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Formation of zirconium molybdate sludge from an irradiated fuel and its dissolution into mixture of nitric acid and hydrogen peroxide

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ABSTRACT

In the dissolution step of the PUREX process, a sludge of zirconium molybdate can be generated from zirconium and molybdenum dissolved in the solution. In order to avoid accumulation of sludge, a new cleaning method using a mixture of nitric acid and hydrogen peroxide was developed. In this work, the cleaning method was verified using an irradiated fuel. Cubic crystals of the zirconium molybdate $ZrMo_2O_7(OH)_2$ were formed upon dissolution of an irradiated PWR fuel. The sludge deposition continued after the completion of the fuel dissolution. A small amount of fissile material was incorporated into the sludge. Although almost the same amount of insoluble residue was present, the sludge could be dissolved with a HNO_3–H_2O_2 solution at 80 °C within 5 h. The sludge formation on the clean Zr surface is inhibited.

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1. Introduction

In the dissolution step of the PUREX process, spent oxide fuel is dissolved into nitric acid (HNO_3). Most of the fuel constituents, mainly oxides of uranium (U) and plutonium (Pu) are dissolved in this step, while a small amount of alloy remains as insoluble residue. The insoluble residue contains platinum group metals (PGM, i.e. ruthenium (Ru), rhodium (Rh) and palladium (Pd)), technetium (Tc) and molybdenum (Mo) [1,2]. A part of these elements are dissolved in the dissolution step.

It was reported that depending on the nitric acid concentration, a more or less big part of dissolved zirconium (Zr) and Mo precipitate as zirconium molybdate during the dissolution step [3,4]. In this article, the authors call this precipitation "sludge". Since the sludge is a crystalline material, it may grow on the wall of the dissolver. It is better to keep the dissolver free from the sludge, because accumulated sludge may hamper the heat transfer from the dissolver wall to the nitric acid solution.

The mechanisms of generation and adhesion of sludge were investigated in detail and a new method of removing the sludge using an HNO_3–hydrogen peroxide (H_2O_2) mixture was developed by Yasuike et al. [5]. It was reported that sludge in the form of $ZrMo_2O_7(OH)_2$ was generated within 90 min in 90 °C-3 M HNO_3 by dissolution of UO_2–ZrO_2–MoO_2 simulated fuel. The sludge was dissolved in HNO_3–H_2O_2 at 70 °C at a mole ratio of H_2O_2/sludge exceeding 10. The dissolution rate of sludge decreased in the presence of RuO_2 because a part of the H_2O_2 was decomposed.

There are inherent differences between simulant materials and the irradiated fuel to be considered. The generation behavior of sludge can be different because a solution of irradiated fuel contains many elements and chemical species such as nitrous acid. Fines of an insoluble residue possibly existing in the dissolver may change the dissolution behavior of the sludge by decomposing H_2O_2. For these reasons, it is necessary to verify the sludge dissolution technology with HNO_3–H_2O_2 under realistic conditions before utilizing for the industrial dissolver. Therefore, a sludge generation test using an irradiated fuel and dissolution tests on the sludge were carried out.

Before using the irradiated fuel, non-radioactive tests and U/Pu solution tests were also carried out. An objective of the preliminary tests is to estimate the generation rate of sludge. It is unknown what percentage of Mo is dissolved into HNO_3 and how much remains as insoluble residue although the generation rate of sludge is expected to depend on the concentrations of Mo and Zr in the solution. Therefore, the relationship between the generation rate of sludge and the initial concentration of Mo was determined through non-radioactive tests. Dissolution of the sludge was also verified. Experiments using U/Pu solutions were also carried out to observe the effect of U/Pu on the composition of the sludge.

2. Experiments

2.1. Sludge generation from non-radioactive reagents and its dissolution

Standard experimental conditions were determined on the assumption that UO_2 fuel with burn-up of 40 GWd/t would be

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dissolved to make 250 g-U/l of 3.0 M HNO_3 solution, and fission products are completely dissolved. In this case, concentrations of Mo and Zr were estimated to be 1.28 g/l and 1.33 g/l, respectively.

When a part of Mo generated by fission forms insoluble residue, concentration of Mo in the solution is lower than the standard condition, i.e. 1.28 g/l. The amount of Mo which forms insoluble residue varies with fuel burn-up [6]. In order to simulate these situations, initial Mo concentration was varied between 20% and 100% of standard concentration. When Mo concentration is 80% of standard condition, it means 20% of Mo remained in the insoluble residue.

