

STUDY ON GASEOUS EFFLUENT TREATMENT DURING DISSOLUTION TESTS OF SPENT FUEL

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ABSTRACT

Iodine-129 capturing capability of AgS (silver impregnated silica-gel) column was studied in the dissolution tests using 1.5 kg of spent fuel with a burnup of 8,000 MWd/t. Removal of carbon-14 as carbon dioxide by KOH aqueous solution was also studied in the tests. The tests are the first stage of a series of experimental works using spents fuels up with burnups up to 45,000 MWd/t. Iodine evolved during the dissolution and the iodine stripping operation which comprised two steps. Total amount of iodine-129 trapped in AgS columns was found to be about 210 kBq which corresponded to about 62 % of the iodine-129 contained in the 1.5 kg of spent fuel according to an estimation by ORIGEN calculations. No significant amount of iodine was measured in the dissolver solution. Previous tests suggested that rest of iodine adhered to the inner surface of piping between the dissolver and the AgS columns. Total amount of carbon-14 captured during the dissolution tests was measured to be about 2 MBq, which indicated that at least several ppms of nitrogen-14 was initially contained in the fuel.

INTRODUCTION

The dissolution of spent fuel in a reprocessing plant releases volatile species such as iodine as dissolver off-gas (DOG). In addition, carbon-14, which is generated mainly from nitrogen-14 initially contained in fresh fuel, is released as carbon dioxide. Iodine-129 and carbon-14 are long-lived nuclides and it is important to control those nuclides and to develop and demonstrate confinement technology in order to reduce the release to the environment.

In the Nuclear Fuel Cycle Safety Engineering Research Facility (NUCEF) a series of tests are carried out to study the capability to confine the radioactivity within the process (dissolution, off-gas treatment, extraction and effluent treatment) under normal operation using spent fuel. The test uses spent fuel with burnups from 8,000 to 45,000 MWd/t. The paper reports the test results of iodine-129 capturing by AgS which will be used in Rokkasho Reprocessing Plant, and carbon-14 measurement for the development of capturing technology using adsorbent.

EXPERIMENTAL

Figure 1 shows the dissolver off-gas (DOG) treatment apparatus within NUCEF. The dissolution process comprises a dissolver, an iodine stripping tank, a dissolver solution filter, an extraction feed tank, a condenser, a scrubber, iodine adsorbent columns and a blower. The equipment is installed in the α cell within NUCEF except for the iodine adsorbent columns and blower, which have been installed in a glove-box above the α cell. The blower is used to maintain the dissolution process under negative pressure for all operating condition.

Spent fuel was dissolved in the dissolver with 5 N or 4.5 N HNO_3 . The present study used 1.5 kg of PWR spent fuel (8,000 MWd/t, cooling time: about 19.5 years) which has been sheered into short pieces. Dissolution was carried out three times and about 500 g of SF was used in each dissolution. Following the dissolution, all dissolver solution is transported to the iodine stripping tank where NO_2 gas is purged through the dissolver solution to strip remaining iodine. After iodine stripping, the dissolver solution is then filtrated to remove all particulate residue and subsequently adjusted to 3M HNO_3 and 250 g/l uranium concentration as extraction feed solution. Table I lists up the experimental condition used in the dissolution tests.

TABLE I
Dissolution Condition Used in the Present Study

Run No.	U element [g]	HNO_3 Concentration [N] (volume [l])	Temperature [K]	Dissolution time [min]
1	436.05	5 (1.7)	373	200
2	443.01	5 (1.7)	373	200
3	439.90	4.5 (1.7)	373	260

