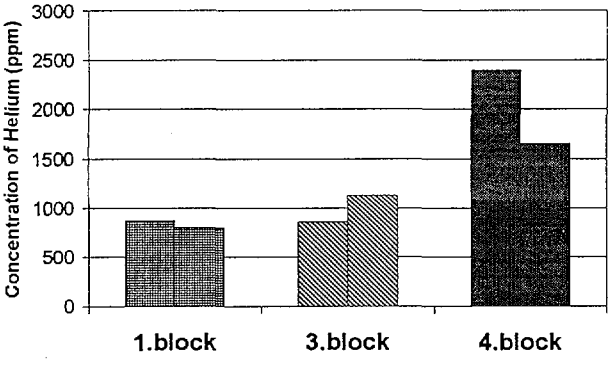


Figure 3d.: Helium in working blocks

It can be seen that largest dissolved noble gas is contained in the block-4. It can be also seen. that there is xenon surplus in the block-3. The noble gas isotope ratios (*Fig. 4a-e*) (the black broken lines mean the values of the several noble gas isotope 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 tritium by beta decay and isotopes emitting alpha particles. The production of alpha particles (^{4}He) is higher by an order of magnitude than the formation of daughter of tritium (^{3}He) (*Fig. 4f*).

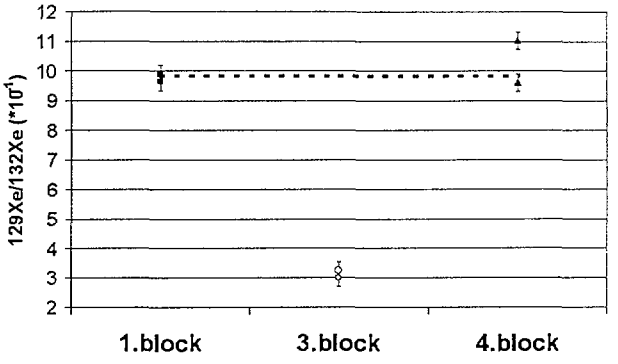


Figure 4a.: $^{129}Xe/^{132}Xe$ in working blocks

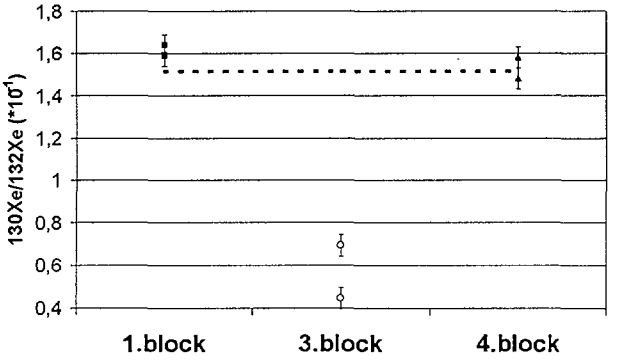


Figure 4b.: $^{130}Xe/^{132}Xe$ in working blocks

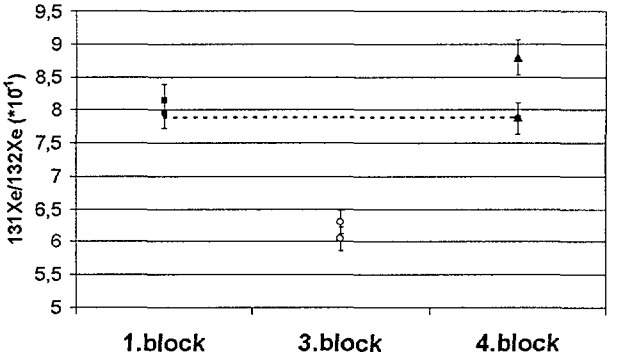


Figure 4c.: $^{131}Xe/^{132}Xe$ in working blocks

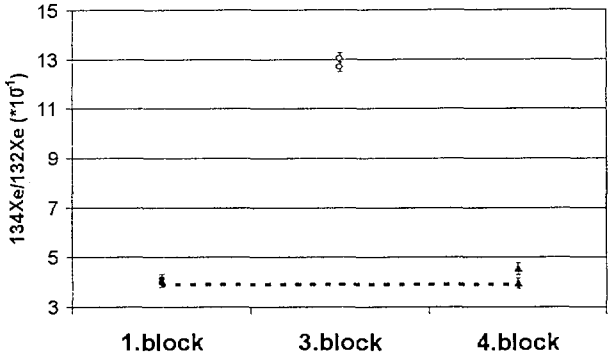


Figure 4d.: $^{134}Xe/^{132}Xe$ in working blocks

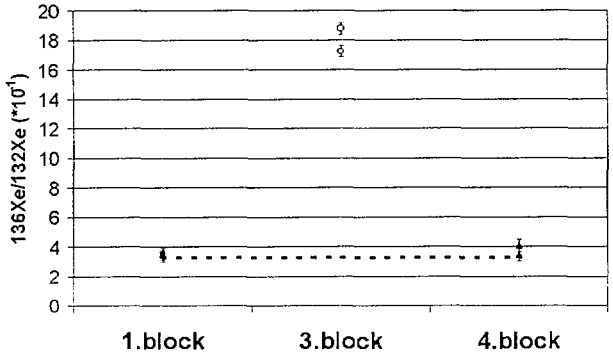