Non-radioactive tests on the sludge generation were carried out in a 300-ml round-bottomed flask equipped with a reflux condenser and a thermocouple. In order to simulate the wall of the dissolver, a test piece of Zr metal was placed in the flask in some of the experiments. As starting materials, sodium molybdate (Na_2MoO_4) and zirconium (IV) dinitrate oxide hydrate ($ZrO(NO_3)_2{\cdot}H_2O$) were dissolved into water individually. Solutions of Mo and Zr as well as HNO_3 and water were mixed in the flask to make a 3 M HNO_3 solution and then heated to 90 °C. Samples of the solution were taken during the test to analyze the concentrations of Mo and Zr, which decrease with sludge generation. The concentrations of Mo and Zr were analyzed by ICP-AES. After the sludge generation tests, the sludge on the Zr test piece was observed by scanning electron microscopy (SEM) and X-ray diffraction (XRD).

Dissolution tests on the non-radioactive sludge were also carried out. A flask was filled with Mo, Zr and nitric acid in standard experimental conditions and then kept at 90 °C for 48 h to generate a sludge. After cooling, this flask with sludge on the inner wall was washed, dried, filled with 4 M HNO_3 containing 2 wt.% of H_2O_2, and then heated to 80 °C. Samples of the solution were taken to analyze the concentrations of Mo, Zr and H_2O_2. The concentration of H_2O_2 was analyzed by redox titration using 0.02 M potassium permanganate ($KMnO_4$) standard solution. The precipitate obtained after complete decomposition of H_2O_2 was observed by SEM.

2.2. Sludge generation from U/Pu solution with non-radioactive reagents

Sludge generation tests using U/Pu solution were carried out in a 250-ml or 50-ml flask. In order to simulate the wall of the dissolver, a test piece of Zr metal was placed in the flask. Solutions of Na_2MoO_4, $ZrO(NO_3)_2{\cdot}H_2O$, U, U–Pu mixture, HNO_3 and water were mixed in the container to make a 3 M HNO_3 solution and then heated to 90 °C. Samples of the solution were taken during the test to analyze the concentrations of Mo, Zr, U and Pu by ICP-MS. The amount of the sample was 1 ml in each time. Since total volume of the solution was much larger, the change in volume of the solution is not so much. A major part of the sludge was dissolved with 3–4 M HNO_3 containing H_2O_2. Detailed conditions are described with results in 3.2.

2.3. Sludge generation from irradiated fuel and its dissolution

An irradiated PWR fuel with burn-up of 60 GWd/t and 4 years cooling time was used for a sludge generation test. The irradiated fuel, 54.2 g, was cut into four pieces of 2 cm in length with cladding and placed in a 250-ml flask. In order to simulate the wall of the dissolver, four test pieces of Zr metal (15 mm × 15 mm × 1 mm) were placed in the flask. Since it was found in non-radioactive tests that the existence of sludge tended to enhance the generation and adhesion of new sludge, "non-radioactive sludge" was previously adhered on three of four Zr pieces by heating in a Mo–Zr–HNO_3 solution. The flask was then filled with 250 ml of 5.8 M HNO_3 and kept at 90 °C for 55 h. One of the Zr pieces with non-radioactive sludge was taken out at 6 h. Samples of the solution were taken at 0, 3, 6, and 24 h to determine the concentrations of the elements by ICP-MS. The solution, claddings, and Zr pieces were taken out from the flask after cooling down. Pieces of Zr were washed with 3 M HNO_3 and water and then dried and weighed. Two Zr pieces kept in the flask for 55 h and a piece taken out after 6 h were observed by SEM, while also one Zr piece with non-radioactive sludge was used for sludge dissolution test.

On the wall of the flask used for the fuel dissolution, sludge and small part of the insoluble residue were adhered. The flask was washed with 3 M HNO_3 and water for some time while keeping adhered sludge on the wall. After being dried and weighed, the flask was filled with 250 ml of 4 M HNO_3 containing 1.8 M H_2O_2 and kept at 80 °C for 5 h. Samples for ICP-MS analysis were taken after 3 h and 5 h. The amount of the sample was 1 ml in each time. After cooling down and removal of the solution, the flask was washed, dried and weighed again.

The sludge on a Zr piece was also dissolved in 100 ml of 4 M HNO_3 containing 1.8 M H_2O_2 at 80 °C for 5 h. After cooling down, the Zr piece was washed again with 3 M HNO_3 and water for some time and then dried, weighed, and observed with SEM.

