

Fuel Fabrication Capability (FFC) Scrap Recycle Report

Chemical Science and Engineering Division

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by

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FUEL FABRICATION CAPABILITY (FFC) SCRAP RECYCLE REPORT

1 INTRODUCTION

The Global Threat Reduction Initiative (GTRI) is currently engaged in the development of an ultra-high density low enriched uranium (LEU) fuel for its high-performance research reactors. The fuel consists of a monolithic uranium-molybdenum alloy containing 10% (weight/weight, or w/w) Mo (U-10Mo) enclosed in Al-6061 cladding with a Zr bonding layer. Significant losses are expected in the manufacturing of the LEU fuel, so a means to recycle uranium from the scrap pieces is needed. Uranium can also be recovered from irradiated U-10Mo fuels. As a result, Argonne National Laboratory (Argonne) is assisting by developing an aqueous processing flowsheet for scrap recovery in the fuel fabrication process and for spent fuel recovery. Experiments are being done to verify predicted dissolution conditions and optimize HNO₃ and HF concentrations for timely dissolution of the fuel without the formation of precipitates, while meeting the requirements needed for further processing by a tributyl phosphate solvent extraction procedure to recover uranium from a nitric acid-based media.

Dissolution conditions will be discussed first for the U-10Mo foils, followed by the U-10Mo foils with Al cladding, and finally the U-10Mo foils with Al cladding and a Zr bonding layer. The goal of the dissolution experiments is to test predicted conditions from earlier reports (Stepinski 2008a,b,c); these conditions were based on literature data and make the necessary adjustments to successfully dissolve the LEU fuels without the formation of precipitates. Final concentrations of all fuel components were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), and the error associated with the values is $\pm 10\%$.

2 U-10Mo DISSOLUTION RESULTS

U-10Mo foil dissolution experiments were initiated to confirm conditions predicted in the literature for the aqueous processing of ultra-high density LEU fuels. The U-10Mo foils received from Idaho National Laboratory (INL) for use in the dissolution experiments contain 89.98% depleted uranium (0.22% U-235) and 10.02% Mo. Small-scale experiments representing 20 g-U/L and 50 g-U/L samples were dissolved in solutions with varying HNO₃ concentrations. The 50 g-U/L samples were dissolved in the presence of 0.5-M Fe(NO₃)₃ to aid in the dissolution of larger amounts of Mo. A negatively charged Fe-Mo complex forms, preventing precipitation of uranyl molybdate (Schulz and Duke 1959; Schulz et al. 1956).

For the 20 g-U/L samples, an orange-red precipitate was observed under all dissolution conditions investigated. The amount of precipitate is too small to collect on the current scale, but it can easily be dissolved back into solution with a little shaking. For the samples that were not shaken, the red-orange precipitate re-dissolved into solution after about 7–8 days of sitting at room temperature. A similar precipitate has previously been observed in the synthesis of uranyl nitrate from UO₂ plates that do not contain Mo. It is possible that a uranium oxide is precipitating out of solution; uranium trioxide is known to form from uranyl nitrate solutions at elevated temperatures (Sheft et al. 1950). However, temperatures of 400°C and greater were not reached under our experimental conditions.

The samples containing 50 g-U/L did not form any precipitates. Ferric nitrate was effective in dissolving larger amounts of Mo in the presence of U without the precipitation of uranyl molybdate. In addition, Fe(NO₃)₃ increased dissolution rates by approximately three- to four-fold. Larger nitrate concentrations are expected to lower the solubility of Al(NO₃)₃ due to the common-ion effect, but this will be confirmed experimentally when the U-10Mo foils with Al cladding are dissolved (Stepinski et al. 2008c).

Conditions have been determined that are reproducible (triplicate) for the dissolution of U-10Mo foils representing a 20 g-U/L solution and a 50 g-U/L solution in the presence of 0.5-M Fe(NO₃)₃. An initial HNO₃ concentration of 3.4–3.5 M for the 20 g-U/L samples and 2.9–3.0 M for the 50 g-U/L samples was effective in dissolving the U-10Mo foils; average dissolution rates were 11 and 38 mg/min-cm², respectively. Final nitric acid concentrations of 3 M and 1.8 M are in the range needed for further processing. A previous report by Stepinski et al. suggests that a final HNO₃ concentration in the range of 2.2–5 M is needed for solutions containing 20 g-U/L, and that 1–4 M is needed for solutions containing 50 g-U/L (Stepinski et al. 2008c). The average Mo and U concentrations for the 20 g-U/L solutions suggest that the small amount of precipitate that formed re-dissolved because the final solutions contain 10.7% Mo and 89.3% U, which is similar to the original composition of the foils. U-10Mo dissolution results are shown in Table 1.

