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Fission gas release behaviour in MOX fuels

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Abstract. As a part of plutonium recycling programme MOX $(U,Pu)O_2$ fuels will be used in Indian boiling water reactors(BWR) and pressurised heavy water reactors(PHWR). Based on successful test irradiation of MOX fuel in CIRUS reactor, 10 MOX fuel assemblies have been loaded in the BWR of Tarapur Atomic Power Station (TAPS). Some of these MOX fuel assemblies have successfully completed the initial target average burnup of ~16,000 MWD/T. Enhancing the burnup target of the MOX fuels and increasing loading of MOX fuels in TAPS core will depend on the feedback information generated from the measurement of released fission gases. Fission gas release behaviour has been studied in the experimental MOX fuel elements (UO_2 - 4% PuO_2) irradiated in pressurised water loop (PWL) of CIRUS. Eight (8) MOX fuel elements irradiated to an average burnup of ~16,000 MWD/T have been examined. Some of these fuel elements contained controlled porosity pellets and chamfered pellets. This paper presents the design details of the experimental set up for studying fission gas release behaviour including measurement of gas pressure, void volume and gas composition. The experimental data generated is compared with the prediction of fuel performance modeling codes of PROFESS and GAPCON THERMAL-3.

1. INTRODUCTION

Approximately 15% of the fission product inventory in irradiated nuclear fuel comprise of noble gases xenon (Xe) and krypton (Kr), in different isotopic states. The behaviour of fission gases plays a dominant role in the fuel performance. The fission gases, if retained in the matrix can cause fuel swelling. If the gases are released it decreases pellet clad gap conductance. The decrease in gap conductance increases the fuel temperature and thereby leading to increased gas release. Enhancing the burnup target of the MOX fuels and loading of more MOX fuels in TAPS core will depend on the feedback information generated from the measurement of released fission gases.

Fission gas release behaviour has been studied in the experimental (UO_2 - 4% PuO_2) MOX fuel element irradiated in Pressurised Water Loop (PWL) of CIRUS. The eight (8) MOX fuel elements examined were irradiated to burnup of ~16,000 MWD/T. These experimental MOX fuel elements contained fuel pellets with a number of design variables to study their effects in fission gas release.

2. MOX FUEL FABRICATION

Mixed oxide (MOX) fuel with UO_2-4% PuO_2 was fabricated for test irradiation in the standardised powder pellet route [1]. UO_2 powder after oxidation-reduction treatment was ball-milled with PuO_2 powder in small batches. For making controlled porosity pellets 0.5wt% of methyl cellulose was mixed with milled powder. MOX powder was pre-compacted at 5 tsi, granulated to 20# sieve size. The final compaction was done at 300 MPa pressure. Sintering was carried out in Ar+5% H_2 mixture at 1680–1700°C for 8 hrs. The sintered pellets were vacuum degassed at 400°C for 8 hrs. Resintering test was carried out at 1700°C for 24 hrs. The pellets of AC-2 cluster were not chamfered whereas all the fuel pellets of AC-3 cluster were chamfered. The Zircaloy-2 clad tubes were of 0.89 mm wall thickness with inside diameter varying from 12.637 to 12.713 mm. Clad tubes and other fuel pin components were out gassed at 220°C for 3hrs before pellet loading. Helium was used as the cover gas. Typical experimental MOX fuel pin is shown in Fig.1.

