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PRODUCTION OF DEPLETED U-10Wt% MO FOILS

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ABSTRACT

Research reactors and medical isotope producers can utilize Low Enriched Uranium (LEU) metallic fuel alloyed with ten weight percent (wt%) molybdenum (U-10Mo) in lieu of highly enriched uranium (HEU) fuels and have conducted feasibility studies confirming the utility of such fuel. The challenge has been to produce suitable fuel from the U-10Mo alloy, in the form of metallic foil. Manufacturing Sciences Corporation (MSC) an *EnergySolutions* Company, under contract with Battelle Energy Alliance undertook a full scale proof of principal foil making process, using depleted uranium, alloyed with 10wt% Mo as a surrogate. Two 305 kilogram (kg) castings were vacuum induction cast, rolled, and made into plate and foil, ranging in thickness from 4 millimeter (mm) to 0.25mm in sizes representing widths and lengths required for the fabrication of full sized fuel elements.

1.0 Introduction

The process for making foil in the U-10Mo system is industrially immature on the scale desired to meet the LEU metallic fuel demands. A contract between MSC and Battelle Energy Alliance provided the funding to produce full sized foils using the surrogate material, depleted uranium (DU), as a substitute for LEU. The processes used, and the manufacturing parameters associated with the processes, are presented here and are intended to help guide USDOE in assessing whether U-10Mo alloy sheet can be reliably produced on a scale needed to meet the needs of this application.

2.0 Experimental Work

The depleted uranium plate and sheet industry is mature, in that millions of kilograms (kg) of DU have been made into plate and sheet for applications including nuclear shielding, ballistic

armor, and physics calorimeters. DU, LEU, and HEU are metallurgically identical, and as such it is prudent to make process discoveries using the surrogate DU. The equipment and infrastructure at *EnergySolutions*' MSC Oak Ridge TN facility, was utilized to produce DU-10Mo foils to take advantage of DU casting, rolling, fabrication equipment and knowledge resident at the facility.

2.1 Vacuum Induction Casting

The vacuum induction melting (VIM) furnace was a double walled, water cooled, steel shell, pumped with twin Stokes 412 MB rotary vane pumps each with a 615 roots blower. Induction power came from an Ajax 300 kilowatt- 1000 hertz solid state power supply delivering electrical energy to a water-cooled copper coil, 1050 (mm) in diameter by 900 mm tall. The mold chamber below the melt chamber was a double walled water cooled steel shell equipped with a graphite felt insulated 3-zone heating package capable of delivering 350 kilowatts of resistance heating power to molds placed within the furnace. Vacuum levels in both chambers were maintained below 250 millitorr during the melt.

The alloyed metals were vacuum induction melted and cast using a cylindrical graphite crucible shown schematically in Figure 1. The crucible was a "step" design where the reduced section contained the alloy agent (Mo) and the expanded section contained the uranium.

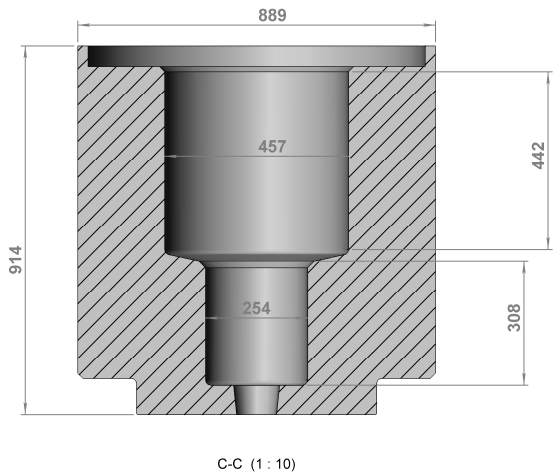

Figure 1: Schematic cross section of the stepped, graphite melting crucible
(note: all dimensions in mm)

Prior to charging the crucible with metal, a two layer titanium carbide (TiC) - yttrium oxide (Y_2O_3) coating was applied to a warm graphite substrate to prevent graphite / uranium interaction. The crucible was heated to approximately 125°C ,and spray coated with TiC using a high volume low pressure (HVLP) spray gun. The TiC was then over-coated with Y_2O_3, again using HVLP techniques. [The two coat system was designed to prevent the Y_2O_3 from reducing to deleterious yttrium carbide (YC) when temperatures exceed 1550 °C during melting.]

