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CONSIDERATIONS IN THE DEVELOPMENT OF A PROCESS TO MANUFACTURE LOW-ENRICHED URANIUM FOIL FUEL FOR THE HIGH FLUX ISOTOPE REACTOR

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ABSTRACT

A reference flow sheet is the one of the first planning steps in the development of a manufacturing capacity for low enriched uranium foil fuels and can be used to develop a work structure, a critical path schedule and identify development needs. The reference flow sheet presented is specific to the High Flux Isotope Reactor and is used to estimate the change in HFIR operating cost due to fuel conversion.

1. Introduction

The U.S. nonproliferation policy "to minimize, and to the extent possible, eliminate the use of HEU (highly-enriched uranium) in civil nuclear programs throughout the world" [1] has resulted in the conversion (or scheduled conversion) of many of the U.S. research reactors from HEU to low-enriched uranium (LEU)—low enriched meaning uranium having a ^{235}U wt % of 20 or less. However, some U.S. reactors operating with HEU have not converted to LEU because there is currently available no suitable LEU fuel that will allow these reactors to meet their mission requirements. Of these, the highest power density core and the most challenging to convert to LEU is the High Flux Isotope Reactor (HFIR) with its unique, continuously-graded, involute-curved fuel plates.

To minimize power peaking, the fuel distribution in the current, HEU fuel is contoured in a radial direction in each of the 540 fuel plates that comprise a HFIR fuel assembly. One assembly contains a single outer element with 369 fuel plates and a single inner element with 171 fuel plates.
During the current and immediate past fiscal years, a design for a new fuel for HFIR based on a

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uranium/molybdenum alloy (10 wt % molybdenum alloyed to uranium, termed U-10Mo) was developed. This design is documented in [2] and [3]. The geometry of the LEU HFIR fuel plates and elements would be unchanged from the current HEU design but the fuel region inside the plate would be changed from the current U_3O_8-dispersed-in-aluminum-particles to a metal sheet of U-10Mo – hereinafter termed a foil. The LEU fuel is believed to maintain the HFIR neutron source performance at the level obtained from the current HEU fuel, given that the reactor power is increased from the current level of 85 MW to 100 MW. Qualification of the fuel is currently being performed at the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL). Manufacture of the fuel will be the responsibility of a to-be-determined contractor, but specifications for the configuration and quality of the fuel are the responsibilities of the reactor operator. The reactor operator also bears financial responsibility for the production of the fuel. To estimate the financial impact on the HFIR operating budget of manufacturing the proposed LEU fuel, a study was undertaken to develop a conceptual production process — also termed a reference flow sheet — under the assumption that the LEU fuel would be qualified for use in the HFIR. The study will soon be published as [4]. This paper is an excerpt from [4] and presents that part of the manufacturing process that is unique to HFIR – the creation of a contoured LEU foil.

2. Description of fuel foil

In the context of this study, a foil is a thin sheet of alloy approximately 8 cm (3 in.) wide and 50 cm (20 in.) long by 300 Φm (12 mils) thick. The molybdenum content would be 10% by weight, and the uranium enrichment would be minimally less than 20% (19.75%). A photograph of a depleted uranium/molybdenum foil is shown in Fig. 1.

Fig. 1. Prototypic foil fabricated at the BWXT/Y-12 Plant.

A foil fuel appears to offer the best option for using an LEU fuel in HFIR because of the density of the alloy; a high-uranium loading is required to maintain the reactor performance at the level currently achieved with HEU fuel. Based on the previously reported neutronic studies [2], [3], the enriched uranium loading in a HFIR LEU fuel will have to increase from the current 9.4 kg ^{235}U/assembly in the HEU fuel to ~17 kg ^{235}U/assembly in an LEU fuel. The total fuel loading in HFIR will increase from ~ 10 kg/assembly for the current U_3O_8 HEU fuel to ~ 90 kg for the U-10Mo LEU fuel. In addition, to meet the performance objectives of HFIR, the fuel profile within each LEU fuel plate will have to be graded not only in the radial direction as currently required for HEU (see Fig. 2) but also in the axial direction to minimize flux peaking at the ends of the fuel plates.

