Analysis of Fission Gases Released in the Void Volume of Irradiated CANDU type Nuclear Fuel

Marin MINCU, Octavian UTA, Razvan MARINESCU

Institute for Nuclear Research Pitesti, Romania, Campului Str., Nr. 1, POB 78, 115400
marin.mincu@nuclear.ro

Abstract

The gaseous fission products tend, due to their nature and their insolubility, to produce changes in the fuel pellet volume and increase pressure in the void volume of irradiated fuel elements. Considering the impact that these changes can have in terms of nuclear safety, it is necessary to study the behavior of fission gas during irradiation. In order to study the production and release of fission gas during irradiation and post-irradiation experiments, various experiments can be designed. This paper presents the installation for cladding puncture and analysis of fission gases released into the void volume of irradiated fuel elements and recent experimental results obtained on CANDU type fuel elements. The concordance with the result provided by the computation codes will be also discussed.
The installation for cladding puncture and fission gas analysis was designed and manufactured at RATEN ICN Pitesti. It is used for:

- Measurement of the pressure and volume of gases in the void volume of the fuel rod;
- Measurement of the fuel rod internal void volume;
- Determination of the chemical composition of fission gases, including isotopic composition of the fission gases where applicable.

The paper contains also a description of the method used for the analysis of fission gases. A special attention is paid to the calibration method used for gas analysis by quadrupole mass spectrometry. A dedicated device was designed in order to mix pure gases in different concentrations for the calibration of the mass spectrometer.

Key words: fission gases, nuclear fuel.

1. Introduction

The behavior of the fission gases in irradiated fuel pellets plays a significant role for nuclear fuel performance. Fission gases are released from fuel pellets by a temperature-dependent process. At an early stage of the research in this field it was assumed that the release process is a diffusion process. In the meantime, it was discovered that burn-up and fission rate have large effects on the measured diffusion coefficient. A defect-trap model for fission gases release was developed to explain these measurements. According to this theory, the rate of escape of fission gases is inversely proportional to the number of defects in the material structure [1][2].
Structural changes occur during irradiation of ceramic UO_2, namely grain growth, swelling and creep [7]. The common cause of irradiation swelling of ceramic fuels is the formation of fission gas bubbles. The fission gas is formed as isolated atoms. Those diffuse and gather into small clusters, which in turn diffuse at a slower rate. The diffusion coefficient is dependent on the bubble radius.
Irradiation-induced swelling is a significant cause of the fuel cladding failure.

In order to study the production and release of fission gases, various experiments can be designed. This paper presents the installation for cladding puncture and analysis of fission gases released into the void volume of irradiated fuel elements and recent experimental results obtained on CANDU type fuel elements.

2. Description of the installation

The installation for puncture and fission gas measurement was designed to be able to measure:

- the pressure and volume of gas inside the fuel rod;
- the fuel rod internal void volume;
- the isotopic composition of the fission gas;
- the chemical composition of fission gas.

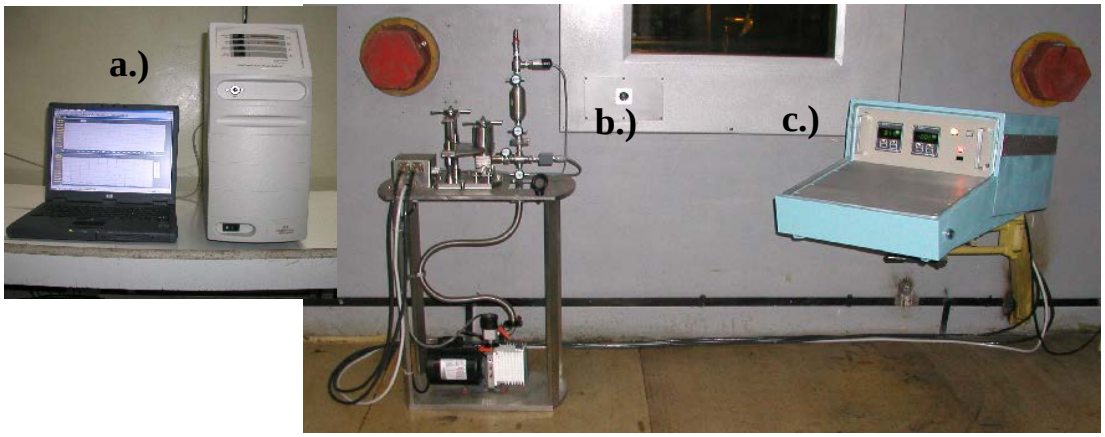

Fig. 1 The installation for puncture and fission gas measurement prior to be installed in the hot cell

As presented in figure 1, the installation consists of:

a. Quadrupole mass spectrometer SRS-QMS 200;
b. Puncture tool (installed in the hot cell);
c. Control panel (outside the hot cell).