Seventy five kilograms of Molybdenum in the form of 6 millimeter (mm) irregular lump (99.9% pure) was placed in the reduced section of the crucible. An overcharge of 1 wt% Mo was

included to account for flotation losses during melting. Six hundred kilograms of depleted uranium scrap metal was placed in the enlarged section.

The loaded crucible was placed in the induction furnace and the dual cavity book molds installed. The furnace was sealed and the vacuum induction melting sequence commenced.

The peak crucible temperature reached was 1550°C and the final metal temperature prior to casting was 1450°C. The casting run yielded two ingots with approximate dimensions of 40 mm by 523mm by 806 mm each weighing approximately 305 kg. The ingots were stamped with the heat identity of 805A and 805B respectively

Bulk compositional samples were removed from ingot 805A using a band saw. The samples were tested using inductively coupled plasma (ICP), and found to be 9.6 wt% Mo.

2.2 Rolling

Rolling was accomplished on a 4 high reversing mill with 900 mm diameter back-up rolls and 460 mm diameter work rolls with a 1 meter face width. Servo hydraulics screw downs capable of 22,240 kilo-newton (kN) of separating force controlled the mill opening.

Ingot 805A was heated to 650°C in a molten carbonate salt bath, using methods typically employed for depleted uranium and its alloys. The first rolling pass was 5% true strain; the second rolling pass was also 5% true strain. During the second pass cracks appeared at the edges of the ingot, perpendicular to the rolling direction and propagated approximately 50mmm to the interior before changing directions and continuing parallel to the rolling direction, resulting in edge fragmentation A broken piece from that ingot was harvested and rolled from a preheat temperature of 670°C, and failed in a similar way. Salt bath rolling was abandoned and replaced with rolling in a 304 stainless steel can of the design depicted in Figure 2.

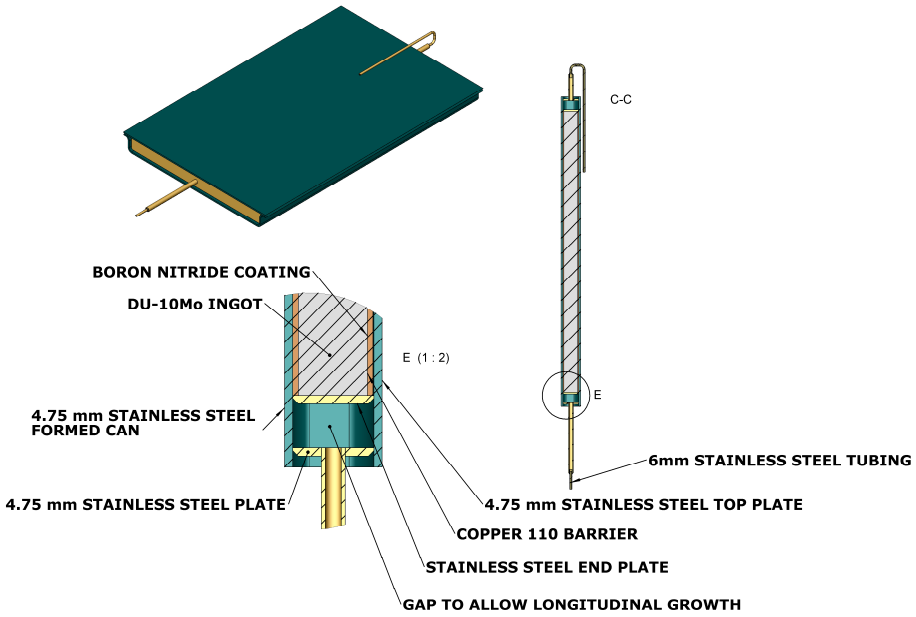

Figure 2: Schematic cross section of the stainless steel can design used to roll the DU-10Mo alloy from 40 mm to 4 .2mm