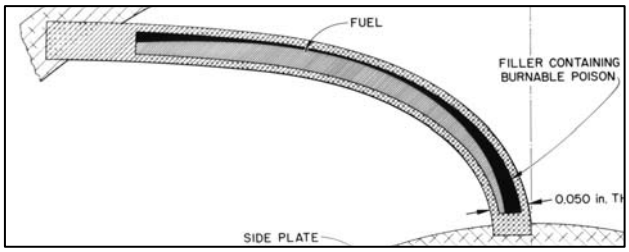

Fig. 2. Fuel profile inside current HEU inner element fuel plate.

Uncertainties in the performance of the fuel are currently being resolved by irradiations at the ATR. Accommodating fission gases during irradiation is the primary concern. Uranium metal containing > 7% Mo seems to be capable of retaining the fission gases in the fuel structure in small bubbles (i.e., submicron) necessary for predictable, low magnitude swelling during irradiation. Reaction of the U/Mo with the aluminum in the cladding or matrix must be avoided to retain the structure necessary for retention of these fission gases in small bubbles. A diffusion barrier between the fuel meat and cladding will likely be required to meet this requirement. Silicon and several other materials show promise as a diffusion barrier. Unique to HFIR are the added stresses and strains that may be present at the bond interfaces during forming and irradiation; these stresses due to forming the relatively sharp radii of curvatures required for the involute-shaped fuel plates (see shape in Fig. 2). In foil fuel, the bonding of the fuel meat to the cladding is the major area of uncertainty.

3. Sizing of fabrication operations

In the current HFIR HEU process, manufacturing the flat plates represents about 60 - 70% of the cost of a fuel assembly. Comparison of the foil and current dispersion fuel fabrication is shown in Fig. 3 in a simplified process flow diagram. In the foil process, there are more steps with the attendant increased in-process inventory levels and "hold points" (in-process inventory storage). Furthermore, there are significant differences in complexity-of-operations for the foil process when compared to the current dispersion fuels. The blend down, alloying and initial rolling steps are common to all LEU fuels and not unique to a HFIR foil fuel.

For the HFIR foil fuel, the production requirements for the various process steps were determined by the facility operator's planned annual consumption of fuel plates (540 plates/assembly times 10 assemblies/year) and assuming a yield for each of the various process steps. The production requirements for the various process steps required to fabricate a LEU fuel for HFIR are shown in Table 1. The yields assumed in this study, also shown in Table 1, were relatively optimistic values based on the judgments of experts familiar with similar process steps when in a full production mode. In development and startup production, the yields will likely be lower than the values listed in the table. As can be seen, the overall yield through the process is ~ 50%.

4. Rolling operations for foil fuel

For HFIR, two foil thicknesses will be required; about 290 Φm (~0.012 in.) thick for the inner

fuel elements and about 410 Φm (~0.016 in.) for the outer elements. The "as rolled" width of the finished foils for both HFIR elements will be wider (about 9 cm [~3.5 in.]) than the finished fuel core specifications to allow a margin for trimming.