Several different equations have been cited in the literature for the formation of uranyl nitrate from U metal with molar ratios of HNO₃/U ranging from 2.8 to 6 (Pierce 2003). For the 20 and 50 g-U/L samples, an average molar ratio of 5.2 HNO₃/U was obtained, which is within the range reported in the literature. The Mo may also be consuming a small amount of HNO₃, which helps explain why the ratios are at the higher end of the reported range.

TABLE 1 Results for U-10Mo Fuel Dissolutions^a

Mass (g)	U (g)	Mo (g)	Fe (M)	Initial [HNO ₃] (M)	Final [HNO ₃] (M)	Dissolution Rate (mg/min-cm ²)	Precipitate	Final Mo (g/L)	Final U (g/L)
0.39	0.35	0.04	0	3.5	3.1	11	yes	2.7	22
0.44	0.4	0.044	0	3.4	3	13	yes	2.3	20
1.04	0.94	0.10	0	3.4	2.9	8.9	yes	2.2	19
0.52	0.47	0.050	0.5	3	1.8	29	no	6.3	57
1.14	1.03	0.110	0.5	2.9	1.8	41	no	5.8	47
1.15	1.03	0.120	0.5	2.9	1.8	44	no	6.1	51

^a Final Mo and U concentrations are $\pm 10\%$.

Fagueras et al. (1961) and Ferris (1961a) studied the solubility of Mo in the presence of uranyl nitrate, ferric nitrate, and many other metal salts. Based on their data, U-10Mo foils up to 25 g-U/L can be dissolved in ≥ 3 -M HNO₃ without precipitation; in the presence of 0.5- to 1-M Fe(NO₃)₃, up to 90 g-U/L can be dissolved in 2-M HNO₃ without precipitation (Fagueras et al. 1961; Ferris 1961a). Solutions containing approximately 20 g-U/L formed a red-orange precipitate when the initial HNO₃ was between 3 and 6 M. This contradicts what has been reported and/or predicted based on previous literature reports (Stepinski et al. 2008c). However, the precipitate can easily be re-dissolved with a little shaking or time. A larger piece of U-10Mo fuel (1.7 g) was dissolved in an attempt to obtain a more significant amount of precipitate; however, the amount of precipitate remained too small to collect. For samples containing 50 g-U/L in the presence of 0.5-M Fe(NO₃)₃, no precipitation was observed for the entire range of HNO₃ concentrations investigated (2–6 M). The results that we obtained for the 50 g-U/L samples with 0.5-M Fe(NO₃)₃ agree with what has been predicted.

Previous studies have shown that dissolution rates for U-10Mo foils are about 100 mg/min-cm² in the presence of 6-M HNO₃ and 2 mg/min-cm² in the presence of 2-M HNO₃ (Schulz and Duke 1959; Ferris 1961b). The dissolution rates that we observed are within the range expected for initial HNO₃ concentrations of 3.4–3.5 M for the 20 g-U/L solutions and 2.9–3.0 M for the 50 g-U/L solutions in the presence of 0.5-M Fe(NO₃)₃. The dissolution rates are 11 mg/min-cm² for the 20 g-U/L samples and 38 mg/min-cm² for the 50 g-U/L samples.

3 U-10Mo WITH Al CLADDING DISSOLUTION RESULTS

Each of the five U.S. high-performance research reactors requires slightly different ratios of Al cladding to U for the U-10Mo foils. For example, the Missouri University Research Reactor (MURR) requires U-10Mo foils with Al/U molar ratios of 1.9–7.7, while the Advanced Test Reactor (ATR) at Idaho National Laboratory needs foils containing Al/U molar ratios of 4.9–24.8 (Stepinski et al. 2008a). In a previous literature report by Stepinski et al. (2008a), the average molar ratio of Al/U based on the number of plates required each year, the plate geometries, and the amount of Al cladding required for the U-10Mo fuels for the National Institute of Standards Reactor (NIST), MURR, and ATR was utilized to predict dissolution conditions. That value assumes a molar ratio of 4.9 Al/U (Stepinski et al. 2008a); however, the U-10Mo foils with Al cladding that were received from Idaho National Laboratory contain molar ratios of 18.8 and 111 Al/U. Small-scale experiments are difficult to complete with these types of foils because 67% and 92% of foil mass is Al cladding. Large foils would need to be dissolved in small volumes to obtain U concentrations of 20 or 50 g-U/L. In addition, the primary goal of the dissolution experiments was to test predicted conditions, which were based on the fuel having a molar ratio of 4.9 Al/U.