3. Results and discussion

3.1. Sludge generation from non-radioactive reagents and its dissolution

Fig. 1 shows a typical condition of the flask after the sludge generation test using non-radioactive reagents. The inner wall of the lower half of the flask was covered with a white precipitate. The outer wall of this part was in contact with the furnace. The precipitate was not floating but adhering to the wall of the flask while it came off easily by using an ultrasonic bath. Fig. 2 shows the X-ray diffraction pattern of the precipitation, which completely agrees with that of $ZrMo_2O_7(OH)_2$.

Fig. 3 shows changes in Mo concentration in 3 M HNO_3 with an initial Zr concentration of 1.3 g/l at 90 °C. The initial concentration of Mo was varied in order to simulate its distribution between the solution and the insoluble residue. In Fig. 3, 100% "Mo dissolution" means all Mo generated as FP is dissolved and nothing remains in the insoluble residue. In the case of 20% "Mo dissolution", only 20% of Mo generated as FP is dissolved and 80% remains in the insoluble residue. The concentration of Mo in the solution decreased with generation of $ZrMo_2O_7(OH)_2$ sludge precipitation. The concentration of Zr decreased by half of the Mo decrease in moles, exactly.

In the case of 100% Mo dissolution, about 30% of Mo was removed from the solution by generating sludge within 6 h. The

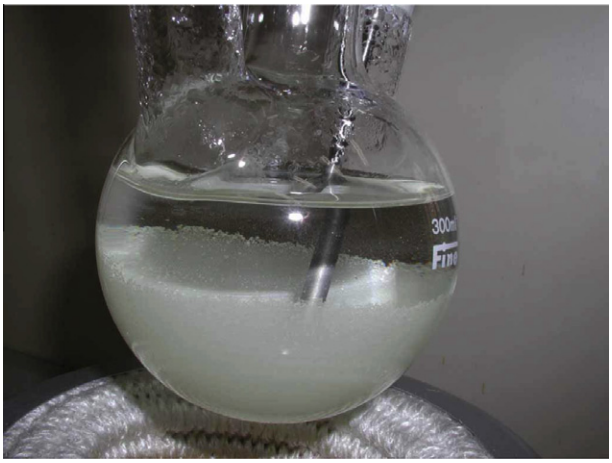

Fig. 1. Photograph of sludge generated from non-radioactive reagents.

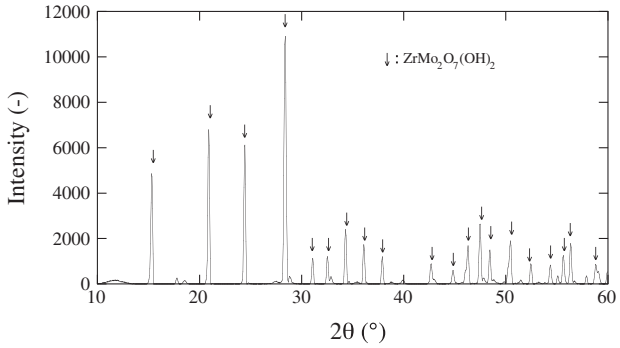

Fig. 2. XRD pattern of sludge generated from non-radioactive reagents.

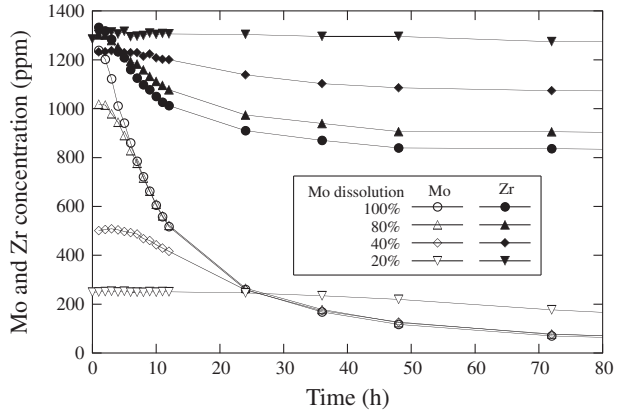

Fig. 3. Decrease in Mo concentration with generation of sludge (when Mo dissolution is 80%, 20% of Mo in an irradiated fuel remains as insoluble residue).

sludge generation rate decreased with Mo dissolution, i.e. the initial concentration of Mo. Almost no sludge was generated in the case of 20% dissolution. Fig. 4 shows the amount of the sludge generated within 24 h. Formation of the sludge continued more than 24 h in every case. Therefore, a longer heating period after completion of fuel dissolution is required to obtain representative amount of sludge. For the irradiated fuel, it was decided to keep the solution at 90 °C for 55 h, which was the longest time reasonably achievable.