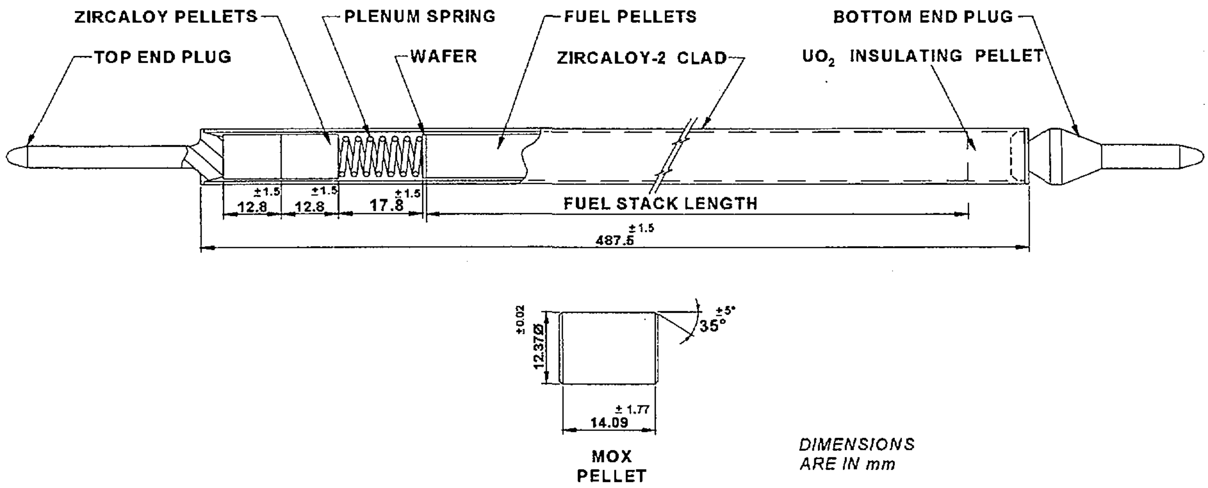

Fig. 1. A typical experimental mox fuel pin.

3. IRRADIATION DETAILS

Experimental MOX fuel pins were test irradiated in the pressurised water loop (PWL) of CIRUS reactor to a target burnup of ~16,000 MWD/T. The irradiation details of AC- 2 and AC-3 clusters are given in Table 1.

4. FISSION GAS RELEASE STUDIES

A new fission gas measuring set up was designed and fabricated to study the behaviour of fuel elements with low void volume and low fission gas releases.

4.1. Fission Gas Collection System Design

The puncture chamber and fission gas collection system was designed with low dead volume for precise determination of void volume of the fuel element. The puncture chamber, which can accommodate both MOX and PHWR type fuel pins, was designed with a very low internal volume. A contoured neoprene pad was provided at the fuel loading port of the chamber to ensure leak tightness. A tungsten carbide-tipped centre drill was used for puncturing the fuel pin. The puncture chamber was connected to the gas collection and analysis system by means of stainless steel tube through a hot cell service plug. The entire gas collection and analysis system was designed using 2 mm bore stainless steel tube to keep the overall system volume low. The vacuum system comprising of a rotary vane pump, capable of pumping down the system to a pressure of 1×10^{-1} Torr was used.

Table 1. Irradiation Details of MOX Fuel Clusters

Cluster ID	Coolant temperature, °C	Coolant pressure, psi	Neutron flux, $n/cm^2/sec$		Pin ID	Average burnup, MWD/Te	Relative power Generation	Heat flux, W/cm^2
			Fast flux	Thermal flux				
AC-2	204	1,500	$5*10^{12}$	$5*10^{13}$	TP-1	16,265	1.18	93
					TP-2		1.15	
					TP-3		1.15	
					TP-4		1.15	
					TP-5		1.18	
AC-3	204	1,500	$5*10^{12}$	$5*10^{13}$	TP-6	16,000	1	110
					TP-7			
					TP-8			

A capacitance diaphragm gauge, with a measuring range of 0.1 to 1100 mbar was used to measure the gas pressure. The system was fitted with two calibrated measuring flasks of volumes of 251 cc and 501 cc. A dual column gas chromatograph (GC) with thermal conductivity detector was used for the measurement of released fission gas composition. Helium was used as carrier gas. A quadrupole mass spectrometer (QMS) was used for the estimation of isotopic composition of the gases.

4.2. Experimental

The schematic diagram of the fission gas analysis system is shown in Fig. 2. The surface of the fuel pins was cleaned thoroughly with emery paper and mild organic cleaning aids to get a leak tight contact between Neoprene pad of the puncture chamber and the fuel pin.