In figure 2 is schematically presented how the installation for puncture and fission gas measurement was mounted in the hot cell.

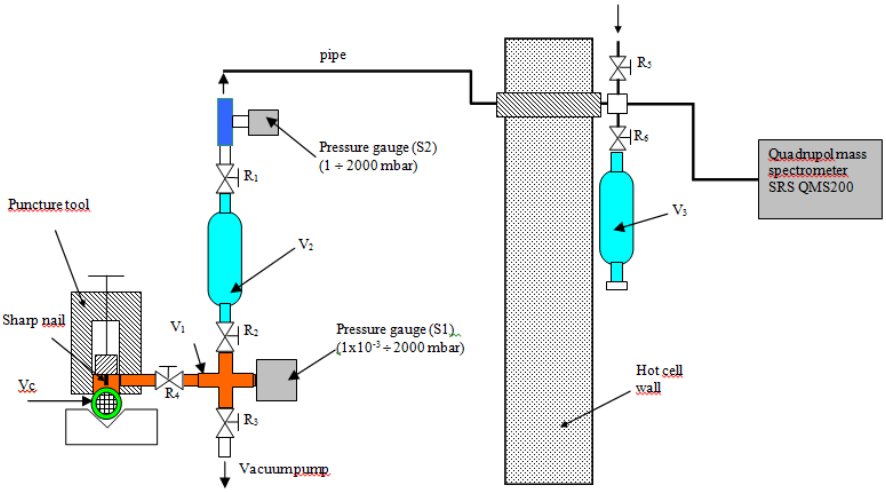

Fig. 2 *Simplified design of installation for puncture and fission gas measurement*

3. Method of operation

The fuel rod is placed into the puncture tool and vacuum-sealed by a rubber gasket.
The volume V_1 (see fig. 2 and 3) must be measured prior to puncture the fuel rod. The technique used is to pressure the standard volume V_2=148.5 cm^3 to a measured pressure P_2, to pump down the volume V_1 to a measured pressure P_1 and then to expand gases from V_2 into the unknown volume V_1. The equilibrium expansion pressure P is then measured. The volume V_1 is evaluated using the ideal gas law.
In practice, the unknown volume is pumped down to less than 0.03 mbar. In this case, the initial pressure P_1 will be approximated to zero.

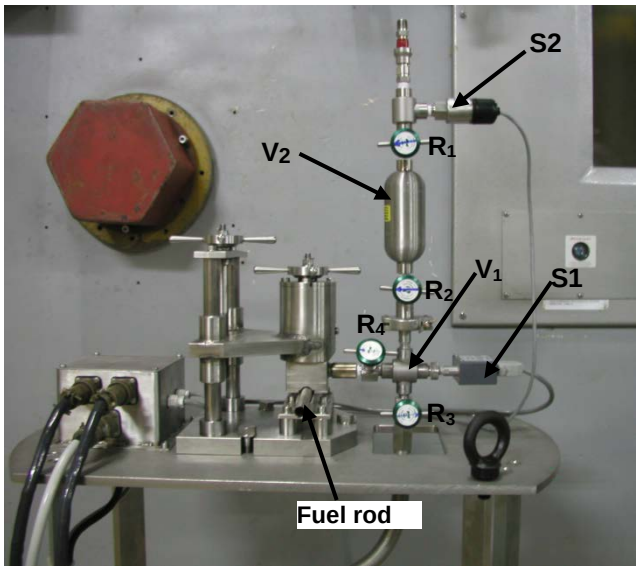

Fig. 3 Puncture tool
R_1, R_2, R_3, R_4,-manual valves
V_1 - Unknown volume
V_2 - Standard volume (delimited by R_1 and R_2)
S_1- Pressure transducer (10^{-3}÷2000 mbar)
S_2- Pressure transducer (1÷2000 mbar)
V_c – fuel rod internal void volume

In Table 1 is presented an evaluation of the unknown volume V_1. The pressure can be measured with a precision of ± 1 mbar. In order to increase the precision of unknown volume measurement, we need to repeat the measurement.