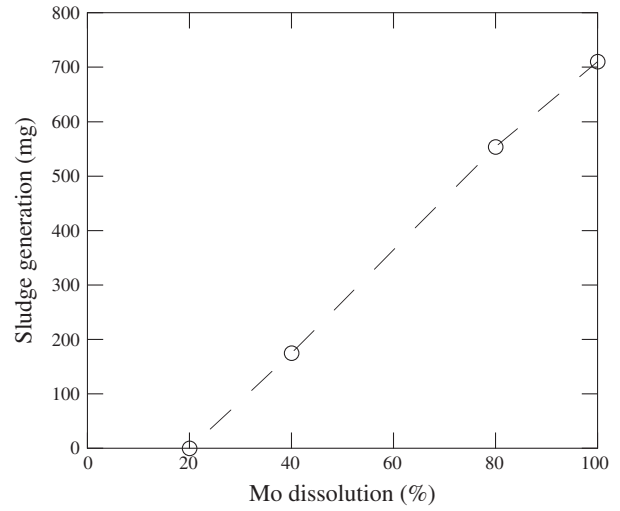

Fig. 4. Dependence of sludge amount in 24 h on Mo dissolution.

In the cases of 40–100% Mo dissolution in Fig. 3, it can be seen from the decrease in Mo concentration that the sludge formation is slower at the beginning. In order to clarify this inhibition effect, a control experiment was carried out using a flask which had an inner wall already covered by sludge from previous experiment. The experimental results are compared in Fig. 5. In this figure, plots "without previous sludge" are the same as 100% "Mo dissolution" in Fig. 3. When previous sludge was present, a fast decrease of the Mo concentration indicates that the sludge generation started almost immediately and at high rate, decreasing at low Mo and Zr concentrations. From these results, it can be concluded that a nucleation process is the rate-determining step in the beginning. Therefore, Zr pieces previously covered with sludge from non-radioactive reagents were used in the irradiated fuel experiment. Concerning the industrial dissolver, this result means that more frequent washing of the dissolver wall could lead to less adhesion of the sludge.

Fig. 6 shows an SEM image of the sludge sparsely growing on a Zr piece that was taken out at an early stage of the test. The sludge shows cubic crystals of 1–10 μm. The crystals on the Zr piece also seem to be randomly distributed. Fig. 7 shows an SEM image of the sludge densely adhering on a Zr piece after 48 h of heating. The size of the crystals remains 1–10 μm but the number of crystals in-

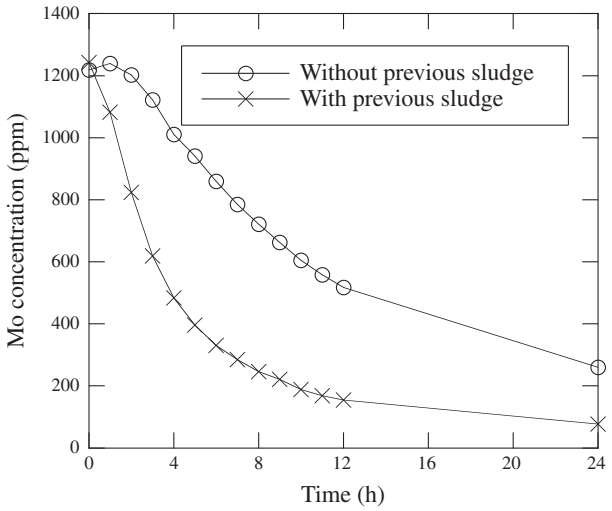

Fig. 5. Effect of previous sludge on the generation rate.

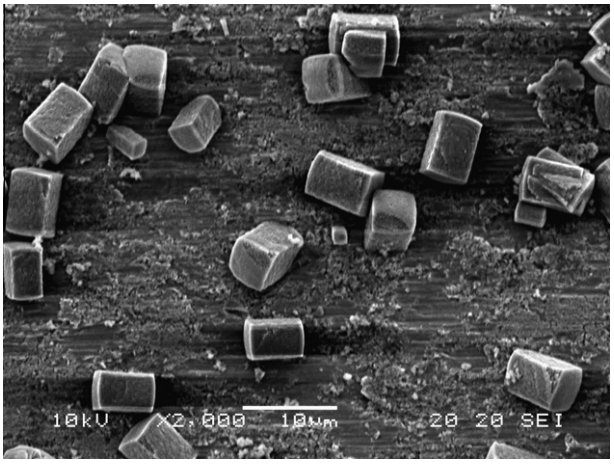

Fig. 6. SEM image of sludge at an early stage.