Hot rolling of ingot 805B was accomplished by canning the ingot in a 304 stainless steel outer skin (can), 4.75 mm thick with a 3 mm thick copper interlayer between the uranium and the steel to prevent the uranium-iron eutectic reaction from occurring, when rolling temperatures exceeded 725°C. To facilitate copper removal after rolling, boron nitride (BN) was used as a parting agent between the copper and uranium. At each end of the can a labyrinth arrangement was constructed. Both labyrinths had 6 mm diameter tubing welded to them, to accommodate an argon gas supply. During preheating, one liter per minute of argon was allowed to flow through the 6 mm tubing, through the uranium filled cavity, exiting via the second 6mm tube.

A preheat temperature of 760°C was applied using an electric resistance heated box furnace. A type K thermocouple attached directly to the stainless steel can, indicated when the rolling temperature was reached. A 2 hour minimum soak at 760°C insured the DU-10Mo alloy was at the desired rolling temperature.

Once the ingot / can reached rolling temperature, it was rolled according to the schedule shown in Table 1. All rolling occurred parallel to the long axis of the casting.

Table1: Rolling schedule to reduce the U10-Mo alloy core from 40mm to 22mm

Material idenity	Rolling direction	Prepass length (mm)	Prepass width (mm)	prepass temp °C	thickness used to generate pass schedule	desired true strain per pass	estimated seperating force (kN)	Mill set	calculated post pass thickness (mm)
805B	sr	897.0	533.0	760°c	55.88	0.05	2222	48.82	49.13
					53.01	0.05	1555	47.38	48.05
					50.82	0.1	3110	39.91	41.20
					46.72	0.1	6442	29.71	31.40
					42.72	0.15	8442	20.30	21.24
					35.93	0.15	8442	14.46	15.39

After the fifth pass the stainless steel can broke open at a transverse weld joint on the leading edge of the can. Rolling continued through pass 6, resulting in a rolled DU-10Mo plate, 22 mm thick by 1400 mm long by 500 mm wide . Immediately after rolling the DU-10Mo / can combination was water spray cooled to minimize oxidation during cooling.

Once cooled, can removal and size reduction by shearing was attempted, taking advantage of MSC's 30 mm plate shear. The results were poor. The DU-10Mo alloy cracked leaving fragments and deep cracks along the sheared edge that propagated into the bulk plate. Band sawing proved efficient and sectioning of the rolled plate by sawing, avoided the shear induced cracks and yielded several pieces 200mm long by 260mm wide. The 200 mm by 260 mm pieces were canned in a smaller version of the initial can; preheated to 760°C; and rolled parallel to the long axis of the cast ingot using the rolling schedule shown in Table 2.

Table 2: Rolling schedule to reduce the U10-Mo alloy core from 22mm to 12.1 mm

Material idenity	Rolling direction	Prepass length (mm)	Prepass width (mm)	prepass temp ℃	thickness used to generate pass schedule	desired true strain per pass	estimated seperating force (kN)	Mill set	calculated post pass thickness (mm)
805B	sr	300.0	250.0	760℃	37.80	0.05	2222	31.61	31.93
					37.16	0.05	1555	32.31	32.98
					32.39	0.1	3110	23.23	24.52
					30.48	0.1	6442	15.01	16.70
					28.24	0.15	8442	7.84	8.78
					25.30	0.15	8442	5.31	6.24

The can again broke open during the 5th pass. Rolling continued through pass 6. Water spray cooling was again employed at the completion of the rolling cycle to minimize alloy oxidation. Can removal via band sawing yielded a rolled slab 12.1 mm thick by 360mm long by 260 mm wide.