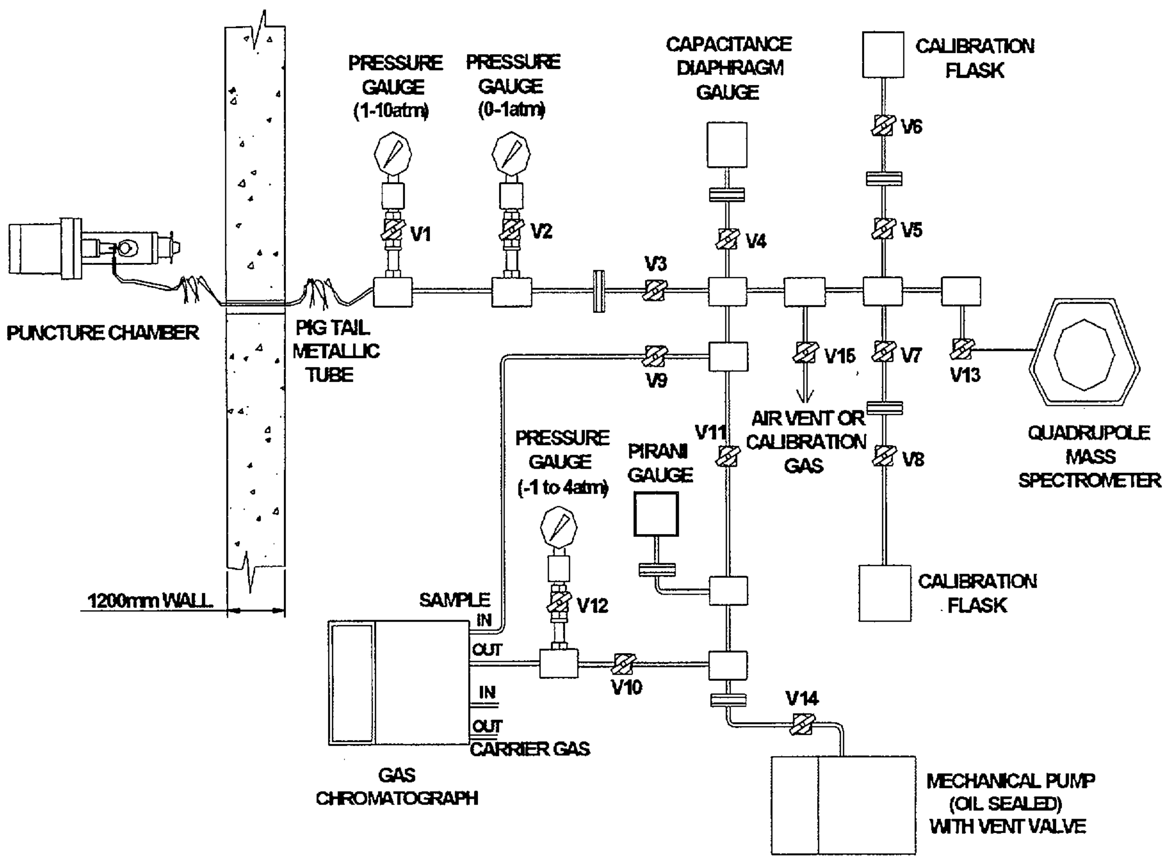

Fig. 2. Schematic diagram of fission gas collection & measuring system.

4.3. Estimation of System Volume

The fuel element was fitted to the puncture chamber and gently tightened against the neoprene pad. The system with the fuel element fixed in the puncture chamber was evacuated to a pressure of ~ 0.1 Torr. The exhaust of the rotary pump was fed to the hot-cell. All the shut-off valves except valves V10, V9 and V13 were kept open during this operation. Closing V5 and V7 valves the calibration flasks were isolated. The pressure (P_1) at which the flasks were isolated was noted from the capacitance diaphragm gauge. The system was isolated from the rotary pump by shutting off the valve V11. Valve V14 was not used for isolating the system from the pump since the same pump was used to evacuate the injection port of the GC. The system was filled with air to a measured pressure (P_2) and expanded to one of the calibration flasks (volume V_1) at pressure P_1. The resultant system pressure P_3 is noted. Enough time was allowed for the stabilisation of the pressure during above operations. The system volume (V_2) was calculated using the relation:

$$V_2=V_1*(P_1-P_3)/(P_3-P_2) \qquad \text{--------------------------}(1)$$

The experiment was repeated thrice to get concordant values.

