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Co-Rolled U10Mo/Zirconium-Barrier-Layer Monolithic Fuel Foil Fabrication Process

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CONTENTS

FIGURES

Co-Rolled U10Mo/Zirconium-Barrier-Layer Monolithic Fuel Foil Fabrication Process

1. FOIL FABRICATION FOR MONOLITHIC FUEL

Integral to the current UMo fuel foil processing scheme being developed at Idaho National Laboratory (INL) is the incorporation of a zirconium barrier layer for the purpose of controlling UMo-Al interdiffusion at the fuel-meat/cladding interface. A hot "co-rolling" process is employed to establish a ~25-µm-thick zirconium barrier layer on each face of the ~0.3-mm-thick U10Mo fuel foil (Figure 1). A flow chart of the foil fabrication process is provided in Figure 2.

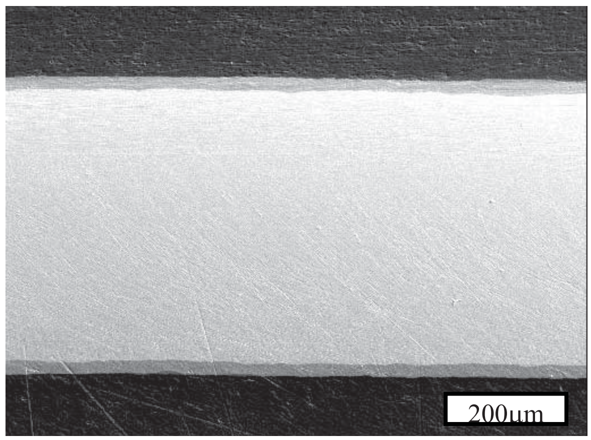


Figure 1. Cross sectional scanning electron microscope (SEM) micrograph of a Zr co-rolled U10Mo fuel foil.

Process steps distinctive to co-rolled zirconium monolithic fuel are summarized here and explained in detail below:

- Encapsulating a uranium-molybdenum alloy-coupon into a low carbon steel can assembly with a zirconium barrier material sheet/foil
- Hot rolling of the canned hot-rolling assembly to produce a bonding zirconium clad fuel foil
- Annealing and cold rolling of the Zr co-rolled fuel foil to obtain the desired thickness of "fuel meat"
- Shearing and surface preparing the Zr co-rolled fuel foil for the clad bonding.

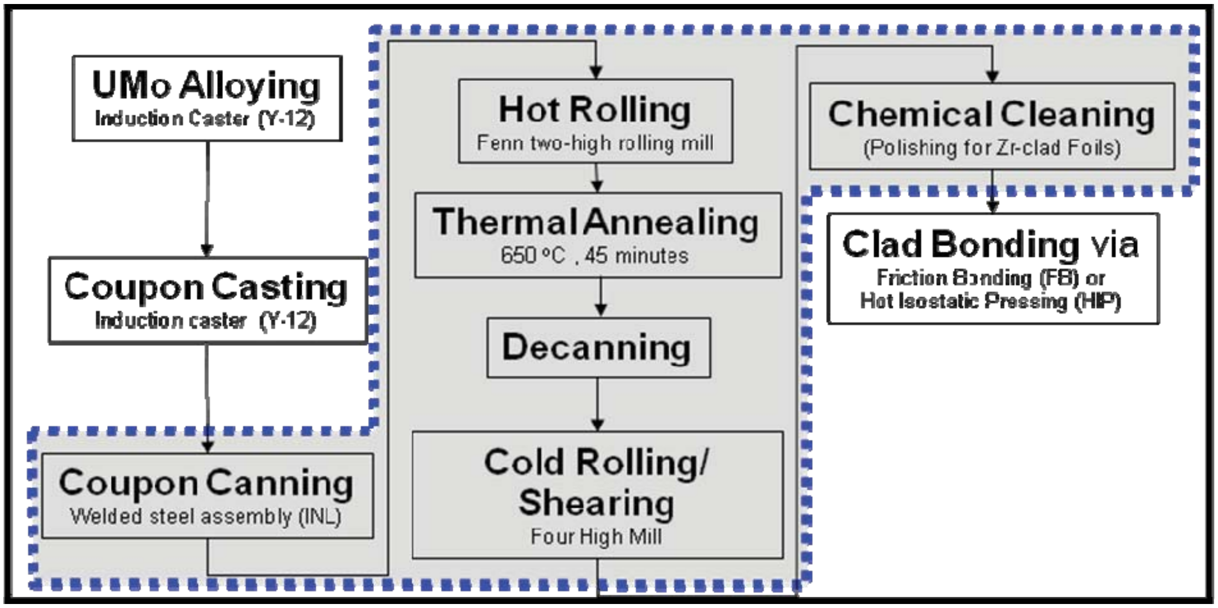


Figure 2. Processing steps associated with the fabrication of U10Mo/Zirconium co-rolled monolithic fuel foils.