The 12.1 mm thick rolled plate was further sectioned into 2 pieces approximately 260mm wide by 175 mm long. Each sectioned piece was canned in the usual way; preheated again to 760 °C; and rolled parallel to the casting direction by the schedule shown in Table 3. Water quenching after rolling was again performed. After band saw sectioning and edge trimming, alloy pieces approximately 6.5 mm thick by 260mm wide by 250 mm long by were recovered for further processing.

Table3: Rolling schedule to reduce the U10-Mo alloy core from 12.1mm to 6.5 mm

Material idenity	Rolling direction	Prepass length (mm)	Prepass width (mm)	prepass temp ℃	thickness used to generate pass schedule	desired true strain per pass	estimated seperating force (kN)	Mill set	calculated post pass thickness (mm)
805B	sr	275.0	250.0	760℃	27.31	0.05	2222	21.63	21.65
					25.82	0.05	1555	21.53	21.54
					24.47	0.1	2222	17.80	17.83
					22.34	0.1	3110	14.14	14.17
					20.26	0.15	3332	10.93	10.98
					17.83	0.15	5110	5.38	5.44

Canning in the usual way and subsequent rolling by the schedule in Table 4 yielded a plate 4.2 mm thick plate by 380 mm long by 250 mm wide. The 4.2 mm thick material was considered the “master” plate, and became the source material to make the varying thickness required to demonstrate the feasibility of foil making.

Table4: Rolling schedule to reduce the U10-Mo alloy core from 6.5mm to 4.2 mm

Material idenity	Rolling direction	Prepass length (mm)	Prepass width (mm)	prepass temp ℃	thickness used to generate pass schedule	desired true strain per pass	estimated seperating force (kN)	Mill set	calculated post pass thickness (mm)
805B	sr	340.0	250.0	760℃	19.05	0.05	2222	17.448	17.82
					17.82	0.05	889	16.579	17.00
					17.00	0.14	2222	14.109	14.83
					14.83	0.14	2444	12.172	12.89

The can design was changed to a "picture frame" style, using low carbon steel. Yttrium oxide replaced BN as a parting agent; the copper was eliminated as an inner layer; as was the argon cover gas. A schematic of the can design is depicted in Figure 3.

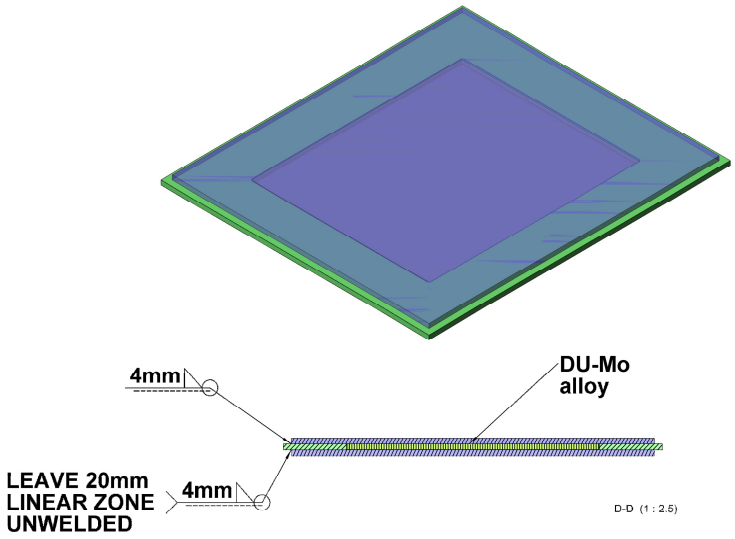

Figure 3: Schematic depiction of the carbon steel "picture frame" style can.

A 4.75 mm carbon steel plate (A36) was cut into a frame with inside dimensions approximately 252 mm by 307 mm to accommodate a 4.2 mm thick by 250 mm wide by 305 mm long DU-10Mo alloy plate. The external dimensions of the frame were 350 mm by 405 mm, making the frame width approximately 50mm. The 4.2mm thick DU-10Mo alloy plate was placed inside the frame and two 4.75 mm thick carbon steel "skins" were fillet welded to enclose the DU-10Mo alloy plate. A 20 mm portion was left unwelded to provide a vent for air to escape during the heating process. The can was heated to 760°C and rolled for two passes according to the schedule in Table 5.