Figure 4e.: $^{136}Xe/^{132}Xe$ in working blocks

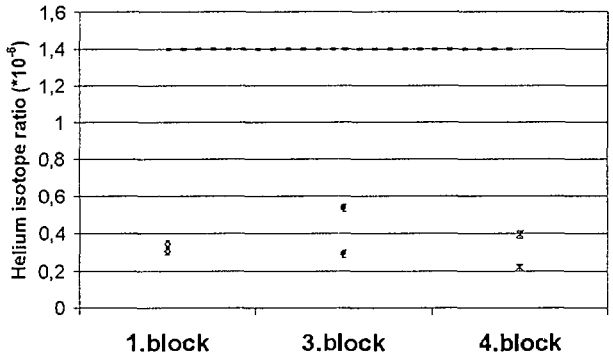


Figure 4f.: $^{3}He/^{4}He$ in working blocks

Xenon isotopes	Concentration (ppb)	
	Air origin	Uranium fission origin
^{129}Xe	1.44	-
^{130}Xe	0.27	-
^{131}Xe	1.13	1.69
^{132}Xe	1.44	3.05
^{134}Xe	0.56	5.27
^{136}Xe	0.47	7.60
Total	5.31	17.61
Proportion	23.17 %	76.83%

Table 2.: Ratio of xenon from air and uranium

On 17th March 2001 noble gas samples were taken from the block-2, which was shutting down. It can be seen that the argon content, similarly with xenon and krypton content, increase during the shut-down period (*Fig. 5*). It means there is air penetration, because the isotope ratios of xenon and krypton (*Fig. 6a-c*) 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).

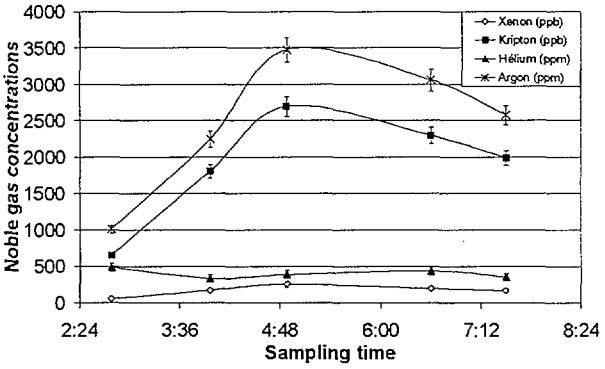


Figure 5.: Noble gas concentrations in the primary water during the shut-down

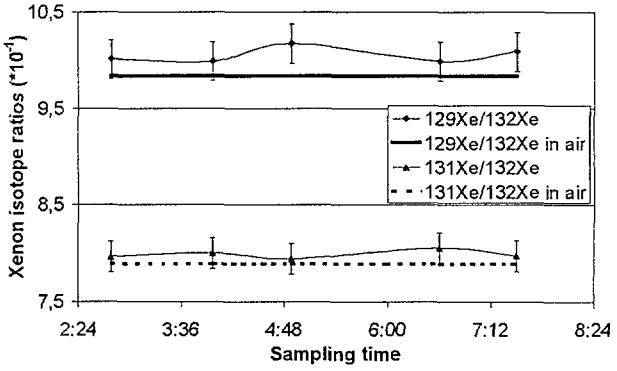


Figure 6a.: $^{129}Xe/^{132}Xe$ and $^{131}Xe/^{132}Xe$ isotope ratios in the primary water during the shut-down