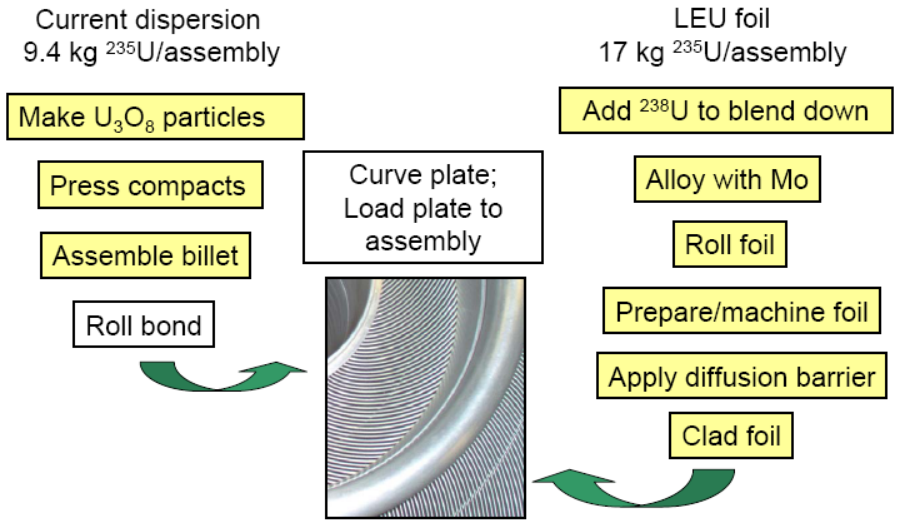

Fig. 3. Simplified comparison of process steps for current HEU fuel and foil-based LEU fuel.

Table 1. LEU foil fuel production requirements for HFIR

Processes (reverse order from Fig. 3)	Needed output plates	Yield (%)	Needed input plates	U-10Mo (kg)
Plate assembly	5,400	90	6,000	1.039
Diffusion barrier	6,000	90	6,667	1,154
Foil preparation	6,667	85	7,843	1,358
Rolling foil	7,843	85	9,227	1,598
Alloying	10,458	85	10,856	1,880
Blend-down	—	75	—	2,506
Inventory buildup (50% greater than annual for first few years)	—	—	16,283	3,759

The reference process for the foil rolling step is shown in Fig. 4. In this process, the individual foil blanks of ~250 mm thick would be hot and cold rolled to the desired thickness for subsequent processing. Rolling of uranium metal into foils to <250 mm thick is well-documented technology, and some demonstration, full-size uranium fuel foils have been rolled. However, the rolling of uranium foils with the dimensional control that will be required in a production process has not been demonstrated. Uranium metal alloys work-harden rapidly during rolling, and the billets must be annealed at ~ 930EC between each hot rolling pass. A 10 – 20% reduction-per-pass hot rolling schedule can be achieved to a thickness of ~500 Φm. The finished foil thickness is achieved by cold rolling. In cold rolling, after only 50 Φm (~0.002-in.)

reduction in thickness, annealing is required. A salt bath to maintain the temperature at the required 930EC has been used for annealing uranium alloys between rolling passes and is the reference annealing process. In a dedicated LEU production line, other annealing techniques (i.e., infrared heating) may be more efficient and should be considered in the future. The thickness of the finished foils needs to be controlled to within at least ~ 25 Φm (0.001 in.) within each foil and among foil-to-foil production. At this stage in development, no information is available concerning the statistical variation of the foil rolling process.

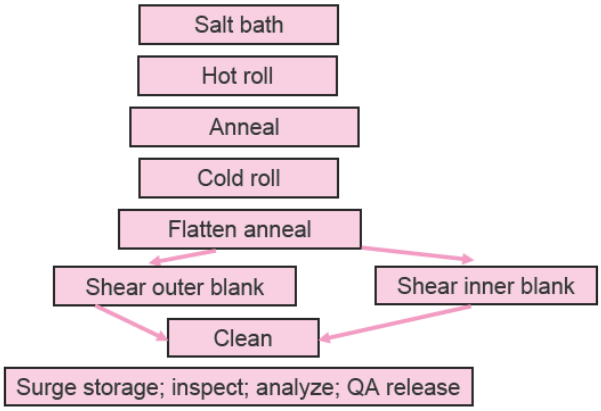

Fig. 4. Conceptual foil rolling operations.

After cold rolling, the fuel foils must be flattened. In the reference process, flattening is accomplished by the phenomena of creep, using dead weights on top of the fuel foils at the annealing temperature. This is analogous to the process used in the current HEU fabrication process to flatten aluminum fuel plates after cold rolling. After flattening, the fuel foils would be cut to their finished dimensions by using a blanking die and shearing.

Inspection of the finished foils would consist of dimensional and surface inspection covering 100% of the plate surface and destructive metallographic examination of samples for grain structure. The weight of each finished foil would be measured and recorded for accountability.

