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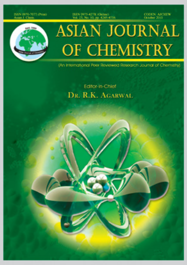

Development of a Measurement System for Fission Gases Released from Spent Pressurized Water Reactor Fuel during Heat Treatment†

JUNG-WON LEE*, JANG-JIN PARK and GEUN-IL PARK

Recycling Process Demonstration Research Division, Korea Atomic Energy Research Institute, Daeduk daero 989-111, Yuseong-gu, Daejeon 305-353, Republic of Korea

*Corresponding author: Fax: +82 42 8688824; Tel: +82 42 8688268; E-mail: jwlee3@kaeri.re.kr

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For an evaluation of the release behaviour of fission gases during DUPIC (direct use of spent pressurized water reactor fuel in CANDU reactors) fuel fabrication processes, a γ-ray on-line monitoring system was developed. The system is composed of a γ-spectrometer located outside a hot cell and a tubular furnace located inside the hot cell. The γ-detector was aligned with the sample located in the tubular furnace in the hot cell to measure the radioactivity of the sample through the hole of the shield plug in the hot cell wall. Using this monitoring system, a quantitative analysis of the release behaviour of fission gases was conducted during heat treatment of spent pressurized water reactor fuel. It was observed that Cs-137 was not released during oxidation at 500 ºC, but almost 99 % of Cs-137 was released during sintering at 1800 ºC. Verification test of the measuring system was performed by comparing experimental results with chemical analysis data.

Key Words: Fission gas, DUPIC, γ-Ray, Cesium release.

INTRODUCTION

Since 1991, DUPIC (direct use of spent pressurized water reactor fuel in CANDU reactors) fuel cycle technology has been developed at KAERI (Korea Atomic Energy Research Institute) for the reuse of spent PWR (pressurized water reactor) fuel as a fuel for CANDU (Canada deuterium uranium) reactors[1-3]. For the fabrication of DUPIC fuel, as shown in Table-1, a spent PWR fuel material undergoes heat-treatments such as oxidation for powdering, OREOX (3 cycles of oxidation at 450 ºC in air and reduction at 700 ºC in 4 %-H_2/Ar) for enhancing the powder properties and sintering for making dense pellets. During heat treatments, some of the volatile and semi-volatile fission gases are released from the spent fuel material. It is important to understand the release behaviour of fission gases for producing a good quality DUPIC fuel along with powder properties. The DUPIC fuel technology has been well demonstrated by establishing an optimal process using typical spent PWR fuel[4]. For further improvements, the effect of fuel burn-up on the DUPIC fuel fabrication should be verified as one of the key process parameters.

This paper describes the release behaviour of the fission gases such as C-14 and Kr-85 during the oxidation and OREOX processes in terms of the spent fuel burn-ups and the release characteristics of cesium under different thermal treatment conditions.

TABLE-1
HEAT TREATMENT CONDITIONS FOR
DUPIC FUEL FABRICATION

Process	Operating conditions
Oxidation	Oxidation: 500 ºC, 5 h, Air, 5.5 L/min
OREOX	Oxidation: 450 ºC, 2 h, Air, 5.5 L/min Reduction: 700 ºC, 7 h, Ar/4 % H_2, 13 L/min Passivation: 80 ºC, 4 h, Ar/2 % O_2, 2 L/min Heating rate: 4 ºC/min
Sintering	Sintering temp./time : 1,800 ºC /10 h 4 % H_2/Ar, 8.0 L/min, Heating rate: 5 ºC /min

EXPERIMENTAL

Table-2 shows the spent fuel characteristics used in this experiment. Fig. 1 shows a schematic diagram of the fission gas measurement system, which was installed in a glove box in the hot cell area. The release behaviour of the Kr-85 fission gas was analyzed by using this on-line measuring system. The Kr-85 gas in a gas stream flows into a Marinelli shaped tank, which is equipped with a NaI-type γ-detector. Quantitative analysis for the release fraction of the C-14 fission gas during

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each step was performed using a scrubbing method. Effluent gas from the oxidation process flowed into the 1.5 M NaOH solution at a constant gas flow rate. Liquid samples with the same volume were taken at a specific time interval. The C-14 activity for each sample was analyzed using an LSC (liquid scintillation counting) method. Release fractions of the fission gases were obtained by comparing the cumulative activities from the experiments with an initial inventory calculated using the OREGEN code. Fig. 2 shows a schematic diagram of the equipment for measuring the Cs fission product release. The γ-spectrometer system (GAMMA-X^{TM} HPGE, coaxial photon detector system, ORTEC gamma vision 32 GMX series) was aligned with a fragment of the spent fuel material or a green pellet located in a tubular furnace. The release fraction of Cs was obtained from the count rate change of a γ-spectrum due to the release of cesium during the thermal treatment of a sample. The cumulative release fraction of cesium after the completion of each test was calculated by comparing the count rate of cesium from the spent fuel material before and after the test. The released Cs was completely trapped in a fly-ash filter located at the end of the tubular furnace.

