MEASUREMENT OF FISSION GAS RELEASE FROM PHWR FUEL PINS

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Introduction

Approximately 15% of the fission product inventory in irradiated nuclear fuel, comprises of noble gases Xenon (Xe) and Krypton (Kr), in different isotopic states. These gases, if released to the fuel-clad gap and/or to the fuel element plenum, reduce the thermal conductivity of the cover gas and retard the heat removal from the fuel. Consequently the fuel temperature increases, releasing more gas to the plenum. Moreover, high thermal gradient in the fuel during power generation and the thermal shocks experienced by the fuel elements due to the movement of control rods, produce cracks in the normally brittle ceramic fuels. These cracks also act as fast conduits for the release of fission gases to the fuel-clad gap.

The measure of fission gases released in the fuel element testifies to the performance of the fuel. In general, fuels are designed with sufficient porosity to enable them to retain most of the generated gases within the matrix. During a postulated Loss Of Coolant Accident (LOCA), depressurization of the coolant occurs. When the coolant pressure drops to a value less than the fission gas pressure of the fuel pin, the clad creeps out, resulting in the ballooning of the clad. Coolant flow in the channel further reduces, thereby increasing the temperature of the fuel. Thus, knowledge of internal pressure of the fuel pin, is also a safety requirement. Xe and Kr have different isotope ratios when produced from fission of uranium or plutonium. Therefore, the isotope composition of the fission gases provides additional information on their source of production.

This article describes the fission gas measurement setup (installed at the PIED hot cells) and the experimental details and also presents the results obtained from the tests carried out on fuel pins from 19-element UO2 PHWR fuel bundles irradiated to different burn ups from 2,500 MWd/tU to 15,000 MWd/tU in various reactors. Results from one ThO2 fuel bundle irradiated to nominal burn up of 11,000 MWd/tTh are also included for comparison. The pattern of fission gas release is compared with the data from Ontario Hydro 37-element fuel pins.

Description of the Apparatus

Puncture Chamber and Gas Measurement System

The schematic diagram of the fission gas measurement system is shown in Fig. 1 and the photograph of the actual system is shown in Fig. 2. The hot cell windows can be seen in the background.

The system consists of a remotely operated leak-tight puncture chamber of very low dead volume, placed inside a hot cell (shown in Fig. 3 (a)). The salient feature of the chamber is that, it is adaptable to fuel pins of MOX and PHWR designs. The chamber is connected to the collection system located in the operating area by a 2mm bore stainless steel tube, passing through 1200 mm thick shielding wall of the hot cells, as shown in Fig. 3 (b). The fission gas collection system with a substantially low dead volume has been fabricated, with all components made of stainless steel. The low dead volume of the system, enhances the sensitivity of volume and pressure measurements of released fission gases.
fission gases from PHWR fuel pins, which are of low void volume. Fission gas pressure measurements are carried out using a capacitance diaphragm gauge operated within the range of 0.1 Torr to 1100 Torr. The experimental details and the methodology used in the quantitative estimation of released fission gases are described elsewhere[1].

**Gas Chromatograph (GC)**

A dual column gas chromatograph, packed with molecular sieve in a 6 mm diameter stainless steel column of 3 m length was used, to estimate the composition of the released fission gas. Helium was used as the carrier gas. A thermal conductivity detector with tungsten-rhenium filaments was used for detection. Since the fission gases being injected to the GC from the measuring system are at sub-ambient pressure, pre-calibration of the number of molecules of the constituent gases against the peak areas was carried out before the test, by injecting Xe-Kr-He gas mixture with known composition and at known pressure.
A quadrupole mass spectrometer with a mass range of 1-300 amu was used, for estimating the isotope ratios of the fission gases Xe and Kr. The gases were ionized using a tungsten gas tight ion source and detected using Channeltron / Faraday detector. The gas from the measuring system is admitted to the analyzing chamber of the QMS through a heated non-discriminating gas inlet capillary system. A turbo molecular pump backed by a dry diaphragm pump provided a very low pressure, less than $10^{-5}$ mbar. Analysis was carried out in Multiple Ion Detection (MID) mode, with 0.5 second dwell time for each mass number. The isotope ratios of Xe and Kr were estimated from ionization currents of the respective isotopes. The mass calibration of the spectrometer was carried out, by admitting Xe, Kr and He gas mixture of known chemical and natural abundance isotope ratios.