Table 1. Evaluation of the unknown volume V_1

P_1 [mbar]	P_2 [mbar]	P [mbar]	Unknown volume $V_1=V_2(P_2-P)/(P-P_1)$ [cm^3]
0,065	968	806	29,85
0,062	949	790	29,89
0,061	965	803	29,96
0,061	958	797	30,00
0,061	960	799	29,93
0,06	962	801	29,85
0,059	964	802	30,00
0,06	951	792	29,81
0,058	953	793	29,96
0,055	955	795	29,89
0,049	963	800	30,26
0,06	964	802	30,00
0,051	943	785	29,89
		Average volume:	29.94 ± 0.10

It is necessary to measure unknown volumes with a high precision because the internal void volume of a CANDU fuel rod is less than 2 cm^3 and a standard deviation of 0.1 cm^3 lead to a relative error greater than 5%.

Cladding puncturing

In order to measure the pressure of the gas in the void volume of the fuel rod we have to puncture the cladding and to expand the gases in the volume V_1 that was measured before. Prior to puncturing, the system is pumped down to less than 0.03 mbar. The fuel rod is punctured mechanically by turning the screw on the plunger drive. The fission gas will then expand into the volume V_1. The final pressure P_4 is measured and recorded. The fission gas is expanded a second time into the volume V_2. The equilibrium pressure P_5 is then measured and recorded. At this step, a significant fraction of gas from the internal void volume of the fuel rod is captured in the volume V_2.
For the case presented in this paper, it was measured P_4= 64 mbar.
The fission gas collected in volume V_2 is brought to the QMS 200 mass spectrometer by means of a tube passing through the hot cell wall.

4. Fission gas analysis by QMS 200 mass spectrometer

SRS QMS 200 is a quadrupole mass spectrometer that operates under high vacuum. The analog scanning mode was used for measurements. This mode allows the detection of fractional masses and provides the direct view of the peak shapes and of the resolution of the instrument.
It can be assumed that the total spectrum is a linear combination of the spectra of different species that are present in the mixture. In mathematical terms, the assumption stated above can be written as the following linear equation:

$$H_M = \sum_g h_{Mg}$$

where: g is an integer variable that indexes the gases present
M is an integer variable that represents the mass numbers
H_M= total peak height of the spectrum at mass number M
h_{Mg}= peak height contribution from gas g at mass M

h_{Mg} is related to the fragmentation pattern, sensitivity and partial pressure of gas g by the equation:

$$h_{Mg} = \alpha_{Mg} S_g P_g$$

where: α_{Mg}= fragmentation factor of gas g at mass M
$S_{g=}$ partial pressure sensitivity of gas g
P_g= partial pressure of gas g in the system.

The above equations are combined to obtain the system of equations:

$$H_M = \sum_g \alpha_{Mg} S_g P_g$$

The software of the spectrometer uses a multiple linear regression to calculate the partial pressures P_g. Obviously, accurate results can only be obtained if the constants α_{Mg} and S_g are well known for the mass spectrometer.
A dedicated device was designed and manufactured at ICN Pitesti in order to determine the factors α_{Mg} and S_g for the SRS QMS 200 mass spectrometer, as presented in figure 4. This device is able to mix gases in known proportions [2] [4]. The sensitivity factors S_g were determined by measuring mixed gases in proportions similar with that obtained from irradiated fuel rods.
Fragmentation factors were determined by analyzing pure gases He, Xe and Kr.