TABLE-2
SPENT FUEL CHARACTERISTICS USED IN THIS EXPERIMENT

	G23 Assembly		Q43 Assembly	K23 Assembly	
Rod No.	5B	K10A	F16	B16	M03
Initial enrichment	3.21 %			4.2 %	
Burn-up (MWd/tU)	27,300	35,000	37,000	58,000	65,000
Decay time (yrs)	16	18	9	9	3
NPP	Gori # 1		Gori#2	Uljin # 2	

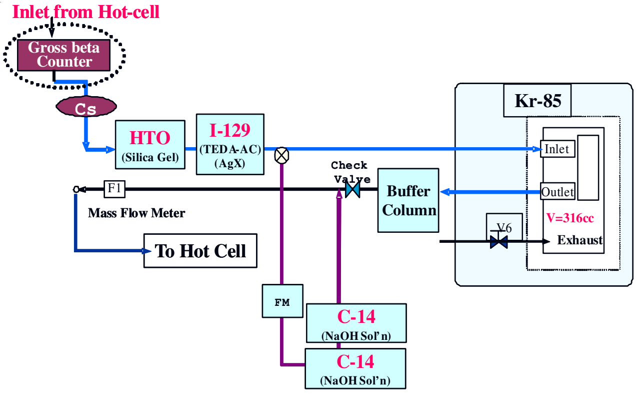

Fig. 1. Schematic diagram of the measuring system for fission gases released from the oxidation and OREOX processes

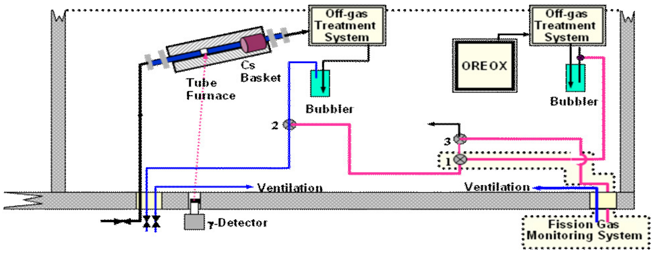

Fig. 2. Schematic diagram of the measuring system for the cesium release

RESULTS AND DISCUSSION

Release behaviours of Kr-85 and C-14 fission gases: The release behaviours of Kr-85 during oxidation and the OREOX process are shown in Figs. 3 and 4. As shown in these figures, Kr-85 started to be released at 350 ºC during oxidation of the spent fuel. However, during the OREOX process, no release of Kr-85 was observed in the 1st oxidation step of the 1st cycle of OREOX, but there was a release in the 1st reduction step at 700 ºC (Fig. 4). Release of Kr-85 was also detected in the 2nd OREOX cycle. The release rate of Kr-85 increased sharply in the early stages and then decreased to a non-detectable level. These results imply that the release behaviour of Kr-85 is strongly dependent on the temperature history, not the gas atmosphere. This also means that a longer process time is required to completely release Kr-85 from spent fuel with a higher burn-up.

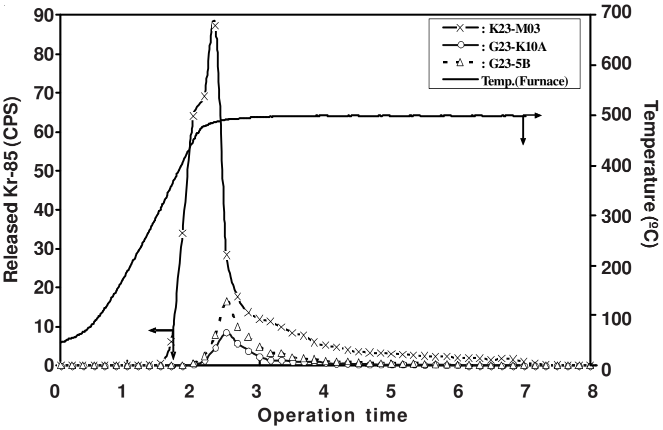

Fig. 3. Release behaviour of Kr-85 with fuel burn-ups during the oxidation process

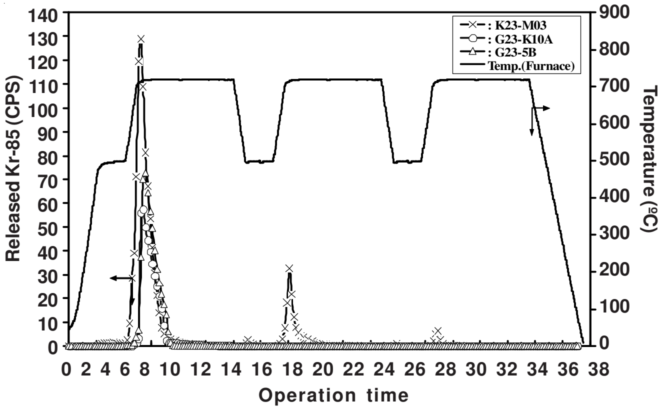

Fig. 4. Release behaviour of Kr-85 with fuel burn-ups during the OREOX process

The release behaviours of C-14 during oxidation and the OREOX process are shown in Fig. 5. The behaviour was similar to that of Kr-85. After starting its release at *ca.* 300 ºC during the oxidation step, a sharp increase of the release rate at the 1st reduction step up to 700 ºC during the OREOX process was observed. The maximum activity of C-14 in the off-gas stream during the OREOX process was about 7 to 10 times higher than that at the oxidation step of 500 ºC. It was also observed that a higher burn-up spent fuel requires a longer operation time for a complete removal of C-14 from the spent fuel. Conclusively, it was confirmed that Kr-85 and C-14 were completely removed during the oxidation and OREOX processes.