**Fuel Pins from PHWR Fuel Bundle**

A typical 19-element PHWR fuel bundle is shown in Fig. 4. Puncture tests were carried out on dismantled fuel pins from the 19-element UO$_2$ fuel bundles, with the average burn up in the range of 2,500 -15,000 MWD/tU received from different reactors. Fuel pins were selected from the outer, intermediate and the central regions of the bundles.

**Results**

Fission gas release data from the fuel pins from two UO$_2$ fuel bundles which were discharged after an extended burn up of about 15,000 MWD/tU from Kakrapar Atomic Power Station (KAPS) are given in Table 1(a).

Location of the pin in the respective bundles, percentage fission gas release, ratio of Xe to Kr and the inside pressure inside the cladding are given in the Table. The burn up of the individual fuel pins is calculated from the power factor, obtained from the reactor operator.
Table 1(a): Fission gas release data from high burn up UO$_2$ fuel pins from PHWR

<table>
<thead>
<tr>
<th>Bundle average burn up, MWd/TU</th>
<th>Location of the pin</th>
<th>Pin burn up, MWd/TU*</th>
<th>Fission gas release, %</th>
<th>Xe / Kr ratio</th>
<th>Internal pressure at 300 K, MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.161 (KAPS-2)</td>
<td>Outer</td>
<td>16,070</td>
<td>16.9</td>
<td>12.2</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>18.9</td>
<td>12.1</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>16.2</td>
<td>12.2</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>13,645</td>
<td>1.0</td>
<td>8.8</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.0</td>
<td>10.3</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td>Center</td>
<td>12,890</td>
<td>0.1</td>
<td>9.5</td>
<td>0.3</td>
</tr>
</tbody>
</table>

* Power factors: 1.06 for outer pin, 0.9 for middle pin and 0.85 for outer pin

Table 1(b): Fission gas release data from low burn up UO$_2$ fuel pins from PHWR

<table>
<thead>
<tr>
<th>Bundle average burn up, MWd/TU</th>
<th>Location of the pin</th>
<th>Pin burn up, MWd/TU*</th>
<th>% fission gas release</th>
<th>Xe / Kr ratio</th>
<th>Internal pressure at 300 K, MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>7,671 (NAPS-1)</td>
<td>Outer</td>
<td>8,130</td>
<td>8.8</td>
<td>11.5</td>
<td>0.6</td>
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<td></td>
<td>7.8</td>
<td>10.7</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>6,990</td>
<td>0.1</td>
<td>9.5</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.2</td>
<td>8.5</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>Centre</td>
<td>6,520</td>
<td>0.1</td>
<td>10.1</td>
<td>0.1</td>
</tr>
<tr>
<td>4499 (KAPS-1)</td>
<td>Outer</td>
<td>4,670</td>
<td>0.2</td>
<td>10.5</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.2</td>
<td>9.0</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
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<td></td>
<td>0.2</td>
<td>8.4</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>Middle</td>
<td>4,410</td>
<td>0.1</td>
<td>7.5</td>
<td>0.2</td>
</tr>
<tr>
<td>2,600 (NAPS-2)</td>
<td>Outer</td>
<td>2,760</td>
<td>0.5</td>
<td>7.7</td>
<td>0.2</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>0.8</td>
<td>11.6</td>
<td>0.2</td>
</tr>
</tbody>
</table>

* Power factors: 1.06 for outer pin, 0.9 for middle pin and 0.85 for outer pin
Results obtained from normally discharged bundle and other low burn up bundles are given in Table 1(b). Fuel pins from the ThO$_2$ fuel bundle did not show any perceivable gas release. However, mass spectrometric analysis of the residual gas inside the fuel pins was carried out and a distinct difference in the isotope ratios of fission gases was observed between UO$_2$ and ThO$_2$ fuel pins.

Release data from all the tested fuel pins along with the data available from the pins from the 37-pin fuel bundles from the Ontario Hydro Generating Station [6], are plotted in Fig. 5. The locations of the fuel pins in the bundle are identified in the plot as shaded, partially shaded and open symbols, respectively for the outer, intermediate / middle and the inner / central fuel pins. Higher release for the outer fuel pins is clearly noticeable in all the fuel bundles. The release from the outer pins of the 19-element Indian PHWR fuel bundles, discharged after extended burn up of about 15,000 MWd/tU, is much higher than that observed from the outer elements of the 37-element fuel bundles of the Ontario Hydro Generating station.