Fig. 4 Device for gases mixing and SRS QMS 200 mass spectrometer calibration

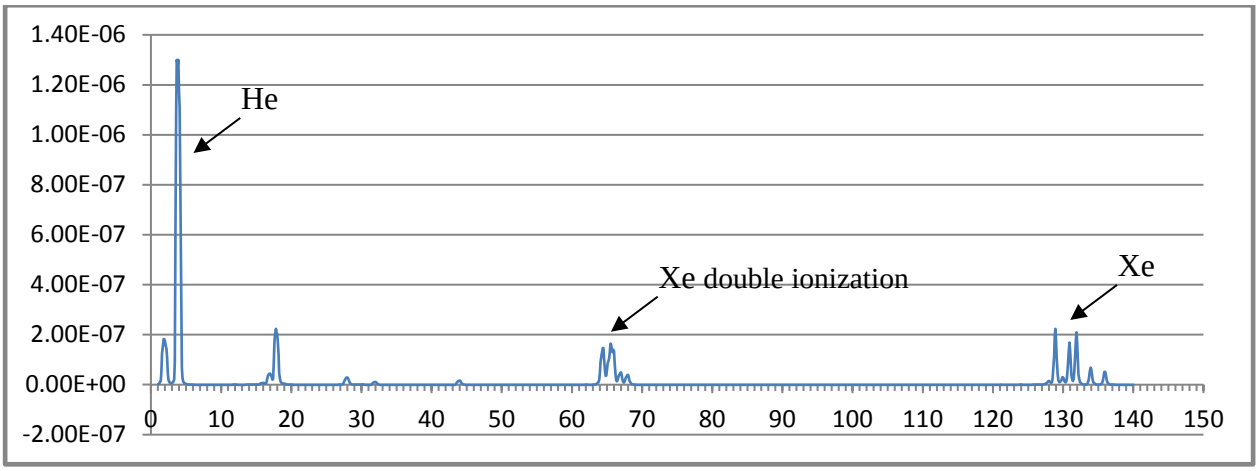

Fig 5. *Spectrum of a He-Xe mixture (He/Xe=0,12 partial pressures ratio)*

In figure 5 is presented a spectrum for a He-Xe mixture. The ratio of partial pressures He/Xe was measured using the pressure gauges mounted on the calibration device.

The fission gases collected in the volume V_2 (fig. 3) were analyzed by mass spectrometry using the analog scanning mode. A spectrum obtained for a CANDU fuel rod is presented in figure 6. It can be seen that a very small quantity of fission gases was released in the void volume of the fuel rod.

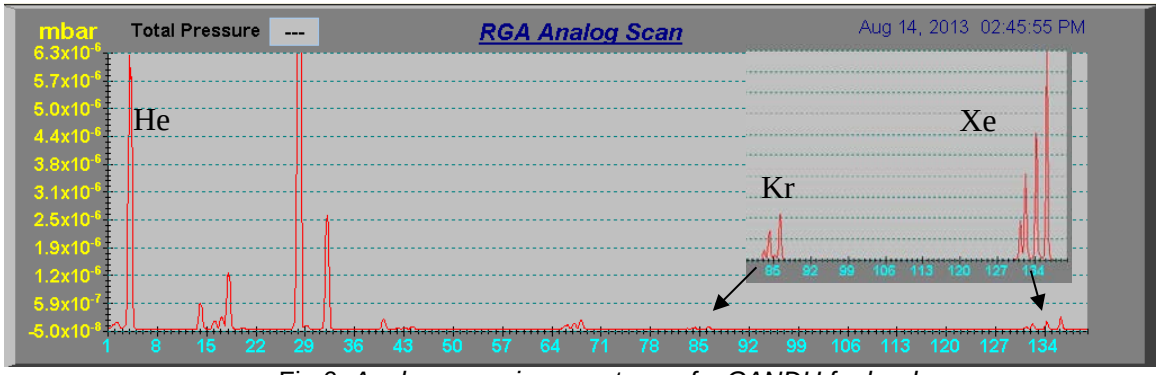

Fig 6. *Analog scanning spectrum of a CANDU fuel rod*

In Tables 2 and 3 and Figures 7. and 8. are presented detailed mass spectra of Kr and Xe. Isotopic compositions of Kr and Xe were calculated using the following relation:

$$C_k(\%) = \frac{P_k}{\sum_i P_i} x100;$$

where: P_k – partial pressure of isotope k;
P_i – partial pressure of isotope i;
C_k – concentration of isotope k;