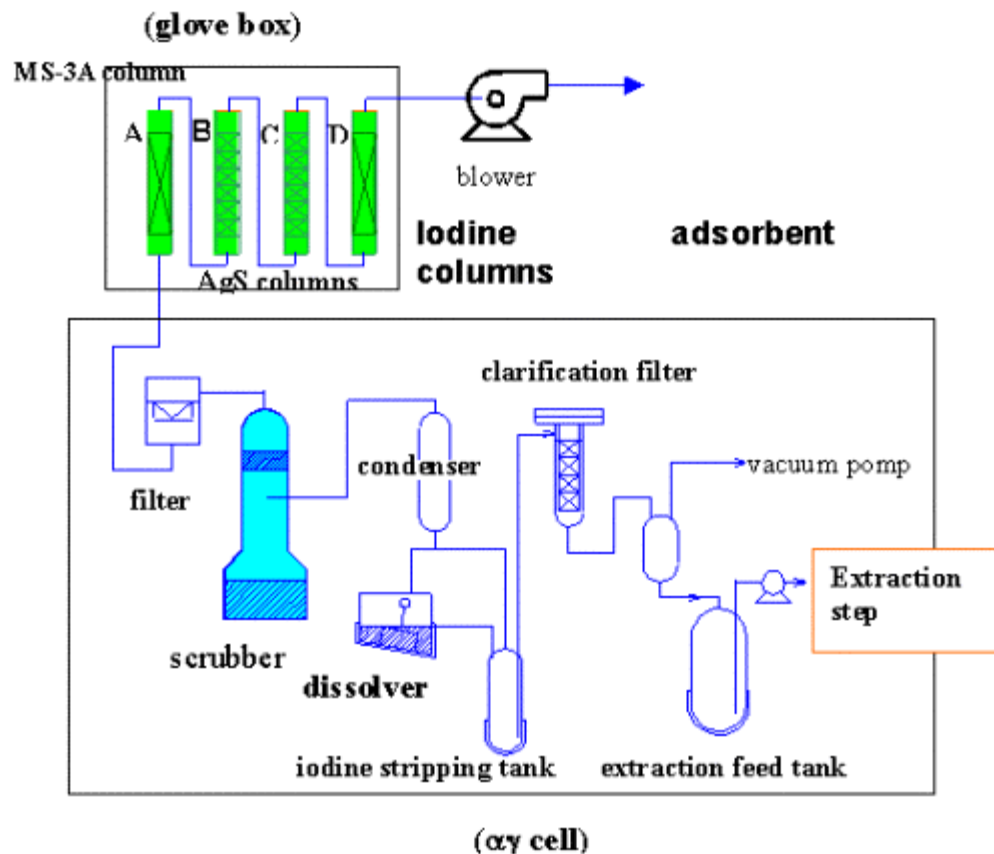


Figure 1 The dissolver off-gas treatment apparatus with the miniature scale reprocessing plant in $\alpha\gamma$ cell within NUCEF-BECKY

Iodine stripping is carried out in two-steps operation which has been developed by Sakurai et al.(1). In the first-step operation KIO_3 is added to all dissolver solution collected in the iodine stripping tank heated up at 373 K and then the solution is maintained at the same temperature for two hours. In the second-step operation 50 % NO_2 gas is introduced into the tank at a flow rate of 170 l/h and maintained at 373 K for two hours.

Dissolver off-gas and iodine stripping off-gas via the scrubber pass through a HEPA filter and then into the glove-box located above $\alpha\gamma$ cell where iodine is collected by the adsorbent columns which are installed in the glove-box. In order to reduce the adhesion of the iodine to piping associated with the off-gas treatment system, all piping is maintained at 323 K. The first column (Column A) contains 130 g of MS-3A to remove H_2O contained in the off-gas. The column was maintained at 333 K. Other three columns contains AgS (silver impregnated silica-gel) particles and capture iodine by maintaining the column temperature at 423 K. The first two of those three columns (Column B and C) have seven cartridges respectively and each cartridge (44 mm o.d. and 15 mm in length) accommodates 3.4 g of AgS. The fourth column (D) also contains AgS but not divided in cartridges. The off-gas flow rate in the dissolution and iodine stripping process is approximately 400 l/h. The iodine collection efficiency of the columns was over 99.9 %, which was confirmed in the test prior to the present study.(2)

The AgS adsorbent was supplied as AC6120 from SÜD CHEMIE AG. More than 98 % of the adsorbent particles were sized between 1 and 2 mm. The bulk density was 719 kg/m^3 and its silver content was 12.1 wt%.

Measurement of carbon-14 amount is carried out using a CO_2 trap containing 2M KOH aqueous solution which is connected to a bypass line located between the column C and D. The off-gas flow rate in the CO_2 trap (i.e. in the bypass line) was 6 l/h. The effect of NO_x gas contained in the off-gas on the CO_2 capturing efficiency of the trap was checked prior to the experiment. It was necessary to reduce the NO_x concentration lower than 1 % to avoid the adverse effect. During the spent fuel dissolution tests the concentration of NO_x was maintained at low level by the scrubber and was found to be 0.025 %.

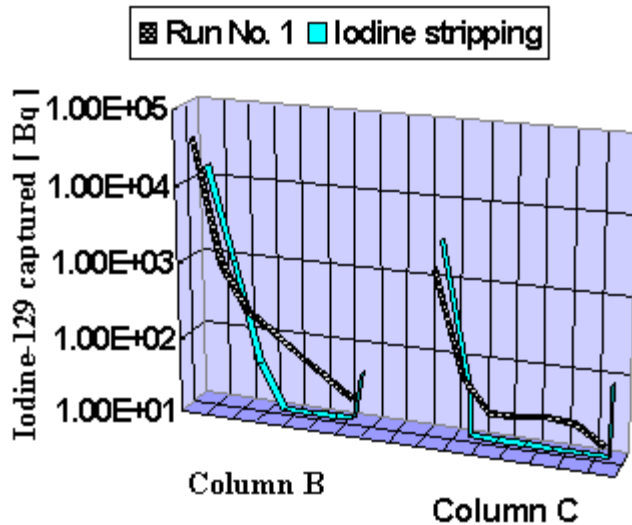


Figure 2 Concentration profiles of Iodine-129 captured by AgS Columns (Columns B and C)

RESULTS AND DISCUSSION

Iodine Captured by AgS

Figure 2 shows concentration profiles of iodine-129 captured in AgS columns (B and C) during the first dissolution test and iodine stripping operation, respectively. Each cartridge has 3.4 g of AgS and adsorption capacity of AgS is 135 mg-iodine/g-AgS (3). Therefore, each cartridge can capture 0.459 g of iodine, which corresponds to 3 MBq of iodine-129. In the present study, as shown in the figure each cartridge did not reach the saturated concentration of iodine.