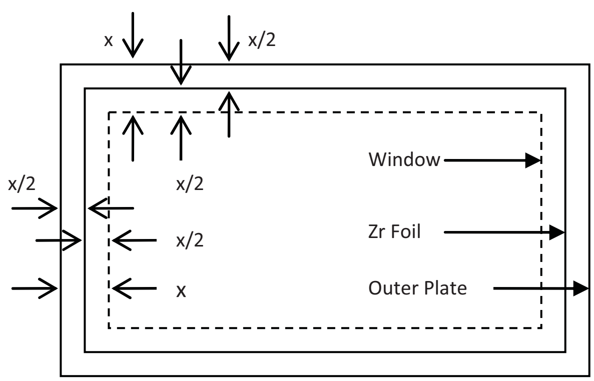
This document serves to present the currently established process methodology utilized in the fabrication of U10Mo/Zirconium co-rolled monolithic fuel foils.

2. ROLLING CAN ASSEMBLY: MATERIAL PREPARATION

2.1 Preparation of the Low Carbon Steel Canning Materials

Low carbon steel plate material is used for the rolling can assembly. The can consists of an “inner” “picture frame” piece and a top and bottom cover plate. The outside dimensions of the rolling can assembly is typically 1.0 in. greater than that of the alloy coupon being processed; thus affording a 0.5 in. perimeter of low carbon steel material (Figure 3a). The thickness of the carbon steel plate (1) picture frame is the same thickness as alloy coupon +/- ~0.005 in., and (2) top and bottom covers are at least as thick as the alloy coupon. Thicker top and bottom covers, ~0.005–0.025 in. may be used in order to avoid fly cutting/machining.

The rolling assembly plate material is prepared and assembled in a fashion that aligns the as-rolled direction of the canning materials to be parallel with the intended hot rolling direction. Low carbon steel components are machined to size, edges deburred, and grit blasted to remove any oxide scale. Components are then brushed using a stainless steel bristle brush at 2,900 rpm in order to remove any grit blast media residue and/or embedded particulate (Figure 3c). An orientation stamp is applied at the top edge of one cover plate, serving as a reference for the alloy coupon being processed and the rolling-assembly pass-orientation to be employed.



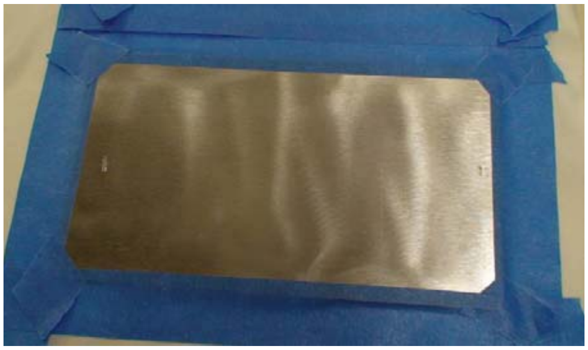
(a)

(b)

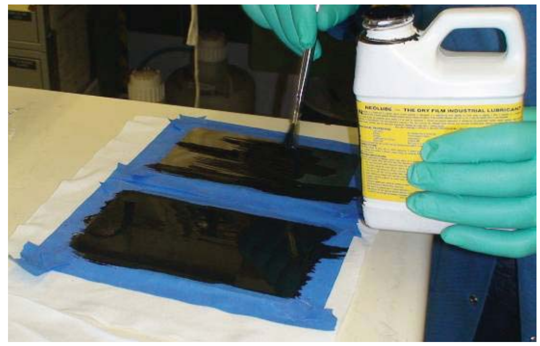
(c)

Figure 3. (a) Rolling can assembly dimensions, (b) Low carbon steel plates used in a rolling assembly, (c) wire brushing of a steel plate.