Table 2. Isotopic composition of Kr

Isotope	Partial pressure [mbar]	Concentration [%]
^{83}Kr	3,24E-08	11,21
^{84}Kr	9,44E-08	32,68
^{85}Kr	1,71E-08	5,92
^{86}Kr	1,45E-07	50,19

Table 3. Isotopic composition of Xe

Isotope	Partial pressure [mbar]	Concentration [%]
^{131}Xe	1,37E-07	9,64
^{132}Xe	2,77E-07	19,49
^{134}Xe	3,89E-07	27,38
^{136}Xe	6,18E-07	43,49

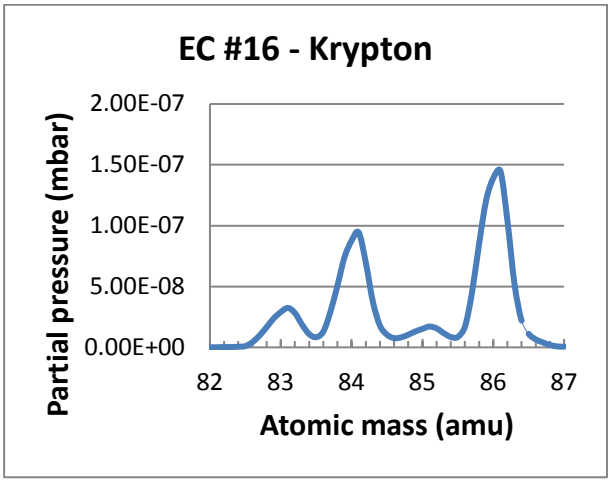

Figure 7. *Isotopic distribution of Krypton*

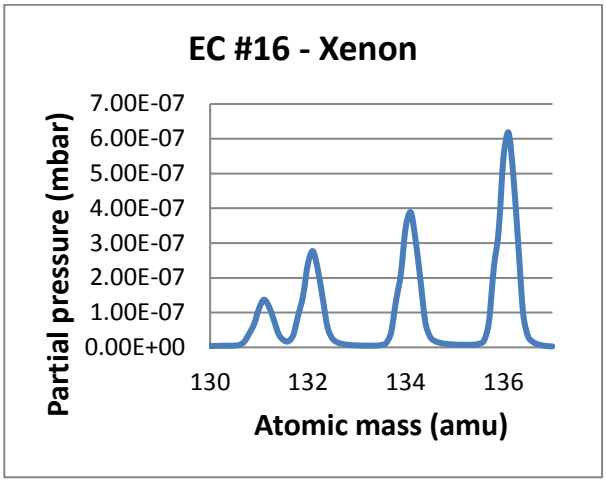

Figure 8. *Isotopic distribution of Xenon*

The measured isotopic compositions of Kr and Xe are in good agreement with those obtained using the computing code ORIGEN as is presented in Figures 9 and 10.