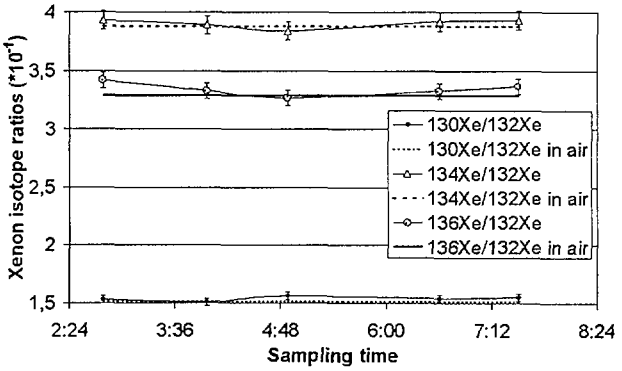


Figure 6b.: $^{130,134,136}Xe/^{132}Xe$ isotope ratios in the primary water during the shut-down

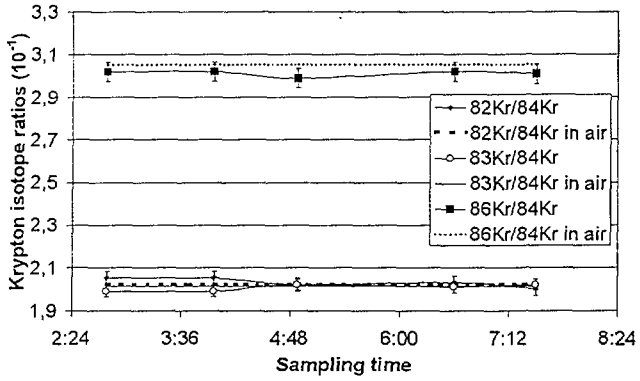
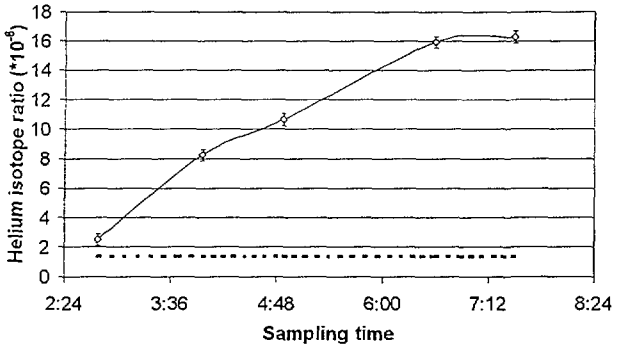


Figure 6c.: $^{82,83,86}Kr/^{84}Kr$ isotope ratios in the primary water during the shut-down

The *Figure 7* shows the helium isotope ratios in function of time during the shut-down. It seems that the isotopes emitting alpha particles are decreased, and the dominant process to formed helium isotopes is the decay of the tritium.



Figures 7.: $^{3}He/^{4}He$ isotope ratios in the primary water during the shut-down

Conclusion

In dissolved gases of primary water from working reactor blocks of Paks NPP the isotope ratio of helium always shows ^{4}He surplus, this effect indicates the dominance of alpha-fission in helium producing processes.

In case of block-3 xenon surplus was detected. Considering the isotope composition results of xenon about 76% of its total amount is uranium fission origin. These results indicate possible leakage of fuel rods.

During the shut-down of block2 the results do not show presence of surplus of typical noble gases of uranium fission processes. This period the production of ^{3}He is more dominant than ^{4}He producing.

Acknowledgement

We would like to thank to Ede Hertelendi, who died in a car accident in 1999.

References

1.Ozima M, Podosek F.A. *Noble gas geochemistry,* 1983