Location of the Zr barrier layer foil is established via scribe lines as described in Step 2 below. Once established, the top and bottom rolling assembly components are degreased using acetone/ethanol, masked, and a “no-stick” graphite coating is applied (Figure 4a). Note that the corner regions are also masked to facilitate spot welding of the Zr barrier foil in desired position. The “no-stick” coating material used is Neolube #2 produced by Huron Industries Inc.—a Colloidal Graphite/Cellulosic Resin Dry Film Conductive Lubricant. The Neolube coating is applied using an acid brush, allowed to air dry, and then residual solvent evolved (after masking taper removal) via a 10 minute hot plate treatment at 100°C (Figure 4b). The “no-stick” coating application process facilitates removal of the Zr co-rolled fuel foil from the rolling can assembly once the hot rolling process has been completed.



(a)



(b)

Figure 4.(a) Masked cover plate (note corner masking), (b) Neolube coating application.

2.2 Zirconium Foil Preparation

The Zr used is Alfa Aesar annealed 99.8% pure (metals basis) foil with a typical thickness of 0.25 mm/0.01 in. The thickness of the starting Zr foil used is selected so that a final barrier layer thickness of ~0.001 in./0.025 mm is obtained. The zirconium foil material is sheared to size, with the orientation of the Zr foil being such that the as-rolled orientation of the Zr foil will be maintained within the rolling assembly (i.e., parallel to the intended rolling direction). The Zr foil pieces are sheared larger that rolling assemblies “picture frame” window, but are smaller than the outer dimension of the cover plate; typically, 0.25 in. allowance with respect to alloy coupon’s dimensions is provided. Edges of the shear Zr foil are deburred using a buffing wheel. Prior to rolling can assembly construction, lay-out lines are scribed on the low carbon steel top and bottom cover plates in order to facilitate “no stick” coating masking of the Zr foil contact areas (as described above).

At least one face of each Zr foil is polished in order to remove any surface oxidation; polished Zr foil faces are, in later steps, placed in contact with the UMo coupon. Zr foil polishing is performed using lint free cloth and a 30µm water-based diamond polishing compound—Amplex Superabrasives WS/HV 1226Y-1 #30 MB (30µm) diamond polishing compound (Figure 5). Approximately 3 minutes per Zr foil sheet is typically adequate for obtaining a bright metallic surface sheen. Polishing compound residue is removed with ethanol, DI water, and lint-free clothes/wipes; adequate post-polishing cleaning is indicated by no appearance of the red polishing compound on the lint-free wipes. After cleaning, foils are handled by personnel wearing clean gloves (typically disposable neoprene gloves) to avoid introduction of hand-oil contamination.

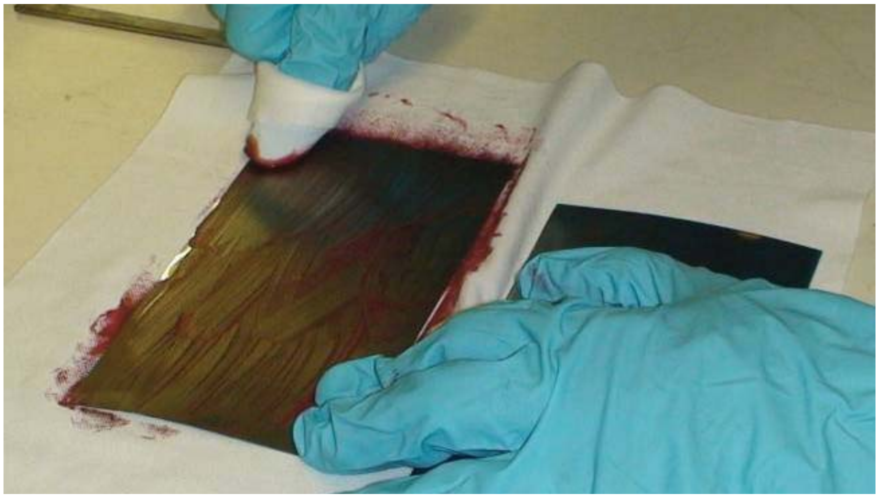


Figure 5. Zirconium foil polishing being performed using lint-free cloth and a 30 um water-based diamond polishing compound.

2.3 Attachment of Zirconium Foils to Cover Plates

Prepared/polished zirconium foils are spot (resistance) welded, at four corners, to the Neolube coated low carbon steel cover plates. Spot welds are made between the Zr foil and bare corner areas of each cover plate. The as-rolled direction of the zirconium foil is oriented parallel with primary rolling direction intended for the assembly.