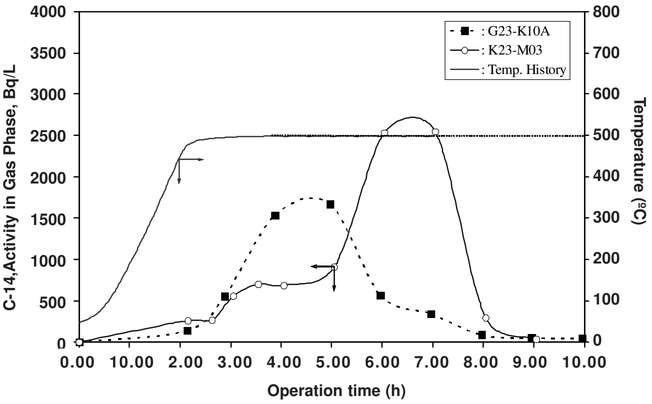

Fig. 5. Release behaviour of C-14 with fuel burn-ups during the oxidation process

Release behaviours of the Cs fission product: The release behaviours of Cs during sintering are shown in Fig. 6. Green pellets were fabricated using OREOX-treated powder and compacted at a pressure range of 80-120 MPa. Cesium was released within a temperature range of 1,100 ºC to 1,450 ºC during the sintering step. The sintering process is generally divided into three stages: an initial stage of sintering (neck formation and growth), an intermediate stage of sintering (network of tubular pores) and a final stage of sintering (pore shrinkage). The onset of densification for sintering occurs at above 1,100-1,150 ºC[4]. Therefore, it is clear that the temperature range of cesium release from a green pellet corresponds to the intermediate sintering stage. It has also been reported that cesium is gaseous at above 1,200 ºC and that a release of cesium occurs by an axial migration in the grain boundary porosity in the central region of the fuel[5]. Based on this result, it was found that a gaseous cesium release during an intermediate sintering stage affected the network formation of the pores, followed by a formation of residual pores in the green pellet. Verification test of γ-ray on-line monitoring system was performed in terms of Cs release percentage from spent fuel by comparing experimental results with chemical analysis data. Spent fuel as fragment type was thermally treated up to 1200 ºC with holding time of 10 h under Ar atmosphere. After heat treatment, spent fuel samples were chemically analyzed. Table-3 shows comparison results of Cs release percentage from spent fuel. It was confirmed that Cs release percentage from this measurement system is within a deviation of 10 % in comparison with chemical analysis method.

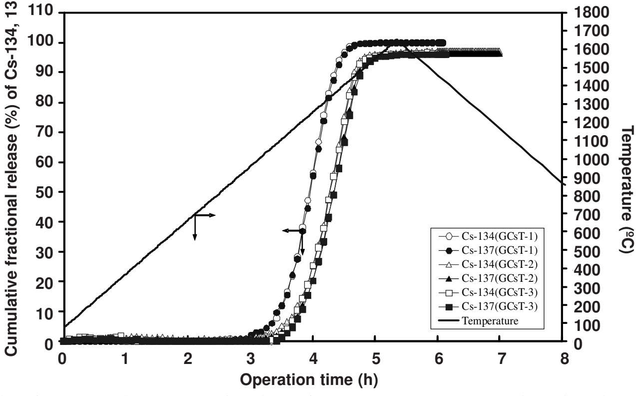

Fig. 6. Cumulative release fraction of Cs-134 and Cs-137 during sintering

TABLE-3
COMPARISON OF Cs RELEASE PERCENTAGE
WITH CHEMICAL ANALYSIS

Spent fuel	Q43-F16 (%)	K23-B16 (%)
Cs release % (This system)	90	88
Cs release % (Chemical analysis)	95	94

Conclusion

The release behaviour of fission gases during heat treatment was studied using a measurement system developed for the DUPIC fuel fabrication. Volatile fission gases of C-14 and Kr-85 were completely released during the OREOX process and showed similar release characteristics. The release fraction percentages of C-14 and Kr-85 were estimated to be 7 to 15 % during the oxidation step and 70 to 84 % during the reduction step. A higher burn-up fuel showed a higher release fraction than that of a low burn-up fuel during the oxidation step. Release of cesium from the spent fuel started at about 800 ºC and release fraction of cesium was estimated to be about 15 % during oxidation at 500 ºC and the subsequent reduction condition at 1,000 ºC. However, about 90 % release fraction of cesium was observed after reaching 1,250 ºC. It was also confirmed that Cs release percentage from this measurement system was within a deviation of 10 % in comparison with chemical analysis method.

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