4.4. Fission Gas Extraction

The fission gas analysis system including the calibration flasks was re evacuated to 0.1 Torr and isolated. The valve V11 was closed to isolate the system from the rotary pump. The drill on the puncture chamber was activated to puncture the fuel pin. The pressure reading on the capacitance diaphragm gauge was observed during the drilling operation. The puncturing of the fuel pin was indicated by the sudden surge in pressure on the gauge. On occasions when the gas pressure exceeded atmospheric pressure, one of the calibration flasks was connected to the system. The increase in pressure in the system due to the release of fission gas from the fuel pin, P_4, was noted. To minimise undesirable air leak to the system, the puncture chamber was isolated from the measuring system by closing the valve V3.

4.5. Gas Chromatograph

A 3-meter long dual column gas chromatograph packed with molecular sieve in ¼ inch OD stainless steel tube 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. The GC and the detector were switched on in advance for system stabilisation. Due to low volume and low pressure of fission gas, the conventional method of purging the GC loop with sample gas was not possible. Since the fission gas pressure on the collection system was sub-ambient, the GC loop had to be evacuated through the sample-loop outlet before admitting the gas through valve V10. The evacuation was ensured by the pressure reading on the gauge connected in the sample out-let path, ahead of valve V12. The valve V10 was closed after evacuation of the loop. The fission gas from the collection system was admitted into the injection loop by opening valve V9 of the sample inlet. The pressure of the gas to be injected to the GC was noted from the capacitance diaphragm gauge before injecting to the GC column. The peak areas were measured from the chromatogram.

4.6. Quadrupole Mass Spectrometer

A quadrupole mass spectrometer with a mass range of 1–300 amu was used for estimating the isotopic ratios of the fission gases. The gases were ionised using a gas tight tungsten ion source and detected using Channeltron/Faraday detector. By opening valve V13, the gas is admitted to the analyzing chamber of the QMS through a non-discriminating gas inlet capillary system. The isotopic ratios of xenon and krypton were estimated from the ionisation currents of the respective isotopes. The spectrometer was calibrated with xenon, krypton and helium gas mixture of known mass ratios.

4.7. Void Volume Estimation of the Punctured Fuel Element

With out disturbing the position of the fuel pin in the puncture chamber, the system volume was estimated as per the procedures described earlier in the section on estimation of system volume. The total system volume included the void volume of the punctured pin. The difference in the volumes in the two cases provides the void volume of the fuel pin.

This procedure of estimating the void volume was checked for repeatability and accuracy by puncturing a few calibration pins of the same geometry. The pins for calibrations had different void volumes and were filled with helium at different pressures. The pressure P_5 of gases inside the fuel pin at ambient temperature is calculated by:

$P_5 = (P_4 - P_0)*$ (System volume+ Void volume)/Void volume -------------(2)

where P_0= System pressure before puncturing the fuel pin and
P_4= System pressure after puncturing the fuel pin.

4.8. Methodology used in Processing Gas Release Data

The quantity of fission gases Xe and Kr in the injected volume was estimated using the gas chromatograph output. The quantity of He in the chromatograph-loop was estimated by using ideal gas laws. The corrections to the system leaks were applied by accounting for oxygen and nitrogen peaks, observed in the chromatogram. The methodology of calculation followed for estimating the percentage of fission gas release is given in the Annex.

5. RESULTS

The summary of the results is given in Table 2. Total fission gas produced, percentage of fission gas released and the ratio of released xenon to krypton for each fuel pin are shown in the table. The average fuel pellet density of each pin and change in the void volume of each pin after irradiation are also given in the table.

5.1. Fission Gas Release Code Prediction

Two fuel-modeling codes are in active use at our research centre. The fuel modeling code GAPCON THERMAL-3 [2] is used by Reactor Engineering Division for fuel design and code PROFESS [3] is used by Post Irradiation Examination Division for analysis and interpretation of PIE data. Blind prediction of fission gas release by both the codes based on fuel design and irradiation parameters are given in Table 3 along with experimentally measured values for 5 MOX fuel elements of AC-2 cluster. As can be seen from the predicted values, both the codes over-predicted fission gas release. The codes also over- predict the fuel centre temperature. Since fuel temperature has a strong effect on fission gas release over prediction by the codes is explainable.