2.4 Preparation of the Fuel Alloy Coupon

Alloy coupon edges and corners are filed in order to remove sharp edges that could potentially cut/breach the rolling assembly during processing. This step is performed in an Argon atmosphere glove box to prevent oxidation of the filings produced (Figure 6). After edge/corner filing, the alloy coupon is removed from the glove box and stored.

Figure 6. Edge filing of a UMo coupon in an argon atmosphere glove box.

Just prior to welding of the rolling assembly, the alloy coupon is chemically cleaned. The cleaning process involves dipping/soaking the coupon in nitric acid solution (70% distilled water/30% nitric acid) for 15–30 seconds, a sufficient time for oxide residue dissolution in most cases. The coupon is then transferred to a DI water rinse bath and then to an ethanol bath. The coupon is removed from the ethanol bath and inspected for complete oxide layer removal. If all areas of the coupon have a clean metal sheen, then the coupon is dried with a lint-free cloth and a final mass measurement obtained. If tarnished areas remain, the cleaning process is repeated. As needed, an acid resistant brush is used to scour the coupon to assist oxide removal. Extended periods of acid soaking are avoided to prevent pitting of the coupon. Cleaned and dried coupons are stored in clean polyethylene zip bags.

2.5 Rolling Can Assembly Lay-Up and Welding

Lay-up of the rolling assembly is typically performed in a fume hood on a lint-free cloth. The lay-up sequences consists of (1) a Zr foil/cover plate is laid "Zr face up" on the working surface, (2) the inner "picture frame" is set on top of the cover plate (Figure 7a), (3) the alloy coupon is placed in the picture frame window (Figure 7b), (4) the second Zr foil/cover plate is placed "Zr foil size down" on the picture frame/alloy coupon layer, (5) the side edges of the lay-up-stack are aligned, and (6) four binder clips (one per side) are put in place to temporarily hold the lay-up together.

The clamped rolling-assembly lay-up is moved to an argon atmosphere glovebox for edge welding. The welding sequence is (1) the rolling assembly lay-up is mounted vertically in a vise and two corners tack-welded applied via a Tungsten Inert Gas (TIG) torch and a low carbon steel filler material, (i.e., Radnor Welding Products ER70S2 TIG wire), (Figure 7c), (2) the assembly is repositioned and the remaining two corners are tacked, (3) the edges of the assembly are then individually TIG welded using filler wire/rod (Figure 7d), (4) the welded assembly is allowed to cool, and (5) the final thickness of the welded rolling assembly is recorded (needed for rolling schedule preparation). Note that the long edges are typically welded first, then the shorter edges. Also note that two pieces of copper plate ~2.5 in. × 4 in. × 0.25 in. are located on each side of rolling assembly during welding for thermal dissipation.

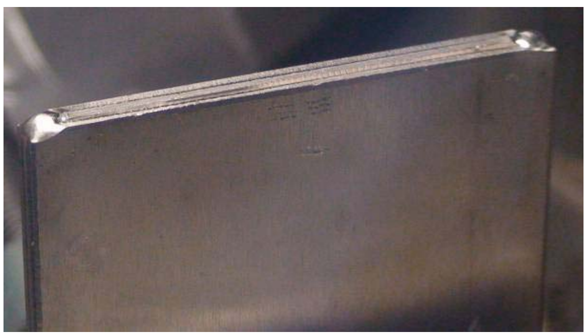
2.6 Preparation for Hot Rolling

The rolling mill rolls of the FENN® two-high rolling mill are preheated for several hours, or overnight, using 80°C (175°F) heating blankets (Figure 9a). The plate-heating box furnace is preheated to 650°C. The rolling assembly is placed on edge (in a plate-rack) inside the preheated box furnace for 30 minutes prior to rolling (Figure 9b).

During or just prior to preheating of the rolling assembly, the heating blankets are removed from the rolling mill rolls. The mill is started and allowed to run for a period of approximately 20 minutes. During this time (1) the rolls are brought into contact, leveled, and the zero-position established, (2) the rolling scheduled is programmed into the controller, (3) the speed of the rollers adjusted to have a linear feed rate of 16.4 ft/min. (30% setting), and (4) the rolls are coated lightly with vacuum oil (Figure 10). Note that the vacuum oil is periodically reapplied during the hot rolling process.