The waste streams generated in rolling of flat foils would primarily consist of rejected fuel foils and trimmings from the blanking of finished flat foils. Some part of this waste stream might be considered clean scrap and recycled in the melting step in the alloying process. Other parts would be considered disposable waste.

5. Foil preparation for assembly

The foil preparation step for HFIR differs from other reactors because of the requirement that the flat fuel foil must be contoured in both in the radial and axial directions to minimize the nuclear flux peaking at the edges and ends of the fuel plates during operation in the reactor. Minimizing this flux peaking is necessary to allow HFIR to maintain the current, HEU fuelled level of performance. The need for the axial fuel profile is unique to an LEU HFIR fuel because the current HFIR HEU dispersion fuel is contoured only in the radial direction.

Achieving the required contours on 290- and 410-Φm-thick flat foils has not been demonstrated. A simple rolling process may not be capable of achieving the required dimensional control on the non-symmetrical radial profiles required. Regardless, the axial contour required would necessitate a separate forming step. Other conventional metal forming techniques (i.e., forging or swaging) might be applicable in forming the contours, but these techniques have some of the same drawbacks as rolling. At this stage of development, machining the required profile appears to be the most direct approach for success within the likely dimensional tolerances that will be allowed.

In considering machining of the required profiles on the flat foils, the applicability of the HFIR HEU-type curve contour (see Fig. 2) to such thin foils was not apparent and would require a more complex machining operation to form the required curved contour. With the thin foils, a curved contour would approach a straight line for all practical purposes. In the HFIR HEU dispersion fuel, the one-sided radial contour, which is formed by a powder metallurgical technique, may not be the best configuration for a HFIR LEU fuel foil. A foil symmetrical about its axial midplane may be easier to machine or form, and such a symmetrical foil would provide significant benefits in the subsequent diffusion barrier application, cladding preparation, bonding, and involute forming steps. Comparable, industrially manufactured metal foils with tapered edges symmetrical about its axial midplane include double-edged razor blades. Razor blades are produced by the millions worldwide.

After determining that symmetrical straight contoured fuel foils would not have any negative impacts on a LEU fuel performance in HFIR, the foils contours shown in Fig. 5 – corresponding to continuous profiles described in [3] - were selected for the reference foil designs. The end contours would be formed by machining in a separate operation.

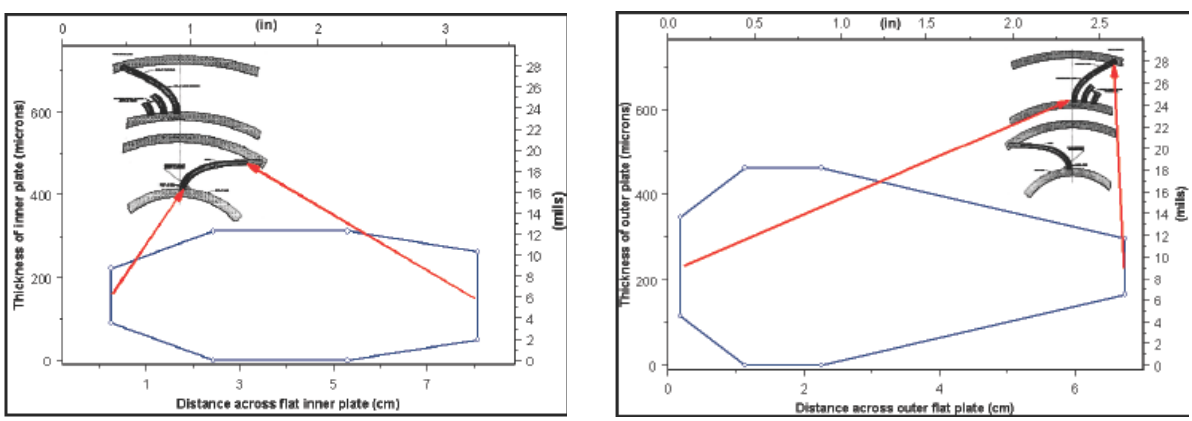

Fig. 5. Flat edge profiles; inner element fuel plate (left) and outer element fuel plate (right).