6. DISCUSSION

Fission gas release behaviour of standard enriched uranium fuels of Tarapur Atomic Power Station had been studied on 14 fuel elements in the burn up range of 3,200 to 24,000 MWD/Te [4]. The fission gas release ranged from 0.9% to 13.6%. Only one fuel element had fission gas release less than 1%. Nine fuel elements have release fraction ranging from 1 to 5% and the remaining 4 fuel elements had fission gas release in the range of 9 to 13.6%.

Table 2. Released Fission Gas Measured from Experimental MOX Fuel Pins

Cluster ID, burnup& heat flux	Pin No.	Average pellet density, % TD	Post irradiation change in void volume, %	Fission gas produced, cm^3 at STP	Fission gas released, cm^3 at STP	Internal gas pressure, Atmospheres (G)	Xenon to krypton ratio	Fission gas released, %
AC2 16,265 MWD/Te 93 W/cm^2	TU8	94.56	(-) 3.1	47.03	0	0.3	0	0
	TP1	96.27	(-) 20.12	296.18	0.36	0.61	14.44	0.12
	TP2	96.34	(-) 19.53	287.39	0.29	0.56	13.90	0.10
	TP3	94.32	(-) 18.33	282.57	0	0.65	0	0
	TP4	96.26	(-) 19.35	288.16	0.45	0.60	16.38	0.16
	TP5	96.20	(-) 12.47	296.08	1.09	0.58	12.70	0.37
AC3 16,000 MWD/Te 110 W/cm^2	TP6	95.92	(-) 12.1	245.79	20.59	7.37	13.92	8.38
	TP7	95.57	(-) 10.2	245.75	25.88	8.30	13.98	10.53
	TP8	95.81	(-) 17.0	285.84	22.55	7.97	13.93	9.17

(-) Negative sign indicates decrease in void volume

Table 3. Comparispon between Predicted and Released Fission Gas for AC-2

Pin No.	Post irradiation examination		PROFESS FG Model-3		GAPCON-THERMAL-3	
	Estimated centre temperature, °C	Fission gas release, %	Centre temperature, °C	Fission gas release, %	Centre temperature, °C	Fission gas release,%
TP-1	#	0.12	1642	2.1	1440	0.92
TP-2	#	0.10	1606	2.02	1325	1.10
TP-3	#	0	1608	1.77	1380	2.33
TP-4	< 1120	0.16	1586	1.75	1380	0.95
TP-5	< 1120	0.37	1674	2.56	1525	1.14

#Ceramography is awaited

Extension of burnup and additional loading of MOX fuel assemblies to the TAPS core require demonstration of fission gas release in MOX fuel in the same range as that of enriched UO_2 fuels. Fuels in the commercial Power reactors like TAPS showed strong correlation between heat rating and power ramp with the amount of fission gas release. Fuel pins with low heat rating maintained low level of release even at high burnup whereas higher release was observed in high rated fuel pins even at low burnup. Similar behaviour has also been experienced in experimental fuel pins of AC-2 and AC-3 clusters. These observations suggest that fuel centre line temperature is a major parameter in controlling fission gas release. Experiment conducted in Halden Reactor [5] puts 1% release as threshold for which an empirical relationship of centre line temperature and burn up has been derived purely based on experimental data. The correlation is remarkably successful in predicting fission gas release behaviour for a variety of fuel of different design and manufacture including MOX fuel. Using the Halden correlation for a burnup of 16,265 MWD/T, the threshold temperature for 1% release works out to be 1235°C. From the examination of autoradiographs of the fuel cross section, the estimated centre temperature for AC-2 cluster is <1120°C, which is lower than the threshold and hence <1% fission gas release in the MOX fuel pins in AC-2 cluster is expected. The estimated fuel centre line temperature of AC-3 cluster is 1330°C, which is higher than the threshold temperature calculated from Halden correlation and therefore, higher fission gas release measured in the fuel pins in AC-3 cluster is not surprising.