(a)

(b)



(c)

(d)

Figure 7. (a) Cover plate and picture frame components, (b) alloy coupon in picture frame window, (c) tacked corners of rolling assembly, (d) finished edge-weld.

Figure 8. Welded rolling assembly ready for hot rolling process.

3. HOT ROLLING

3.1 Hot Rolling Schedule

The rolling schedule used is based on the thickness of the rolling assembly, which can be directly measured throughout the hot rolling process. The percent reduction is based on per-pass thickness reduction and limited to values that are, based on experience, not expected to result in greater than 100 K lbs load force on the mill. The per-pass reduction range generally used is 5 to 20%. Higher percentage passes are typically used in the first half of the schedule to facilitate Zr barrier layer bonding. Cross-roll passes are used in the initial rolling sequence when additional coupon/foil width is desired. Without cross-roll passes, the obtained fuel foil will be approximately 110% of the starting width.

In the second half of the schedule, cold work of the assembly becomes a factor and smaller reductions need to be performed. Rolling speed is also reduced in the second half of the schedule in order to avoid rippling of the rolling assembly. Zero-passes are used as a means of keeping the maintain the target reduction. Namely, a repeat pass at the same roller spacing is used to compensate for reductions that are less than prescribed based on compliance of the mill. An example of a hot rolling schedule is provided in Appendix A.

(a)

(b)

Figure 9. (a) Roller heaters in place prior to hot rolling, (b) plate heating box furnace.

3.2 Hot Rolling Sequences

A preheated rolling assembly is removed from the box furnace, and given one to four passes, as quickly as possible, through a two-high rolling mill, as called for by the rolling schedule. Two operators, minimum, should be participating in this operation: “the pitcher” retrieves the assembly from the furnace using tongs and feeds the assembly into the rollers and “the catcher” grasps the exiting assembly with tongs and applies tension to assist in the pass (helping to maintain flatness of the assembly) (Figures 10a and 10b). A third operator is usually present for the purpose of incrementing the rolling spacing and recording in-process thickness measurements.

(a) (b)

Figure 10. (a) Application of vacuum oil to the rollers, (b) Assembly exiting rolling mill with operator applying tension.

After each pass sequence is complete, the assembly thickness is measured using calipers, the assembly inspected for weld and/or surface cracks, and as needed the orientation stamp is reapplied. Each assembly is then placed back in the furnace and reheated at 650°C for 5–15 minutes. After the reheat step, the process is repeated.

Experience has shown that maintaining the forward-end orientation of the rolling assembly throughout the process leads to relatively uniform thickness foils; regions of greater thickness, if any, typically become established on the trailing end of the assembly and can be removed during the follow-on cold rolling operation. Top-to-bottom rotation of the assembly is usually preformed between pass sequences in order to maintain straightness of the rolling assembly.

Twenty to thirty hot rolling passes are typical for preparation of a 0.25–0.50-mm-thick U10Mo foil. Figure 11 provides an example of assembly thickness reduction over time.

