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PRELIMINARY RESULTS FOR THE CO-ROLLING PROCESS FABRICATION OF PLATE-TYPE NUCLEAR FUEL BASED IN U-10Mo MONOLITHIC MEAT AND ZIRCALOY-4 CLADDING

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ABSTRACT

The fabrication process of plate-type nuclear fuel with monolithic meat is under development at CDTN. The U-10Mo alloy was chosen as the meat material due to its high density, corrosion resistance and the higher dimensional stability proportioned by the metastable gamma phase, which presents a lesser extension of the breakaway swelling phenomena occurrence during irradiation tests. The monolithic meat was cut from an U-10Mo ingot, that was induction melted at CDTN. The co-rolling process was adopted due to the higher mechanical properties and melting point of the Zircalloy-4 cladding material, which presents a lesser discrepancy in relation to the meat material properties, when compared to the aluminum 6061 alloy. Preliminary plates were obtained by means of the co-rolling process, performed at 650, 800, 950 ^{o}C with total thickness reduction of 80%, followed by a pickling step and cold co-rolling passes. The plates were characterized through bending tests, optical microscopy and radiography. The co-rolling temperature of 800 ^{o}C presented the best results, with a homogeneous distribution of the total thickness reduction between the cover plates and the meat, and the absence of delamination in the bending test samples. It was observed the occurrence of meat thickening in its ends, according to its longitudinal axle, parallel to the rolling direction, that is known as the "dog bone", for the three co-rolling temperatures.

1. INTRODUCTION

The research and development of new materials to be used as fuel elements in research and test nuclear reactors received a major push in the past 35 years through the Reduced Enrichment for Research and Test Reactors – RERTR, proposed by the Department of Energy of USA under the coordination of the Argonne National Laboratory - ANL. The program's goal is to minimize the risk of nuclear weapons proliferation by means of the reduction in the degree of isotopic enrichment in uranium-235, from 90 to 20%, without major changes in the design of the operating reactors [1].

These premises led to the need to increase the uranium density of the fuel in order to recover the loss of fissile material. The first stage of the program development proposed to increase the uranium density of the current fuels used, i.e., UAl_x-Al, U_3O_8-Al and $UZrH_x$ dispersions, achieving densities around 3.7 g/cm^3 [2]. Afterwards, irradiation tests were performed with intermetallic compounds of uranium. At this stage, the occurrence of intense breakaway swelling was observed in compounds with U6Me formula, as well as in some uranium silicides. A good behavior under irradiation was observed for U_3Si_2 dispersions in Al, which was qualified with the density of 4.8 g/cm^3 [3].

The need for increased power and neutron sources with high thermal fluxes continued driving the search for alternatives to a further increase in the uranium density of fuel materials. The use of metallic uranium is not feasible due to the occurrence of intense swelling related to the presence of the α-phase with orthorhombic crystal structure [4], the uranium stable phase at room temperature. Thus, tests with uranium metal alloys with additions of transition elements which stabilize the crystal structure of body-centered cubic crystal lattice (γ-phase) at room temperature, such as Mo, Nb, Zr and Ti, were performed. The U-Mo alloys have been one of the most investigated due to the retention capacity of the γ-phase, which rises as the Mo content is increased [5], and their high densities. Dispersions of the U-10Mo (wt%) alloy in aluminum have achieved uranium densities between 8 to 9 g/cm^3, with a good behavior when submitted to irradiation irradiation tests, from the viewpoint of dimensional stability, which is associated with the occurrence of the swelling phenomenon.

Despite the promising initial fuel performance tests, the formation of an interaction layer (IL) in the interface between the Al matrix and the U-10Mo alloy particles was observed. The IL formation is caused by the elements diffusion between the U-Mo alloy particles and the Al matrix. Intermetallic compounds generated at the interaction layer were considered responsible for the reduction of the thermal conductivity, swelling occurrence and porosity at the interface, being detrimental to the nuclear fuel plates performance [6-11].

Aiming to decrease the (U,Mo)-Al interaction kinetics, Si addition to the Al matrix was proposed, since it permits to reduce significantly the IL volume and promote the formation of silicon-rich phases that exhibit better irradiation performance [12-14]. In order to achieve uranium densities higher than 9 g-U/cm^3 and solve the IL formation problem, the development of monolithic fuel plates was proposed [15]. The use of the monolithic fuel provides a sharply decreased fuel surface area per unit mass that could minimize the fuel-aluminum reaction.