Table II summarizes the radioactivity of iodine-129 in the apparatus. It can be seen that MS-3A did not adsorb iodine very much and that Column B adsorbed most of iodine-129 introduced into the apparatus. The result suggests almost all the iodine introduced into the columns were removed in the columns.

Amount of iodine-129 contained in the spent fuel was estimated by ORIGEN calculation. In the calculation burnup was corrected to be 8,000 MWd/t by measuring concentration ratio of Cs-134 and Cs-137 in the dissolver solution. The estimated amount of iodine-129 was 337.5 kBq/1.5 kg-SF. Total amount of iodine-129 captured by the column was about 211 kBq which corresponds to about 62 % of estimated amount.

The fraction of iodine collected in the columns are supposed to be in the range of reasonable amount, because the iodine distribution which had been measured in the previous study using simulated spent fuel solution have shown the iodine collected in the columns to be about 74 % of the total iodine added (2). It was shown that only 0.37 % and iodine remained in the dissolver

solution and almost all iodine was removed to the dissolver off-gas. The other part of the iodine was distributed in the HEPA filter, scrubber, surface of off-gas pipeline, insoluble residue, and other places in the head-end process. In the present study, iodine-129 concentration in the dissolver solution was measured by the extraction of iodine using CCl_4 and the results suggested that there was no significant amount of iodine remained in the dissolver solution, though the iodine contained in insoluble residue has not been measured because of high radioactivity. Sakurai and Takahashi (3) showed less than 3 % of iodine to be found in insoluble residue. The rest of iodine in the present study seemed to adhere to the surface of off-gas piping. And it can be concluded that AgS columns was effective to capture iodine under spent fuel dissolution.

TABLE II
Amount of Iodine-129 Captured in the Adsorbent Columns

Column	Iodine Captured [Bq]					Fraction [%]
	Run No.1	Run No.2	Run No.3	Iodine stripping	Total	
A					703	0.33
B	42,400	84,000	56,100	16,000	199,000	94.44
C	1,960	1,180	2,680	3,310	9,130	4.33
D					1,890	0.90
Total	44,360	85,180	58,780	19,310	210,723	100.00

Measurement of Carbon-14 contained

Carbon-14 trapped in the 2M KOH aqueous solution was measured by β -LSC. The result showed that the 1.5 kg of spent fuel contained about 2 MBq. ORIGEN calculation estimated the concentration of nitrogen initially contained in fresh fuel. As shown in Fig. 3, about 5 ppm of nitrogen-14 was estimated to generate 2 MBq of carbon-14. Therefore, at least 5 ppm of nitrogen-14 was contained as impurity in the fuel.

CONCLUDING REMARKS

Iodine-129 was effectively captured by AgS column during dissolution test of 1.5 kg of spent fuel. About 62 % of iodine-129 contained in the spent fuel was collected, which was based on ORIGEN calculation. Carbon-14 can be trapped as carbon dioxide by KOHaq., when concentration of NO_x gas is lower than 1%.

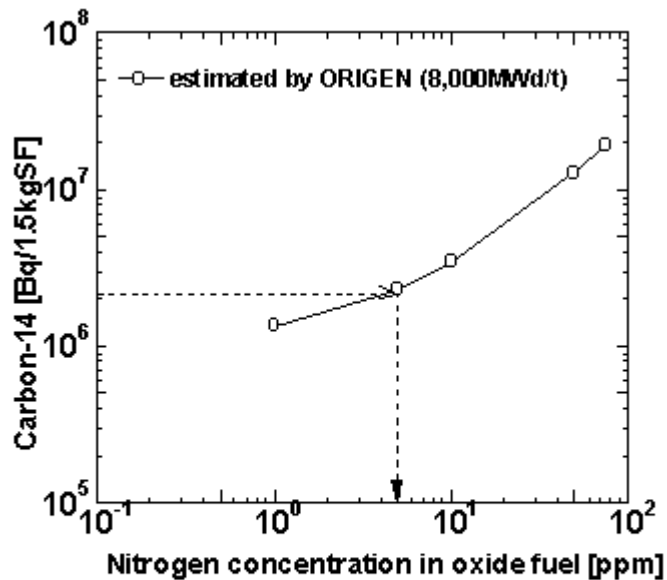


Figure 3 Carbon-14 amount estimated by ORIGIN calculation

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REFERENCES

- (1) Sakurai, T., Takahashi, A., Ishikawa, N., Komaki, Y., Ohnuki, M., and Kato, K., Proc. 23rd DOE/NRC Nuclear Air Cleaning and Treatment Conference, NUREG/CP-0141, CONF-940738, 321(1995).
- (2) Kihara, T., Sakurai, T., Nakano, Y., and S. Fujine, Proc. RECOD'98, France, 1998
- (3) Sakurai, T., and Takahashi, A., JAERI-Review 97-002, 1997 (Japanese)