There are two processes suggested for the dissolution of Al cladding. A two-step procedure uses a caustic solution to dissolve the Al cladding, while a single-step method utilizes a catalyst such as HF to penetrate the aluminum oxide layer and dissolve the cladding (Stepinski et al. 2008a). Results discussed in this report relied solely on the addition of HF to facilitate the single-step dissolution. In an effort to test predicted dissolution conditions, a piece of Al cladding was dissolved first, and the remaining amount of U-10Mo fuel meat needed to obtain a 4.9 molar ratio of Al/U was added to the reaction mixture. Again, the purpose of these dissolution experiments was to test predicted conditions and analyze dissolution rates, consumption of HNO_3 , and formation of precipitates.

First, dissolution conditions were determined for the Al cladding by itself in HNO_3 and HF solutions with and without 0.5-M $\text{Fe}(\text{NO}_3)_3$. The concentration of Al cladding represented what would be required to obtain a 4.9 molar ratio of Al/U for a sample containing 20 g-U/L and 50 g-U/L. It has been suggested that a 1.3 molar ratio of Al/HF would enable timely dissolution of the cladding in a single step (Stepinski et al. 2008c; Jerden et al. 2009). The Al cladding (~0.45 M) was successfully dissolved in a 3.4-M HNO_3 solution containing 0.34-M HF with a dissolution rate of 5.6 mg/min-cm², and no precipitation was observed. However, half of the nitric acid was consumed, so a higher initial nitric acid concentration will be needed to dissolve the U-10Mo fuel with Al cladding and still have a final H^+ concentration in the range of 2.2–5 M, as needed for further processing. Dissolution conditions for the Al cladding were also determined in the presence of 0.5-M $\text{Fe}(\text{NO}_3)_3$. The Al cladding (~1.1 M) dissolved in a solution containing 5.8-M HNO_3 , 0.85-M HF, and 0.5-M $\text{Fe}(\text{NO}_3)_3$, with a dissolution rate of 4.3 mg/min-cm². No precipitation was observed. Results for the dissolutions of the Al cladding are given in Table 2.

Three different sets of dissolution experiments were initiated for samples containing final Al/U molar ratios of 4.9, 5.9, and 7.6. For all dissolutions, a piece of Al cladding was dissolved

TABLE 2 Results for Al Cladding Dissolutions^a

Al (g)	Fe (M)	Cladding Size (cm ²)	Cladding Dissolution Rate (mg/min-cm ²)	Initial HNO ₃ (M)	Final H ⁺ (M)	Initial HF (M)	Final Al (g/L)	Final Fe (g/L)
0.18	0	1.1	5.6	3.4	1.7	0.6	13	0
0.28	0.5	1.4	4.3	5.8	1.3	0.85	29	28

^a Final Al and Fe concentrations are $\pm 10\%$.

first, and a U-10Mo foil was added immediately after the cladding dissolved. A small amount of a red-orange precipitate formed as the solution cooled to room temperature. Again, the amount of precipitate was too small to collect on the current scale, but the precipitates re-dissolved within one week. Concentrations of 3.4- and 5.8-M HNO₃ were tested, along with 0.6- and 1-M HF concentrations, and the cladding and fuel meat dissolution rates increased in the presence of larger amounts of HF. Dissolution conditions were examined using higher concentrations of HF because more HF will be required when Zr is present, and the solubility of Al, U, and Mo in the presence of higher HF concentrations has not been thoroughly investigated. Increasing the HF concentration from 0.6- to 1-M HF did not lead to the formation of larger amounts of precipitates. Again, the red-orange precipitate formed under all three dissolution conditions and was no longer present after a week. The final H⁺ concentration for the last two dissolutions is in the range needed for further processing (2.2–5 M) (Stepinksi et al. 2008c). The results for the dissolution experiments with Al cladding and U-10Mo fuel meat added for solutions containing approximately 20 g-U/L are shown in Table 3.

TABLE 3 Results for Al Cladding with U-10Mo Fuel Meat Dissolutions with Al/U Molar Ratios of 4.9–7.6^a

U (g)	Mo (g)	Al (g)	Fuel Meat Dissolution Rate (mg/min- cm ²)	Cladding Dissolution Rate (mg/min- cm ²)	Initial HNO ₃ (M)	Final H ⁺ (M)	Initial HF (M)	Final U (g/L)	Final Mo (g/L)	Final Al (g/L)	Al/U Molar Ratio
0.29	0.029	0.17	23	5.8	3.4	1.4	0.6	19	2.2	13	5.9
0.34	0.034	0.18	25	2.4	5.8	3	0.6	21	2.5	12	4.9
0.25	0.025	0.19	40	12	5.8	2.8	1	22	2.4	19	7.6

^a Final U, Mo, and Al concentrations are $\pm 10\%$.