The reference flow sheet for the preparation of the outer and inner HFIR foils for assembly into the cladding is shown in Fig. 6. Most of this flow sheet is based on engineering assessments of the applicability of comparable processes within the metal processing industry. Essentially none of this technology has been developed for U/Mo foils. Machining is the reference for forming

for the radial (edges) and axial (ends) contours for HFIR fuel foils.

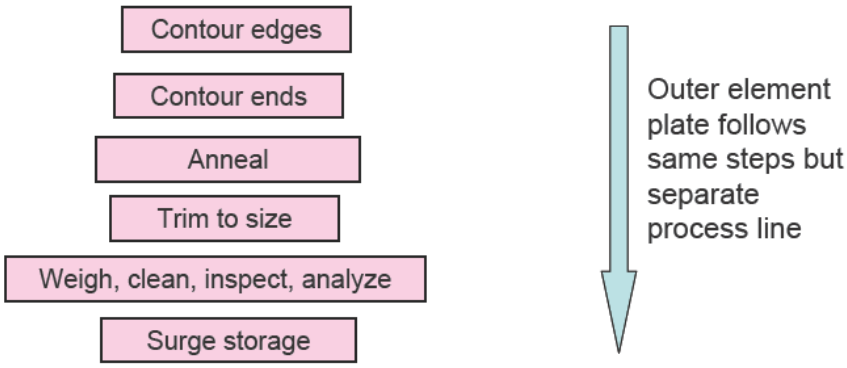

Fig. 6. Procedure for preparing foils for assembly in clad.

Because of the thousands of HFIR LEU foils that will need to be processed annually, contour forming of HFIR foils will require a specially designed computer-controlled automated machine that would include feeding of the foils for edge and end contouring, automatic dimensional inspections, weighting, and surface inspection of the end products. Most likely the machining operation would be done by either cutting or grinding using a cutting fluid. The cutting fluid should minimize uranium contamination in the processing area, but special containment enclosures will be required. The dimensional inspection should be capable of a resolution of 12 Φm (~0.0005 in.) and the weighing to ~ 0.010 g. The foil surfaces should be free of scratches and gouges of some specified depth and length (i.e., about 50 Φm [~ 0.002 in.] deep and 2.5 cm [~1 in.] long). Standards for surface defects will be required.

After forming and inspection, the foils may require trimming to meet a uranium loading tolerance, and the surfaces must be cleaned for application of a diffusion barrier. The reference flow sheet assumes that trimming will be necessary. In the reference flow sheet, the trimming and cleaning of the foil surfaces are accomplished using an acid leaching process. Trimming for weight control of the foils could also be accomplished by machining or grinding the flat surfaces. The proper cleaning of metal surfaces for plating or sputter coating is extremely important to the successful application of a coating. Acid cleaning for this purpose is established technology.

In the processing of a foil, the dimensional inspection data and weight of each foil after contour forming and trimming will constitute the primary information for certifying an individual fuel plate for homogeneity and uranium content. Homogeneity scanning by X-ray attenuation, as currently done for dispersion fuels, should not be required.

The uranium waste streams generated in the foil preparation for assembly step would primarily consist of sludges from machining, sludges for acid leaching, and rejected foils. Most of the foils rejected before application of a diffusion barrier might be considered clean scrap and recycled in the melting step in the alloying process. Some other rejected foils would be considered waste that would likely be considered below economically recoverable quantities and disposed as solid low-level radioactive waste. The sludges would have to be treated and

packaged before discarding as solid low level-waste. Other solid radioactive waste generated in this process would include room air filters, discarded personal protective clothing, and miscellaneous other waste generated within the regulated zone. All of this waste would require packaging and disposal as low-level radioactive waste.