Table 5: Rolling schedule to reduce the U10-Mo alloy core from 4.2 mm to 1mm

Material idenity	Rolling direction	Prepass length (mm)	Prepass width (mm)	prepass temp ℃	thickness used to generate pass schedule	desired true strain per pass	estimated seperating force (kN)	Mill set	calculated post pass thickness (mm)
805B	sr	300	340	760	14.83	0.14	2444	12.17	13.49
					13.49	0.14	5110	10.41	11.58
		397	340	760	11.58	0.14	4443	8.90	10.11
					10.11	0.14	4665	7.58	8.89
		525	340	760	8.89	0.14	5110	6.41	7.78
					7.780	0.14	4668	5.397	6.76
		695	340	760	6.764	0.14	5332	4.514	5.88
					5.880	0.14	5332	3.746	5.11
		919	340	760	5.112	0.14	5332	3.078	4.44
		1060	340	760	4.444	0.14	5332	2.497	3.86

After two passes reheating was required to maintain sufficient heat in the can to continue rolling. The regiment of two passes followed by reheating continued until the rolling can was approximately 5mm thick. Reheating after every pass was required after the overall can thickness was below 5mm. Rolling was stopped due to furnace length limitations, after completion of the 2.497 mm pass. The DU- 10Mo alloy was removed from the can by band sawing and resulted in a1mm thick, 1238mm long by 248 mm wide DU-10Mo plate

Due to preheat furnace length limitations it was deemed appropriate to reduce the carbon steel can frame width from 50 mm to 25mm, and subsequent rolling cans were constructed accordingly, with no deleterious effects.

The 1 mm DU-10Mo alloy plate was sectioned into smaller plates, recanned and rerolled using 1.5mm thick carbon steel (A36) as can construction metal. The design in Figure 3 was again used leaving a 20 mm unwelded vent to allow air to escape during heating. The rolling schedule in Table 6 below, was used to produce 0.5 mm foil, 0.381 mm foil, and 0.254 mm foil in a common finished width of 240 mm with lengths varying from 700 mm to 800 mm

Table 6: Rolling schedules to produce DU-10Mo alloy foils 0.5mm, 0.38 mm and 0.25mm thick.

Material idenity	Rolling direction	Prepass length (mm)	Prepass width (mm)	prepass temp ℃	thickness used to generate pass schedule	desired true strain per pass	estimated seperating force (kN)	Mill set	calculated post pass thickness (mm)	Du-Mo alloy thickness (mm)
805B	sr	varied	290	760	4.444	0.14	5332	2.497	3.864	1
		varied	290	760	3.864	0.14	5332	1.992	3.359	
		varied	290	760	3.359	0.14	5332	1.554	2.920	
		varied	290	760	2.920	0.14	5332	1.172	2.539	
		varied	290	760	2.539	0.14	5332	0.840	2.207	0.508
		varied	290	760	2.207	0.14	5332	0.552	1.919	
		varied	290	760	1.919	0.14	5332	0.301	1.668	
		varied	290	760	1.668	0.14	5332	0.084	1.450	
		varied	290	760	NA	NA	ND	0.076	ND	0.381
		varied	290	760	NA	NA	ND	0.076	ND	
		varried	290	760	NA	NA	ND	0.076	ND	0.254

3.0 Results

The melting and casting of DU- 10Mo alloys that was undertaken delivered expected results, while rolling of the alloy required process development to find a working method. Key features of melting, casting and rolling are discussed to give the reader some insight to the challenges of a production LEU-10 Mo alloy, fabrication plant.

3.1 Vacuum Induction Casting

The vacuum induction casting work was unremarkable in that what was planned met expectations. Some of the important features may assist the reader in duplication of the work at other institutions.