MOX fuel elements in TAPS core will be operating at an average heat rating of about 90 W/cm^2 under normal operating condition. Hence <1% fission gas release is expected. Peak heat flux experienced by MOX fuel in TAPS will be ~110 W/cm^2 where the fission gas release 8–10% is expected. The fission gas release behaviour of MOX fuel is comparable with the gas release from the standard enriched UO_2 fuel pins in TAPS reactor.

Controlled porosity fuel pin TP-3 in AC-2 cluster, operating at heat rating of 90W/cm^2 has shown no release of fission gas. Further irradiation of fuel elements with pellets of similar characteristics to high burn up and at higher heat rating in the power reactor operating condition is needed to generate more data. Similarly, fuel pins with annular pellets in AC-4 cluster have shown no fission gas release in the exploratory irradiation at heat rating of 110 W/cm^2 to a burnup level of 2000 MWD/T. Irradiation to higher burn up levels in power reactor operating condition is being planned to generate in reactor data.

7. CONCLUSIONS

(1) Less than 1% fission gas release was observed in UO_2-4%PuO_2 MOX fuel irradiated with 93 W/cm^2 heat flux at a burn up of ~16,000 MWD/T.

(2) Fission gas release of the order of 8 –10% was noticed in UO_2-4%PuO_2 MOX fuel elements operated at peak heat flux of 110 W/cm^2 at a burn up of ~16,000 MWD/Te.

(3) Fuel pins with controlled porosity pellets operated at the heat flux 93 W/cm^2 showed no fission gas release up to a burn up of ~16,000 MWD/T.

(4) Annular pellets operating at 110 W/cm^2 showed no fission gas release.

(5) Combination of annular fuel pellet design with controlled porosity appears to provide the best promise of achieving high burn up with low fission gas release.

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ANNEX

Total no. of moles of gases present in the injection loop **N=PV/RT** ------(1)
where P is the pressure at which the gas is injected in Torr, V is the loop volume in litres, R is the gas constant (62362 lit.atm), T is the temperature of the sample loop in ° K.
No. of moles of He in the injection loop $\mathbf{n_{He}=N-(n_{Xe}+n_{Kr}+n_{O2}+n_{N2})}$ -------(2)
where n_{Xe}, n_{Kr}, n_{O2} and n_{N2} are the no. of moles of respective gases which can be estimated from the calibration curves of peak area v/s no. of moles of respective gases which have been already generated at identical detector parameters of the GC.

Since the internal pressure of the fuel pin, $P_5= P_{He}+ P_{Xe}+P_{Kr}$, partial pressure of each component gas can be estimated from the mole fraction of the gases as follows:

$\mathbf{P_{He}= P_5* n_{He}/(n_{He}+ n_{Xe}+n_{Kr})}$ ----------------------(3)

$\mathbf{P_{Xe}= P_5* n_{Xe}/(n_{He}+ n_{Xe}+n_{Kr})}$ -----------------------(4)

$\mathbf{P_{Kr}= P_5* n_{Kr}/(n_{He}+ n_{Xe}+n_{Kr})}$. -----------------------(5)

Internal pressure due to fission gases Xe and Kr alone can be estimated from:

$\mathbf{P_{f.g}=P_{Xe}+P_{Kr}}$ ----------------------------(6)

Considering production of 31 cc of fission gas per kg of fuel per MWD burnup[1], the quantity of fission gases generated in the fuel, $V_{f.g}$, was estimated using the equation:

$\mathbf{V_{f.g} = 31*W*B}$ ----------------------------(8)

where $V_{f.g}$ = volume in cc at STP of fission gases generated in the fuel
W = weight of the fuel in kg and
B = fuel element burn-up in GWD/T of UO_2.

The volume at STP of fission gases released was estimated from:

$\mathbf{V_r = Void\ volume*P_{f.g}*(273/300)/P_a}$ -------------------(9)

Where V_r= volume of fission gases in cc at STP present inside the fuel element
$P_{f.g}$ = internal pressure due to fission gases and
P_a = atmospheric pressure
300= temperature of the hot cells, in K

% released fission gases = $\mathbf{V_r*100/ V_{f.g}}$ -------------------(10)

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