Three sets of dissolution experiments were investigated for samples with Al/U molar ratios ranging from 5.1 to 5.5. In the first dissolution experiment, 5.8-M HNO₃ and 1.4-M HF were used with 0.5-M Fe(NO₃)₃. The cladding dissolved, but after the fuel meat was added, a large amount of red precipitate formed. The U-10Mo foil did not dissolve completely, but the precipitate was collected. ICP-MS results indicate that the sample is mostly Fe (80%) and Mo (12%), with smaller amounts of Al (6%) and U (2%). The higher HF concentration does not work well for dissolving the cladding and fuel meat in the presence of large amounts of Fe. In a second dissolution attempt, after about 4 hr, the Al cladding had not completely dissolved, but the fuel meat was added to the reaction and dissolved within 1 hr. The Al cladding had still not dissolved but remained in solution at room temperature over the weekend. It dissolved over the weekend, but the solution was a dark orange-red color (all other solutions were yellow). The initial HNO₃ and HF concentrations were 5.8 M and 0.85 M. No precipitates were observed, but ICP-MS results indicate slightly higher metal ion concentrations than originally anticipated: U = 61 g/L; Mo = 6.7 g/L; Al = 38 g/L; and Fe = 31 g/L. In addition, the final H⁺ concentration was extremely low (~0.1 M), well below the value needed for further processing (1–4 M) (Stepinski et al. 2008c). In a final dissolution attempt, the Al cladding dissolved with a dissolution rate of 4 mg/min-cm², while the fuel meat dissolved much more rapidly, at a rate of 75 mg/min-cm². A slightly lower HF concentration was used (0.77 M), and the final H⁺ concentration of 1.1 M was in the necessary range for the tri-butyl phosphate (TBP) extraction process. The solution was yellow, and no precipitates were observed. ICP-MS values are closer to concentrations expected: U = 52 g/L; Mo = 6.1 g/L; Al = 30 g/L; and Fe = 27 g/L. The results are shown in Table 4.

TABLE 4 Results for Al Cladding with U-10Mo Fuel Meat Dissolutions with Al/U Molar Ratios of 5.1–5.5^a

U (g)	Mo (g)	Al (g)	Initial HNO ₃ (M)	Final H ⁺ (M)	Initial HF (M)	Final U (g/L)	Final Mo (g/L)	Final Al (g/L)	Final Fe (g/L)	Al/U Molar Ratio
0.51	0.051	0.29	5.8	–	1.4	–	–	–	–	–
0.54	0.054	0.30	5.8	0.1	0.85	61	6.7	38	31	5.5
0.47	0.047	0.27	5.8	1.1	0.77	52	6.1	30	27	5.1

^a Final U, Mo, Al, and Fe concentrations are ±10%.

4 U-10Mo WITH Al CLADDING AND Zr BONDING LAYER DISSOLUTION RESULTS

The main concern with dissolving the U-10Mo foils with Al cladding and a Zr bonding interlayer is the formation of an intermetallic U-Zr₂ complex, which is highly explosive (Jerden et al. 2009). Previous studies have shown that the high surface area of the intermetallic phase develops from an unevenly distributed dissolution pattern, and it can be oxidized with explosive potential if its activation energy barrier is overcome (Gens 1958; Swanson 1959; Jackson and Johns 1970). HF can be added to the reaction mixture to prevent this from occurring; however, HF is also required to aid in the dissolution of the Al cladding. A 4:1 HF/Zr molar ratio has been shown to prevent the formation of this explosive reaction (Stepinski et al. 2008c; Jerden et al. 2009). The amount of HF that needs to be added is in addition to the amount required to dissolve the Al cladding.

The final small-scale dissolution experiments were initiated for the U-10Mo foils containing Al cladding and a Zr bonding interlayer. Again, a piece of Al cladding was dissolved first, followed by a U-10Mo foil containing a Zr bonding layer. The U-10Mo foils with Zr obtained from INL contain 83.3% U, 9.3% Mo, and 7.4% Zr by weight. For the first set of experiments for the samples containing 20 g-U/L and 50 g-U/L, the Al cladding was dissolved in the presence of 5.8-M HNO₃ and HF (1.3 molar ratio Al/HF). After the cladding was dissolved, the fuel meat with Zr was added with the correct amount of HF (4:1 HF/Zr) to fully complex the Zr. In second set of experiments, the amount of HF required for dissolving the cladding and Zr was added at the beginning, before the Al cladding had dissolved.