6. Foil coating, aluminum clad preparation and applicaton

The reference flow sheet for aluminum cladding preparation is shown in Fig. 7. The configuration of the aluminum hardware for foil fuel plates will be significantly different from the "picture frame" hardware used in making roll-bonded dispersion fuel plates at a ~8:1 reduction ratio. For foil fuel plates, the friction stir welding or hot isostatic pressure (HIP) bonding processes currently being developed for LEU fuels require that the aluminum hardware be able to receive the finished foils and be close to the overall finished fuel plate dimensions. The reference cladding hardware for the HFIR foil fuel plates will be two matching aluminum strips ~10 cm (4 in.) wide by ~60 cm (24 in.) long, containing contoured recessed areas 150 – 200 Φm (0.006 – 0.008 in.) deep to place the finished foils in a "clam shell" type configuration. A clam shell configuration will require Al-to-Al bonding on only one plane. Picture frame type aluminum hardware with Al-to-Al bonding on two planes might be appropriate for rectangular-shaped foils, but this configuration is not likely applicable to the contoured foils required for HFIR.

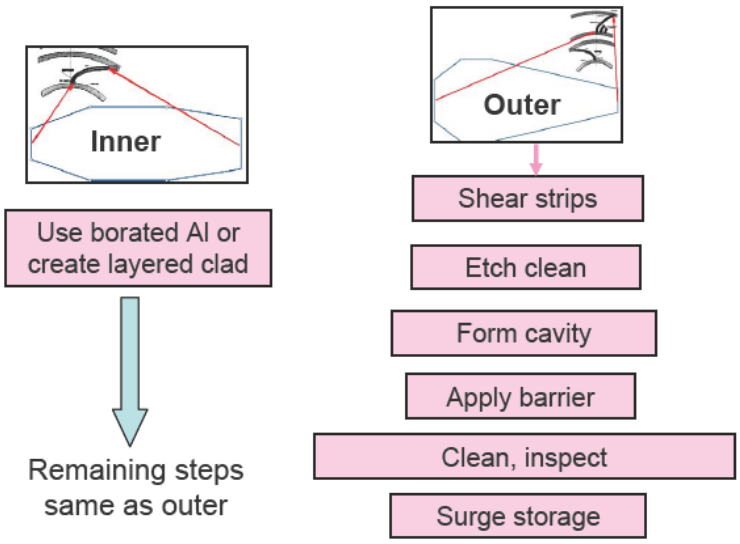

Fig. 7. Process steps for aluminum cladding and foil coating.

The flow sheet also includes the step for applying a diffusion barrier to the surfaces of the formed foils. Silicon (a coating <25 Φm [0.001 in.] thick) has shown promise as a diffusion barrier, but the detail requirements have not been established. A thin coating of silicon (or some other metal such as zirconium) could be applied to the foil surfaces either by physical vapor disposition (sputtering) or by an electroplating process. Electroplating — well known technology for coating metal surfaces — was selected as the reference process. After coating, the thickness and integrity of the coating will require nondestructive inspection to confirm specified thickness and detect any flaws. Several techniques for determining the coating thickness (for instance a capacitance probe) are available commercially.

In preparing the cladding, a burnable poison equal to 0.0164 g ∀ 0.0016 g ^{10}B must be homogeneously incorporated into the cladding for the HFIR inner fuel plates. In the current HEU dispersion fuel, this poison is added to the filler powder as natural boron (20% ^{10}B) carbide powder during the making of the fuel compacts for roll bonding. For a foil fuel, including the burnable poison in the cladding is the reference method and will require the procurement of a special lot of aluminum alloy 6061 TO sheet stock with a boron content of ~ 750 ppm. Procuring such a special lot of aluminum sheet may be difficult and will likely entail the purchase and processing of an entire 10,000-lb billet of the aluminum alloy into the required 760-Φm (~0.030-in.) thick by 10-cm (4-in.) wide by 20-cm (8-in.) long sheets. For the outer fuel plates the 750-Φm-thick (finished plate thickness ~1250 Φm) sheet would be procured as commercial alloy 6061 with the O temper in 102-cm by 204-cm (4-ft by 8-ft) sheets and dedicated for nuclear service. The aluminum sheets for the inner and outer plates would be sheared into strips of appropriate width [~10 cm (~ 4 in.)] and length [~60 cm (~ 24 in.)]. In procuring commercial aluminum plate for the outer plates, consideration should be given to the boron content (specified as < 30 ppm B).