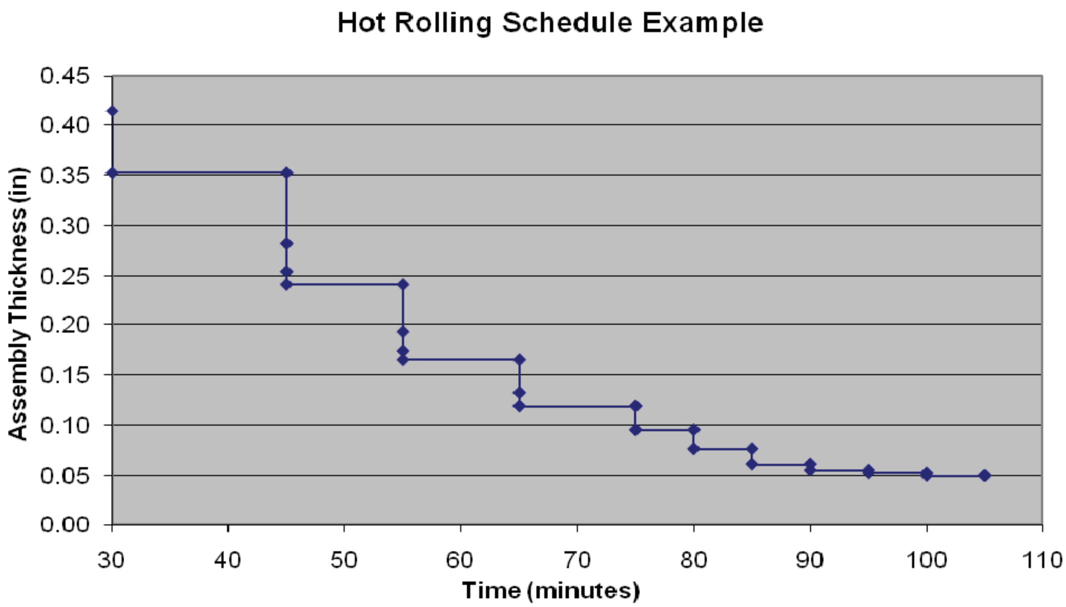


Figure 11. Plot of the assembly thickness versus time during hot rolling.

3.3 Post Rolling Foil Anneal

The co-rolled Zr foil is annealed while still in the rolling assembly to heal a portion of the working-stress defects imparted. The annealing treatment does not induce foil recrystallization, but does allow for some microstructural defect recovery to occur. The annealing treatment has been established as beneficial step prior to cold rolling. The annealing treatment is typically 45 minutes at 650°C. Once performed, the rolling assembly is removed from the furnace and checked for the presence of any radiological contamination (Figure 12).

Figure 12. Rolling assembly after hot rolling and annealing.

3.4 Foil Decanning

The annealed rolling assembly is placed in a fume hood for decanning, or extracting the fuel foil from the assembly. Decanning is performed with the assistance of a 12-gauge electric hand shear, given that the nominal thickness of the assembly is ~0.050–0.060 in. Prior to shearing, a standoff perimeter of ~0.25 in. is outlined around the foil location using a marker pen; the foil location is observable on the rolling assembly via a “shadow image” on each side of the assembly. Two ends and one side of the assembly are subsequently removed from the can using the hand shear. The assembly is then pried open and the foil is extracted (Figure 13).

(a)

(b)

(c)

(d)

Figure 13. (a) 12-gauge electric hand shear, (b) shearing of the rolling assembly, (c) opening of the sheared assembly to reveal the fuel foil, (d) the extracted co-rolled Zr fuel foil

3.5 Hot Co-Rolled Foil Evaluation

Upon extraction from the rolling assembly, the foil is cleaned with an ethanol-soaked cloth in order to remove remnant Neolube/graphite residue. The foil is then weighed to establish the weight percent addition of zirconium established in the rolling process. Subsequently, the foil is inspected for surface blemishes and a series of length, width, and thickness measurements are obtained. Post foil extraction weight and dimensional measurements are taken to support fuel material accountability requirements, provide input for future rolling schedules, and as a basis for establishing the needed follow-on cold rolling schedule. Typically, a cleaned co-rolled foil will smear free of radiological contamination and can be handled outside of a contamination area (i.e., fume hood).

4. COLD ROLLING

4.1 Purpose of Cold Rolling

The primary purpose of the hot rolling activity is to bond the zirconium foil to the UMo coupon as it becomes a fuel foil. The main function of the cold rolling activity is to establish a more homogeneous fuel foil with the desired specific fuel meat thickness. Relative to homogeneity, it has been established that thickness texturing/patterning arises during hot rolling—a likely consequence of hardness variations in the initial coupon coupled with the compliance of the carbon steel rolling assembly. It has been established that 25–50% reduction in the thickness of a co-rolled Zr fuel foil reduces the previously imparted fuel meat thickness variation or ~0.002 in., to a value of, ~0.0005 in. Cold rolling is also used to overcome the limitation of the plate heating furnace used at INL. Namely, the physical length of the plate heating furnace used in conjunction with hot rolling only affords an assembly length of ~34 in. Thus, application of cold rolling is needed to establish full-length ATR fuel plate fuel foils (i.e., 48 in. in length). Appendix B is an example of a cold rolling schedule.

Cold rolling is performed using a four-high Fenn® rolling mill having 1.625 in. diameter working rolls. Hot-rolled foils wide enough to yield two fuel foils are sheared in half prior to a cold roll. This practice allows cold-rolling adjustment of the foil curvature, imparted by stress relieved when the foil is sheared. Namely, a foil having a crown "thicker in the center than the edges" will curve when sheared down the middle. Generally, cold-rolled foils have had any significant crown removed during the cold-rolling sequence and prior to final shearing.