The initial conditions for the dissolutions of samples containing approximately 20 g-U/L and 50 g-U/L are given in Table 5. The first 20 g-U/L sample dissolved fairly quickly with an Al cladding dissolution rate of 9.2 mg/min-cm² and fuel meat dissolution rate of 32 mg/min-cm². However, the small amount of orange-red precipitate that was originally observed re-dissolved within a week. The amount of total HF added was in slight excess of the stoichiometric amounts required for a 1.3 molar ratio of Al/HF and 4:1 molar ratio of HF/Zr. Still, the final H⁺ concentration is in the range needed for further processing. No precipitates were observed for the next 20 g-U/L sample, but dissolution rates were slower (5.1 and 11 mg/min-cm² for cladding and fuel meat) because HF was no longer in excess. The final acid concentration is higher than what would have been expected because less HF was used than in the previous dissolution, and its final H⁺ concentration is 4.7 M compared to 3.9 M obtained in the initial dissolution. Unfortunately, the samples containing approximately 50 g-U/L did not perform quite as well. Precipitation was observed in both dissolution experiments, and ICP-MS results on the identity of the white precipitate are pending. In the second dissolution, the fuel meat would not dissolve completely after 4 hr of refluxing. The amount of Fe(NO₃)₃ and the larger amounts of HF required make it difficult to find dissolution conditions that keep all metal ions in solution (U, Mo, Al, Zr, and Fe). As a result, more work needs to be done to find optimum conditions for dissolution of U-10Mo with Al cladding and Zr in the presence of 0.5-M Fe(NO₃)₃. The final concentrations of all metal ions are shown in Table 6.

TABLE 5 Initial Conditions for Al Cladding and U-10Mo with Zr Fuel Meat Dissolutions

U (g)	Mo (g)	Zr (g)	Al (g)	Fe (M)	Fuel Meat Dissolution Rate (mg/min-cm ²)	Cladding Dissolution Rate (mg/min- cm ²)	Initial HNO ₃ (M)	Initial HF (M)	Final H ⁺ (M)	Precipitate
0.25	0.025	0.022	0.15	0	32	9.2	5.8	0.7	3.9	Yes
0.44	0.044	0.039	0.25	0.5	6.2	3.5	5.8	1.1	0.7	Yes
0.25	0.025	0.022	0.15	0	11	5.1	5.8	0.42	4.7	No
0.43	0.043	.049	0.26	0.5	–	3.2	5.8	0.85	–	Yes

TABLE 6 Results for U-10Mo with Al Cladding and Zr^a

Final U (g/L)	Final Mo (g/L)	Final Al (g/L)	Final Fe (g/L)	Final Zr (g/L)	Final Al/U Molar Ratio
21	2.5	12	0	2.1	4.9
55	6.1	31	27	5.0	5.0
20	2.3	12	0	1.9	5.3
–	–	–	–	–	–

^a Final U, Mo, Al, Zr, and Fe concentrations are ±10%.

5 CONCLUSIONS

Conditions have been optimized for the U-10Mo foils with the exception of the red-orange precipitate that forms and re-dissolves in the 20 g-U/L samples. Dissolving a larger U-10Mo foil did not produce a larger amount of precipitate. The amount of precipitate was too small to collect.

Conditions have not been optimized for the U-10Mo foils with Al cladding and Zr representing 50 g-U/L samples in the presence of 0.5-M $\text{Fe}(\text{NO}_3)_3$. More conditions need to be tested to find the proper HF and HNO_3 concentrations for timely dissolution without precipitation. If precipitation continues to persist, the two-step dissolution route may be necessary.

A more thorough understanding of the HNO_3 consumption when Al cladding and/or Zr are present needs to be obtained. More experiments need to be completed with U-10Mo foils with Al cladding with and without Zr to obtain consistent, reproducible results for HNO_3 consumption.

Future dissolution experiments will test conditions using varying molar ratios of Al/HF because different results would be expected depending on the exact ratio. Dissolving the Al cladding takes a significant amount of time, and foils with Al/U molar ratios of 24.8 would take a much longer amount of time to dissolve compared to foils with Al/U molar ratios of 1.9. For samples containing larger Al/U ratios, the two-step dissolution process may be more appropriate.

Once conditions for all types of foils have been thoroughly investigated and optimized, a scale-up process will follow. This would entail dissolving U-10Mo foils that are approximately 20 g-U and 50 g-U, because 1 L of solution would be required for the feed solution for the TBP-extraction process.

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