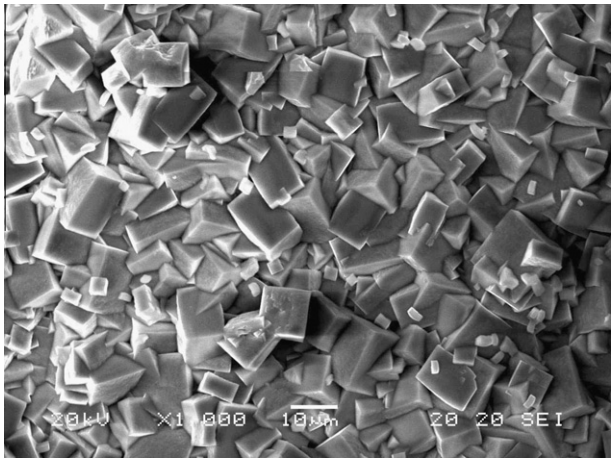

Fig. 7. SEM image of sludge densely grown.

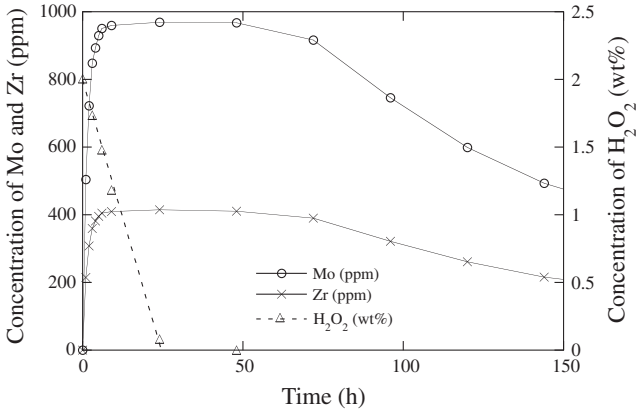

Fig. 8. Dissolution and re-precipitation of sludge in HNO_3–H_2O_2.

creased. Crystals seem to grow on the other crystals as they filled the gaps.

Fig. 8 shows the concentrations of Mo, Zr and H_2O_2 in 4 M HNO_3 solution in contact with 785.4 mg of the sludge at 80 °C. Before contacting with HNO_3–H_2O_2 solution, the appearance of the sludge was similar to Fig. 1 and therefore it was expected that the sludge is dense like Fig. 7 microscopically. It can be seen from Fig. 8 that the sludge was completely dissolved in HNO_3–H_2O_2 within 6 h. The concentration of H_2O_2 decreased linearly even after complete dissolution of the sludge. This result means that a thermal decomposition of H_2O_2 is more dominant than consumption by sludge dissolution when 785 mg of sludge is dissolved in 4 M HNO_3 at 80 °C. The concentrations of Mo and Zr started to decrease again for re-precipitation of the sludge 20–30 h after H_2O_2 has disappeared. Compared with Fig. 3, the decrease of the Mo and Zr concentration is however much slower. This is because process temperature is lower by 10 °C and the Zr concentration is half of that in the initial solution.

3.2. Sludge generation from U/Pu solution with non-radioactive reagents

Table 1 shows the conditions and results of sludge generation experiments using U/Pu solutions. The scale and U/Pu concentrations of "Generation test 1" simulate the irradiated fuel test, while the initial concentrations of Mo and Zr were higher to ensure the generation of sludge. "Generation test 2" was a small-scale test

Table 1
Experimental conditions and results of sludge generation tests using U–Pu solution.

	Generation test 1	Generation test 2
Conditions		
Amount of solution (ml)	250	50
Initial concentration of U (g/l)	190	35
Initial concentration of Pu (g/l)	2.1	2.5
Initial concentration of Zr (g/l)	1.3	2.0
Initial concentration of Mo (g/l)	1.9	2.0
Concentration of nitric acid (M)	3.0	3.0
Temperature (°C)	90	90
Heating time (h)	48	24
Results		
Amount of sludge (g)	1.43	0.20

carried out to verify the result of the first test. This table indicates that 1.43 g and 0.20 g of the sludge were generated in these two experiments, respectively. The weight of sludge may include that of adhered water. These results show that sludge is formed also in the presence of U and Pu. The color of the sludge obtained in these experiments was slightly pink. The variation of elements concentrations observed in "Generation test 1" is plotted in Fig. 9. The concentrations of Mo and Zr decreased with the generation of sludge, while there was no obvious change in U and Pu concentrations.