4.2 Cold Rolling Sequence

Cold rolling is conducted using a roller speed of ~5.4 ft/min and with the rolls lightly lubricated with vacuum pump oil. The initial roller gap setting is established with the rolls in the off mode; incrementally closing the gap until the foil will not pass through the gap. The four-high mill's gap is adjusted with two screw downs. The screw down position is indicated via a pointer and indicator dial/wheel. At each gap setting the foil typically passes three times through the rolls. Typically the gap is reduced two units of measure when the gap is changed, which equates to ~0.0006-in. gap change.

Multiple passes at a given setting are used to reduce foil thickness and also for adjusting/maintaining foil straightness. The foil may be flipped top-to-bottom or end-to-end, as needed, based on need. Tongs are used to apply tension exiting the rolls. Exit tension is used to help maintain a flat foil. Thickness measurements are periodically taken between passes, with more frequent measurements obtained as the foil nears the target thickness.

Note that if edge cracking is observed during cold rolling, the edges of the foil should be sheared away in order to prevent crack propagation (Figure 14).

Figure 14. Photo showing edge cracking of foil.

5. FOIL SHEARING

Foil shearing is used to establish the final length and width of a fuel foil. Shearing is performed using either a bench-top shear or a hydraulic squaring-shear, depending on the length of the foil (Figure 15). Clamping fixtures and guides are used to hold the foil rigidly in place during shearing. Edge quality is important and thus sharp, ding-free blades are required, as is proper alignment of the shear blades.

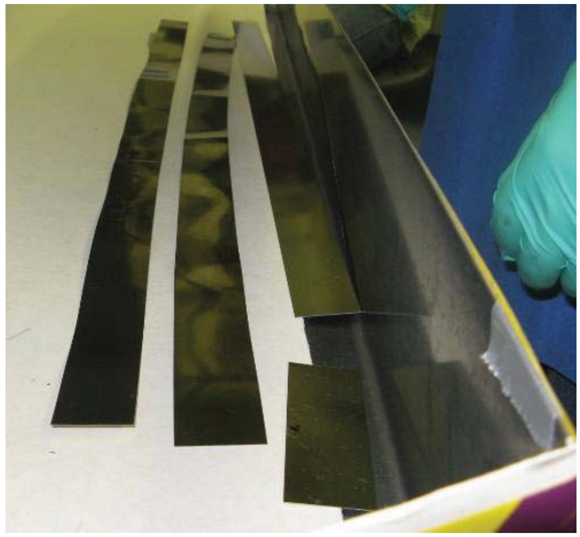


Figure 15. Photo of sheared cold rolled foils.

6. FINAL POLISHING

Prior to clad bonding of the prepared fuel foil, a surface polishing step is employed in order to remove any residual oxidation on the zirconium barrier layer, as well as any surface contamination deposited during rolling operations. The polishing is performed as described in Section 2.2 above. Alternatively, an ethanol-based dispersion of diamond power may be used, thus reducing the possibility of any polishing paste constituent residue on the foil.

Appendix A

Example Hot Rolling Schedule

Appendix A

Example Hot Rolling Schedule

Rolling Schedule

Alloy: XXX

Date: 11/15/09

Description: Co-Rolled Zr 40% HEU10Mo Coupon from Y-12

Pass	Zero Pass	Re-heat after pass	per pass % reduction	Roller setting	Measured Thickness	Reheat Soak (min)	Speed Setting	Max. Force	Notes
	Per Pass Schedule								
				0.415		30			**Initial Soak 30 min @ 650C**
1	☒	☒	15%	0.353		15	30%		cross roll+zero pass to 0.353 target
2	☐	☐	20%	0.282					
3	☐	☐	10%	0.254					
4	☐	☒	5%	0.241		10			
5	☐	☐	20%	0.193					
6	☐	☐	10%	0.174					
7	☐	☒	5%	0.165		10	25%		
8	☐	☐	20%	0.132					
9	☐	☒	10%	0.119		10			
10	☐	☒	20%	0.095		5	20%		
11	☐	☒	20%	0.076		5			
12	☐	☒	20%	0.061		5			
13	☒	☒	10%	0.055		5			zero pass
14	☒	☒	5%	0.052		5			zero pass
15	☒	☒	5%	0.049		5			repeat reheat+zero pass to measured 0.057 target
16	☐	☐	0%	0.049					
17	☐	☐	0%	0.049					
18	☐	☐	0%	0.049					
									Final Anneal 45 Minute @650C