The isotope ratios of released gases, from extended burn up of UO$_2$ fuel bundles and from one ThO$_2$ fuel bundle, irradiated to nominal burn up of 11,000 MWd/tTh are shown in Fig. 6. Published data on ratios available for U$^{233}$, U$^{235}$ and Pu$^{239}$, are also marked in the plot.

**Discussion**

Significantly high fission gas release was noticed in the outer fuel pins of all the fuel bundles, with corresponding increase in the internal pressure in the pin. For the fuel bundle discharged after extended burn up of about 15,000 MWd/tU, fission gas release varied from 16-22% for the outer pins to less than 1% for the central pin. The large variation in gas release is attributed to the higher linear heat rating and higher fuel temperature, experienced by the outer fuel pins with respect to the intermediate and the central pins. The observed fission gas release behaviour, corresponded well with the fuel restructuring revealed during metallography in the fuel cross section [2]. Microstructure in the outer pin showed grain growth region extending up to 50% of fuel radius. High temperature diffusion and grain boundary sweeping are believed to have caused high release in the outer pin [3]. Xenon to Kr ratio also shows a systematic variation from the outer to the central fuel pin, akin to the release pattern. This could be
attributed to the variation in the thermal histories of the fuel pins. The measured values of Xe/Kr ratios varied from 7.5 to 12.2, the maximum being for the peripheral pins. Xe to Kr ratio in the fission gases produced, is about 7 for the fission of U\textsuperscript{235} and about 19 for the fission of Pu\textsuperscript{239} \cite{4}. The release fraction of Kr from the fuel matrix is expected to be higher than that for Xe, by virtue of the higher diffusivity of the former. Therefore, the Xe/Kr ratio in the released gases is expected to be lower than the generation ratio.

In the case of UO\textsubscript{2} fuel, except for Xe\textsuperscript{136}/Xe\textsuperscript{134}, the isotope ratios as measured from QMS, also show a mixed trend of fissions from U\textsuperscript{235} and from Pu\textsuperscript{239}. While the ratios Kr\textsuperscript{84}/Kr\textsuperscript{86} and Xe\textsuperscript{132}/Xe\textsuperscript{134} match close to Pu\textsuperscript{239} fission, the ratios Kr\textsuperscript{82}/Kr\textsuperscript{86} and Xe\textsuperscript{131}/Xe\textsuperscript{134} are suggestive of U\textsuperscript{235} fission. The high value Xe\textsuperscript{136}/Xe\textsuperscript{134} is due to the formation of Xe\textsuperscript{136} from Xe\textsuperscript{135} by neutron absorption, which is not accounted for in the reported yield ratio of Xe\textsuperscript{136}/Xe\textsuperscript{134} \cite{5}. Limited results generated from the isotope measurement of fission gases from ThO\textsubscript{2} fuel show only the closeness of Xe\textsuperscript{131}/Xe\textsuperscript{134} to that reported for U\textsuperscript{233} fission.

Summary

The findings from the fission gas release measurements, carried out on PHWR fuel pins irradiated to different burn ups from 2,500 MWd/tU to 15,000 MWd/tU at different Indian reactors, can be summarized as follows:

1. At all burn ups, the outer elements released significantly more fission gas than the inner ones. For a normal discharge burn up of about 7,500 MWd/tU, the maximum release from the outer pin was 9% , resulting in an internal pressure of 0.6 MPa at room temperature.
2. At burn up of 15,000 MWd/tU, fission gas release from the outer pins was 16-22%, when the middle pin and the central pin released 2% and 1%, respectively. The internal pressure of the outer pins, corresponding to the burn up of 15,000 MWd/tU was in the range of 2.1-2.8 MPa.
3. Xenon to krypton ratio also varied with the location of the pin in the bundle, the maximum being for the outer pin.
4. The measured values of Xe/Kr ratio for natural UO\textsubscript{2} PHWR fuel were between the values reported for the thermal fission for U\textsuperscript{235} and that for Pu\textsuperscript{239}.
5. The isotope ratios of Xe and Kr also showed mixed trend of fissions from U\textsuperscript{235} and Pu\textsuperscript{239} for natural UO\textsubscript{2} fuel.
6. The isotope ratio of fission gas Xe corresponded with that for U\textsuperscript{233} fission, in case of ThO\textsubscript{2} fuel pins.

References