7. LEU fuel plate assembly

The assembly of LEU foil fuel plates through the critical bonding step will require the use of a different technology than has been used for manufacture of the dispersion fuel plates. The reference flow sheet for the assembly of LEU fuel plates is shown in Fig. 8. The inner and outer fuel plates required for HFIR can be assembled in campaigns on a common production line if adequate production capacity is provided. As a minimum, assembly of the finished foils into the clam shell aluminum cladding hardware will require a "clean room" type environment to minimize surface contamination of the bonding surfaces. The reference for assembly is manual, but automated assembly in a controlled environment could be required to control surface contamination for bonding.

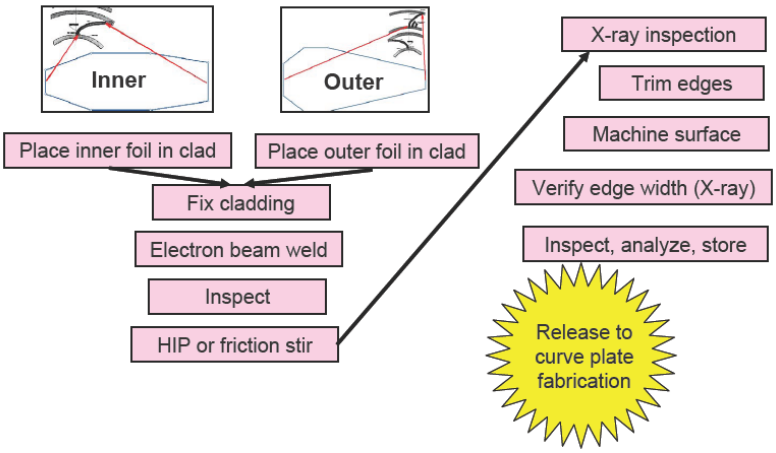

Fig. 8. Proposed process for final assembly of fuel plates.

Hot isostatic pressing (HIP) was selected as the reference for bonding because this process is more adaptable to the high production volumes that will be required for the LEU fuels. Multiple plates can be processed simultaneously by HIP. The friction stir welding process, which is a single-plate incremental process, may provide a bond with characteristics more favorable to the performance of the fuel in the reactor because of the lower time at temperature required to achieve bonding. However, the recognized need by the LEU program for a diffusion barrier probably leads to HIP as a viable process for bonding LEU fuel plates. For a HIP process, the fuel plate assembly must be sealed before HIP. The reference for this sealing is electron beam welding, which will provide an evacuated sealed fuel plate assembly.

8. Costs and implementation schedule

The overall purpose this work was to develop a better understanding of the costs of infrastructure, operating costs, and implementation schedule associated with the fabrication a LEU fuel for HFIR. Deriving these costs and schedule must, at least, be based on an understanding of the manufacturing requirements and steps necessary for such a fuel.

The operating cost of a foil fuel can only be inferred from the current operating cost of fabricating HEU flat dispersion fuel plates and analysis of the added complexity of fabricating foil fuel plates. The operating cost of making a flat foil fuel plate based on the added steps and the complexity of more process steps and the increased number of these steps that must be performed in a contamination zone will certainly be more expensive than for a dispersion fuel plate. The fabrication cost of a foil fuel plate could easily be twice the current cost. In the current manufacturing of HFIR HEU fuel elements containing dispersion fuel plates, fabrication of the flat fuel plates represents about two-thirds of the total costs of an element. In a foil fuel, the costs of the flat plates will likely represent a larger percentage.

9. References

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[4] J. D. Sease, R. T. Primm, III and J. H. Miller, *Conceptual Process for the Manufacture of Low Enriched Uranium/Molybdenum Fuel for the High Flux Isotope Reactor,* ORNL/TM-2007/39, draft, publication expected September 2007.