Vacuum induction melting in a graphite crucible is an indirect melting technique in that the crucible is heated by the induction field; radiant heat transfer and conduction from the crucible melt the crucible contents. In the case of the DU-10Mo alloy the indirect heating, melted the uranium closest to the wall of the enlarged section of the crucible first. This “early” liquid uranium was allowed to drip onto, and react with, the molybdenum lump in the reduced section of the crucible. As melting continued the still solid mass of uranium above the reacting mass in the reduced section, prevented the molybdenum from floating. This geometry allowed sufficient molybdenum / uranium reaction time to get dissolution and produce an alloy of 9.6 wt% molybdenum (87% molybdenum recovery)

The coating strategy of TiC overcoated with Y_2O_3 protected the crucible from a deleterious carbon reaction as witnessed by easy removal of crucible residues following casting.

The 1450 °C casting temperature proved to be sufficient metal superheat to distribute molten metal simultaneously into two mold cavities with approximately equal flow rates, resulting in two ingots whose weights were within 5 Kg of each other.

The cast metal surface finish was smooth permitting direct rolling without machining a fresh surface.

3.2 Rolling

Insufficient preheating temperature was determined to be the failure mechanism of attempts to roll the DU-10Mo alloy from carbonate salt. Successful rolling was accomplished using two canning methods and a preheating temperature of 760° C achieved in an air atmosphere furnace. Rolling from 40mm to 4 mm used a stainless steel can, and rolling from 4mm to 0.25mm utilized a carbon steel "picture frame" type can.

The stainless steel can work hardened during deformation at 760°C and that limitation forced rolling to cease after 60% true strain. The DU-10Mo alloy in the core of the can had ductility beyond that of the stainless steel. Once the rolling can was removed, the major faces of the rolled DU-Mo10 plate had a smooth surface finish. Areas along the edges of the alloy plate in the failure zone of the stainless steel can had embossed features associated with can failure. Those areas had to be excised by sawing to prevent stress concentration areas that would propagate cracks during subsequent rolling cycles.

The sophistication of the stainless steel can for initial rolling, as revealed in later stages of rolling, was unwarranted, and future work would be well served to use low carbon steel (A36), to avoid the work hardening limitation. A can design similar to that used for 4 mm plate should be adopted for 40 mm castings to decrease the number of canning cycles and increase the amount of true strain prior to reheating. The details of that improvement recommendation would require experimentation. The copper interlayer and the BN coating were unnecessary. Yttrium oxide coating of the DU-10Mo, coupled with the uranium oxide layer already present on the alloy, was determined to be a sufficient barrier to avoid the eutectic reaction that was initially feared. The need for inert gas in the can during preheating requires validation. Rolling below 4 mm was successful without inert gas

The carbon steel "picture frame" style can used to roll 4.2 mm alloy plate to 0.25 mm closely matched the strength and deformation characteristics of the DU-10Mo alloy, allowing rolling to occur from a 4.2 mm thick to 0.25mm without destruction of the protective can.

Heating every other pass when overall can thicknesses were between 15mm and 5mm was required to counteract the chill imparted by the rolling mill rolls. Can thicknesses below 5mm required re-heating after every pass to maintain sufficient rolling temperature.

During the later stages of rolling where can thickness were below 2mm it was found that the can had to be fed into the rolling mill with the same leading edge to maintain flatness. If subsequent passes used the trailing edge of the previous pass, wrinkling occurred. The observed phenomenon is unique to the experiences of the MSC team.

Final rolled foils from carbon steel "picture frame" style cans had predictable gage control based on micrometer measurements of the overall can after each rolling pass. The carbon steel can remained intact during rolling, was easily removed after rolling, and resultant foils were flat, uniform in surface finish, and uniform in final thickness.

4. Conclusion

Vacuum Induction melting and casting of DU-10wt% Mo is easily accomplished with existing furnaces, using mature developed technology for uranium casting.

Rolling of DU-10wt% Mo in a stainless steel can from 40 mm to 4 mm was successful although the can design was unnecessarily sophisticated and should be reevaluated in future work

Rolling of DU-10wt%Mo in carbon steel "picture frame" style cans proved successful in making full sized foils for surrogate fuels.