A part of the sludge generated in "Generation test 1" and "Generation test 2" was used for "Dissolution test 1" and "Dissolution test 2", respectively. The conditions and results of these experiments are shown in Table 2. In each experiment, the sludge was dissolved in HNO_3–H_2O_2 solution within 5 h. The ratio of Mo/(Pu + Zr) was close to 2.0 in both tests, supporting the generation of $ZrMo_2O_7(OH)_2$. The amount of Pu contained in the sludge was about 3 mol% of the Zr content. It can be assumed that Pu is chemically incorporated in $ZrMo_2O_7(OH)_2$ because content of Pu did not decrease by washing with acid. Since Zr and Pu are tetravalent while Mo is hexavalent, Pu is supposed to replace Zr. The amounts of Zr, Mo and Pu in the sludge were calculated from the final concentrations with assuming that total volume of the solution including samples taken was equal to the initial volume. Theoretically, 1.37 g of $Zr_{0.97}Pu_{0.03}Mo_2O_7(OH)_2$ should include 280 mg of Zr, 606 mg of Mo and 22.7 mg of Pu, while 0.18 g of that should include 36.7 mg of Zr, 79.7 mg of Mo and 2.98 mg of Pu. The amounts of elements measured in "Dissolution test 1" were close to these theoretical values. On the other hand, the amounts of elements measured in "Dissolution test 2" were much higher than the theoretical values. Since a reflux condenser was used only in "Dissolution test 1", the volume of the solution was supposed to decrease

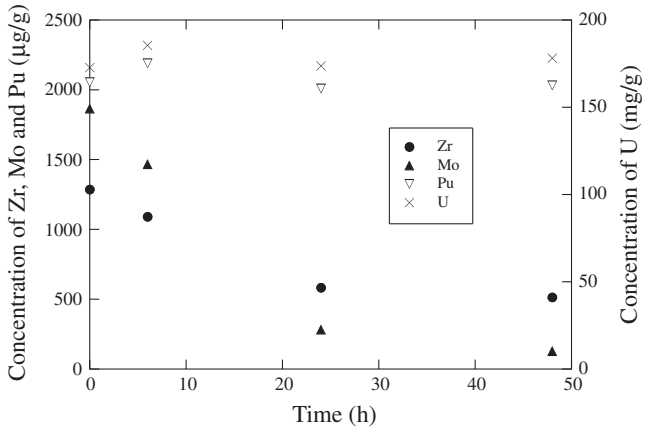

Fig. 9. Change in concentration with sludge generation in U–Pu-Mo–Zr solution.

Table 2
Experimental conditions and results of sludge dissolution tests using U–Pu solution.

	Dissolution test 1	Dissolution test 2
Conditions		
Amount of solution (ml)	250	50
Amount of dissolved sludge (g)	1.37	0.18
Concentration of nitric acid (M)	3.0	4.0
Initial concentration of H_2O_2 (M)	2.0	1.8
Temperature (°C)	80	80
Results		
Dissolution time (h)	5.0	2.0
Final concentration of H_2O_2 (M)	1.15	0.61
Final concentration of Zr (μg/g)	993	922
Final concentration of Mo (μg/g)	1954	2014
Final concentration of Pu (μg/g)	96.9	93.4
Mo/(Pu + Zr)	1.825	2.012
Pu/(Pu + Zr)	0.036	0.038
Amount of Zr in sludge (mg)	246	45.2
Amount of Mo in sludge (mg)	485	99
Amount of Pu in sludge (mg)	24.0	4.58

by evaporation in "Dissolution test 2" and Zr, Mo, Pu were concentrated. When the solution was kept at 80 °C until complete decomposition of H_2O_2, the sludge precipitated again also in these tests.

3.3. Sludge generation from irradiated fuel and its dissolution

Fig. 10 shows the concentrations of U, Pu, Mo, and Zr from the beginning of the dissolution of the irradiated fuel till 24 h. The concentration of U was lower than the original target of 250 g/l because the weight of the cladding was underestimated. As a result, the final concentration of HNO_3 was 3.7 M higher than the original target of 3.0 M. Since it is known that the amount of sludge is lower at higher acid concentration [7], the amount of sludge from this experiment may be less than that from standard condition. The amount of Mo in the solution was about half of the Zr. It is therefore suggested that a significant part of Mo is precipitated in forms of sludge and/or insoluble residue, because nearly equal amount of Mo and Zr are contained in the irradiated fuel.

Fig. 11 shows a photograph of the flask that was washed and dried after removing the solution, claddings, and Zr pieces. The white part of the residue at the bottom should be the sludge, while the smaller black part should be the insoluble residue. The total weight of the adhering materials was 138.9 mg.