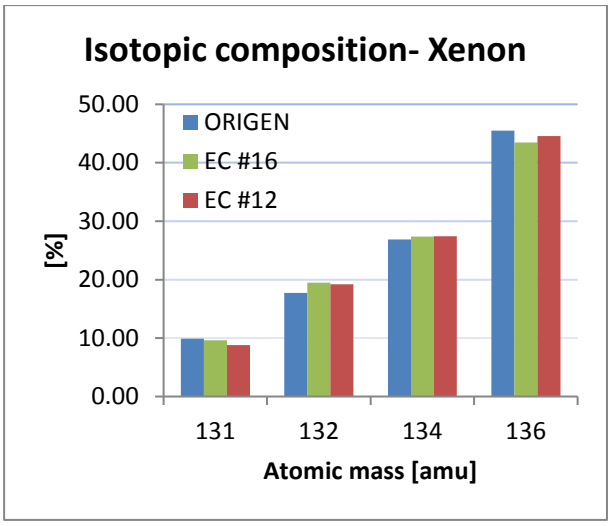

Figure 7. *Isotopic composition of Krypton*

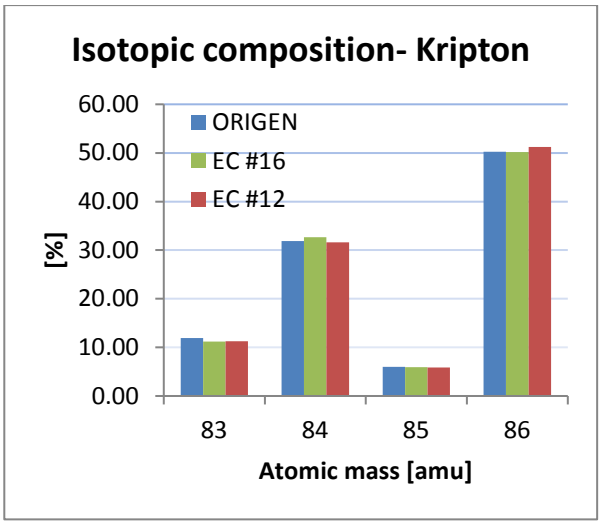

Figure 8. *Isotopic composition of Xenon*

In order to determine the void volume, we have to measure before the volume $V_3=V_1+V_c$ which is obtained after puncturing the fuel rod. The volume $V_3=V_1+V_c= 31.91 \pm 0,13$ cm^3 (see figure. 2) was measured in the same manner as V_1 volume.
The fuel rod internal void volume V_c was determined by subtracting the average volume V_1 out of the average volume V_3.

$$V_c= V_3 – V_1 = 31.91- 29.94 = 1.97 \text{ cm}^3;$$

As described at cladding puncture, the gases from the void volume of the fuel rod are expanded in the volume V_1 that was measured before. Using the laws of ideal gases we can calculate:

$$P_c=P_4V_3/V_c = 64*31.91 / 1,97 = 1036.7 \text{ mbar}$$

Where: P_4 – the pressure after the first expansion.

In order to compare results obtained from different experiments we have to calculate the gas volume under standard conditions of temperature and pressure. It was obtained:

$$V = 0,269\ P_cV_c/T = 0.269*1036.7*1.97 / 300.15 = 1.83 \text{ cm}^3$$

where: P_c – measured internal pressure [mbar];
V_c – measured internal void volume [cm^3];
T - temperature in the hot cell during the measurement [K] ;

5. Conclusions

After vacuum tight test it was found a leakage level of $2.0\text{x}10^{-7}$ std cm^3/sec which assure a good operation for pressures down to $5\text{x}10^{-3}$ mbar that can be obtained by a mechanical vacuum pump.
It is necessary to measure unknown volumes with a high precision because the internal void volume of a CANDU fuel rod is less than 2 cm^3 and a standard deviation of 0.1 cm^3 lead to a relative error greater than 5%.
As far as the pressure measurements are concerned, precise pressure gauges were used for measurements (± 1 mbar).

The measured isotopic compositions of Kr and Xe are in good agreement with those obtained using the computing code ORIGEN.
The calibration of the mass spectrometer has to be made in conditions as close as possible to those used for fission gas measurement. The mass spectrometer is sensitive to inlet pressure.
For the case of CANDU fuel rods irradiated under normal conditions (i.e. without any irradiation incident), a very small quantity of fission gases are released from fuel pellets in the void volume of the element [5].

References

[1] L.O. Jernkvista, A.R. Massiha, and J. In de Betouc - Evaluation of fission product gas release and the impact of fuel microstructure at high burnup
[2] R.M. Carrall – "Fission-Product Release from Fuels", Nuclear Safety, vol. 7, no.1
[3] G.J. Small – Transient Gas Release from Irradiated UO2: Specimen Characterization and Experimental Methods, September 1987
[4] D. Parrat –"Fission Gas Release Studies by means of Fuel Thermal Analyses in Analytical Devices" , IAEA Regional project RER/9/076
[5] J.A.Turnbull, P. Menut, E. Sartori – A REVIEW OF FISSION GAS RELEASE DATA WITHIN THE NEA/IAEA IFPE DATABASE
[6] D.J. Clough, S.J. Gouldstone, R.H. Keep, G.J. Small, A Technique for Determining the Fission Gas Content of Small Samples of Irradiated Reactor Fuel, Harwell Laboratory, July 1986
[7] J.C. Killeen – "Fission Gas Release During Post Irradiation Annealing of Large Grain Size Fuel from Hinkley Point B", Nuclear Electric plc, Barnwood, United Kingdom