The amounts of sludge adhered on the Zr pieces are listed in Table 3 with experimental conditions. As predicted from non-radioactive tests, less amount of sludge adhered on the Zr piece

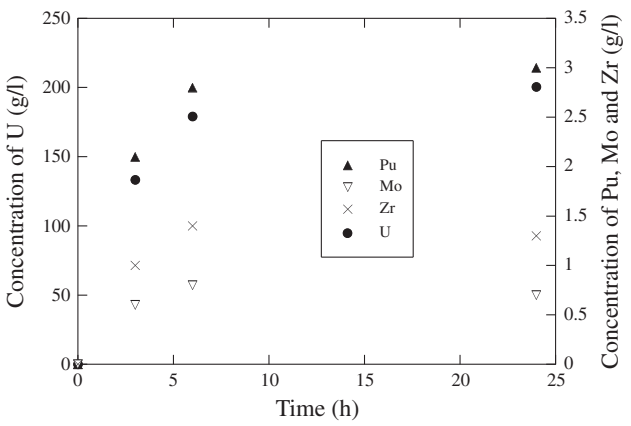

Fig. 10. Change in concentration with dissolution of irradiated fuel.

Fig. 11. Photograph of sludge generated from irradiated fuel.

Table 3
Conditions and results of Zr test pieces used in irradiated fuel tests.

No.	Conditions			Results
	Amount of non-radioactive sludge	Contact time (h)	Usage	Sludge from irradiated fuel (mg)
Zr-1	49 mg	6	SEM observation	3.0
Zr-2	51 mg	55	SEM observation	10.7
Zr-3	Nothing	55	SEM observation	1.0
Zr-4	48 mg	55	Sludge dissolution	20.7

without non-radioactive sludge. The amount of sludge adhered after 6 h on Zr-1 is much less than on Zr-4, which was kept in the solution for 55 h. This result shows again that sludge is generated also after completion of fuel dissolution. The amount of sludge on Zr-2 was about half of that on Zr-4 because only half of the area of Zr-2 was soaked in the solution due to experimental problems.

Fig. 12a and b shows SEM images of Zr-3. The cubic crystals suggest generation of the same sludge as in the non-radioactive tests. The size of the crystals is about 1–10 μm. Oxygen, Mo, and Zr were detected by energy-dispersive of X-ray analysis (EDX). Fig. 12c shows an SEM image of Zr-2. A thicker layer of small dense crystals can be seen at the lower left-hand part of the figure, while larger crystals can be seen at the upper right-hand part. The lower part was the one immersed in the dissolver solution and should therefore be generated from irradiated fuel constituents in the solution. Crystals in this area are denser than those in Fig. 12a and b, indicating that sludge generation was enhanced by non-radioactive sludge. The upper right-hand part of Fig. 12c should be a layer of non-radioactive sludge staying above the surface of the solution. For Zr-1, a deposit similar to the lower left-hand part of Fig. 12c is observed.

When the sludge in the flask was heated in HNO_3–H_2O_2 at 80 °C, there was no vigorous decomposition of H_2O_2 by insoluble residue. Fig. 13 shows the concentrations of Mo, Zr, and Pu during sludge dissolution. No other element was detected significantly. The ratio of Mo/Zr was always about two, confirming the dissolution of $ZrMo_2O_7(OH)_2$. The final concentration of Pu is suggesting that 3.6 mol% of Zr in the sludge is replaced with Pu in accordance with the U/Pu solution tests. In this dissolution test, 68.2 mg of the adhering materials was dissolved. The rest, 70.7 mg, of the adhering materials remaining in the flask was heated again with fresh HNO_3–H_2O_2, but no further dissolution occurred. This part of the precipitation was thought to be insoluble residue.

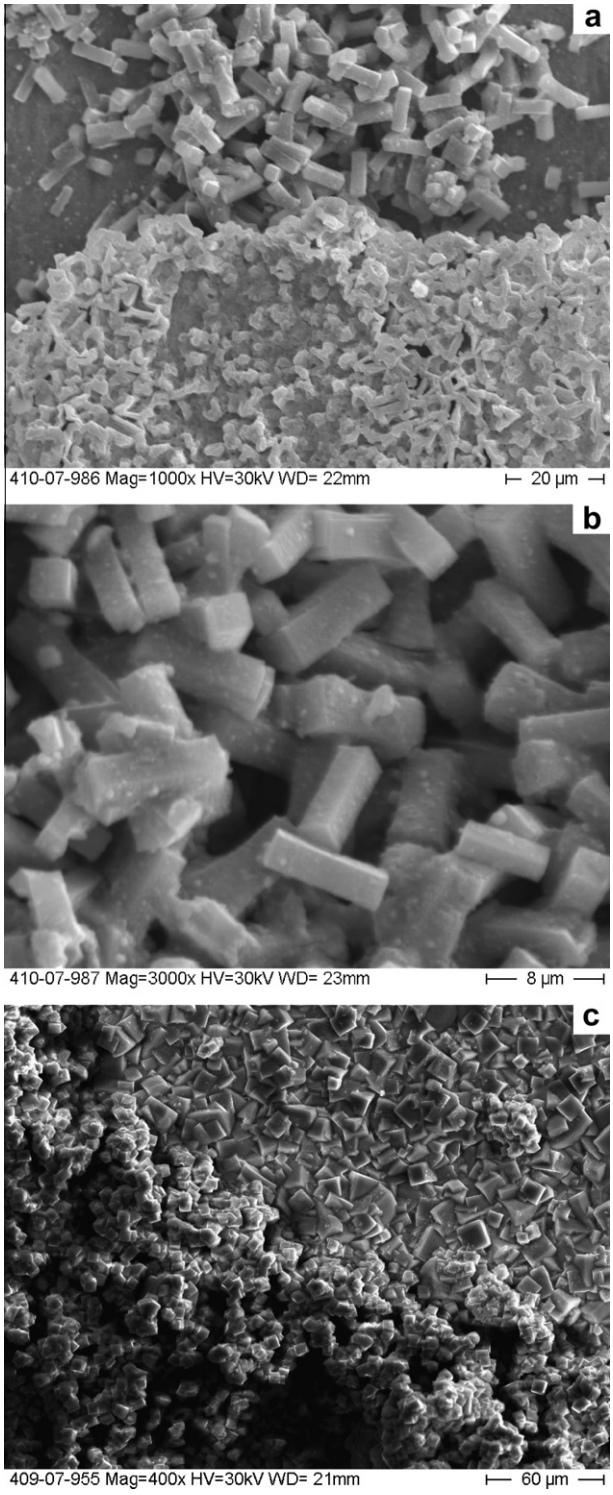

Fig. 12. SEM image of sludge generated from irradiated fuel (a, b: Zr-3, c: Zr-2).

The crystals adhered on the test piece Zr-4 was also completely dissolved by HNO_3–H_2O_2 within 3 h at 80 °C. The weight of the Zr piece after sludge dissolution was completely agreed with the original weight. In SEM analysis, no crystals were found on Zr-4 after dissolution.

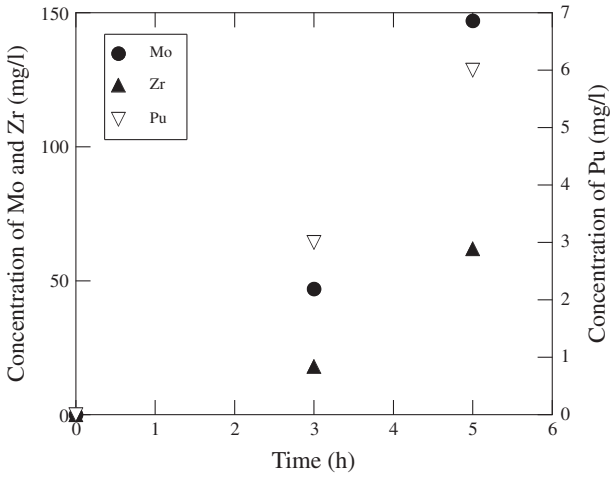

Fig. 13. Change in concentration with dissolution of sludge.

4. Conclusions

(i) The rate of sludge generation was measured in non-radioactive tests to establish suitable conditions for experiments using irradiated fuel.
(ii) The X-ray diffraction pattern of the precipitation completely agrees with that of $ZrMo_2O_7(OH)_2$. This sludge is completely dissolved by a 4 M HNO_3–2 wt.% H_2O_2 mixture in 6 h at 80 °C.
(iii) The sludge crystallization process on a clean Zr surface is inhibited. Therefore more frequent washing of the dissolver walls could lead to less adhesion of sludge and thereby improved efficiency in an industrial process.
(iv) In the sludge generated from U–Pu–Mo–Zr solutions, about 3 mol% of the Zr in $ZrMo_2O_7(OH)_2$ is replaced by Pu.
(v) Similar amounts of sludge are formed upon dissolution of irradiated PWR fuel and ~3.6 mol% of the Zr are substituted by Pu. The sludge could also in this case be dissolved in HNO_3–H_2O_2 solution at 80 °C within 5 h.

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