First irradiation tests were performed in monolithic fuel plates produced through roll bonding process in U-10Mo alloy foils mounted in an Al frame between Al cover plates. Despite the initial promising results obtained in the point of view of reducing the extension of the interaction layer occurrence, discrepancies between the mechanical properties of the U-10Mo foils and the Al cladding material, associated to the excessive reaction between matrix and fuel material, induced by the higher temperatures of the hot roll bonding process, has lead to the adoption of fuel plates fabrication processes based on the application of friction and pressure, like Friction Bonding (FB) and Hot Isostatic Pressure (HIP) processes [16-18]. Additionally, zirconium foils are being employed as diffusion barrier through co-rolling process with the U-10Mo alloy foils [19].

The development of high density uranium alloys as well as the fabrication process of both dispersion and monolithic fuel plates is being performed at CDTN/CNEN, addressing the international concerns on nuclear weapons proliferation. These R & D efforts were motivated by the start up of the Brazilian Multipurpose Reactor, which is foreseen to 2018. Unlike the current materials and process design employed at RERTR program, zircaloy-4 alloy was chosen in this work as cladding material, as an alternative to avoid or minimize the IL occurrence. This approach was already proposed for monolithic U-7Mo fuel plates fabricated through the hot roll bonding process and has shown promising results in irradiation tests with low burn-ups [20,21]. Besides, its higher melting point would allow the fuel plates to be applied at higher neutron fluxes and consequently, higher temperatures. At last, the roll bonding process is expected to be a suitable processing alternative due to the fact that the mechanical properties discrepancy between the cladding and meat materials are lower than that observed when aluminum cladding is employed. The results of preliminary processing of zircaloy-4 clad plate-type nuclear fuels based on monolithic U-10Mo alloy meat at different roll bonding temperatures are presented.

2. EXPERIMENTAL

The U-10Mo ingot was melted in an induction furnace using pure natural uranium and pure molybdenum metallic pellets. The molten alloy was cast in a copper mould resulting in a round ingot with 25 mm diameter and 85 mm height. The meat was composed by machined disks (Fig. 1) cut from the ingot and measured 22 mm in diameter and a thickness of 6 mm. The picture-frame process was employed and the raw plates were composed by a zircaloy-4 frame piece and two cover plates, measuring 100x40x6 mm^3 and 100x40x2 mm^3. The frame piece had a centered hole with 22 mm in diameter, where the U-10Mo monoliths were inserted. Before mounting, the surfaces of the plate components were cleaned by a pickling solution composed by HF, HNO_3 and deionized H_2O, in the volume proportion of 1:6:15. The U-10Mo disk was grinded with SiC sandpaper and washed with acetone. TIG welding process was adopted to fix the cover plates. The top and bottom corners of the raw plate were not welded in order to allow the retained gases to escape from the frame during the first rolling pass.

The raw plates were heated up in a resistive tubular electric furnace at roll bonding temperatures of 650 ^{o}C, 800 ^{o}C and 950 ^{o}C and kept at those soaking temperatures for 30 minutes under argon flow of 5 L/min in order to reduce the zircaloy-4 oxidation. Pasqualini [20] reported the results for monolithic fuel plates roll at 655/675 ^{o}C. In this work higher temperatures were tried in order to study the effect of the discrepancies of mechanical strength of both materials in the dimensions of the plates as well as in the occurrence of defects. The hot roll bonding process was carried out with a total thickness reduction of 80% with an initial thickness of 10 mm. An initial roll bonding pass with a thickness reduction of 10% was applied. A thickness reduction of 15% was applied for the subsequent passes reaching a final thickness around 2.0 mm. After each roll bonding pass the plates were brought back to the tubular furnace for 5 minutes before the next pass. The obtained monolithic plates are presented in Fig. 2. In order to remove the oxidation layer formed during the hot roll bonding process, the monolithic plates were pickled in the HF/HNO_3 solution. The roll bonded plates were then cold rolled to improve the plate flatness, surface finishing and the thickness homogeneity. The thickness reduction employed was divided into two passes of 5% each, totalizing 10%, resulting in finished plate thickness around 1.8 mm.

The resulting plates were cut transversally to the rolling direction into three parts using a diamond plated cut-off wheel. The central specimens containing the monolithic fuel meat were used for characterization in the cross-section as well as in the longitudinal direction in a light microscope. The presence of defects like delaminations was investigated and thickness measurements were performed.

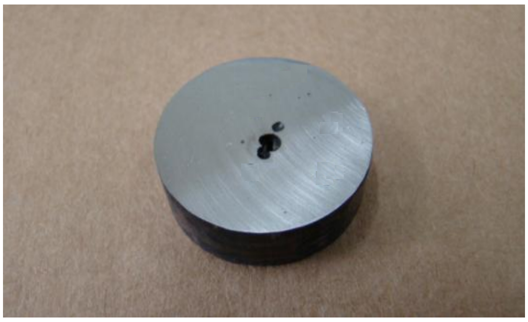

Figure 1. U-10Mo alloy disk to be used as the monolithic fuel meat. Central porosity originated during ingot casting is observed.

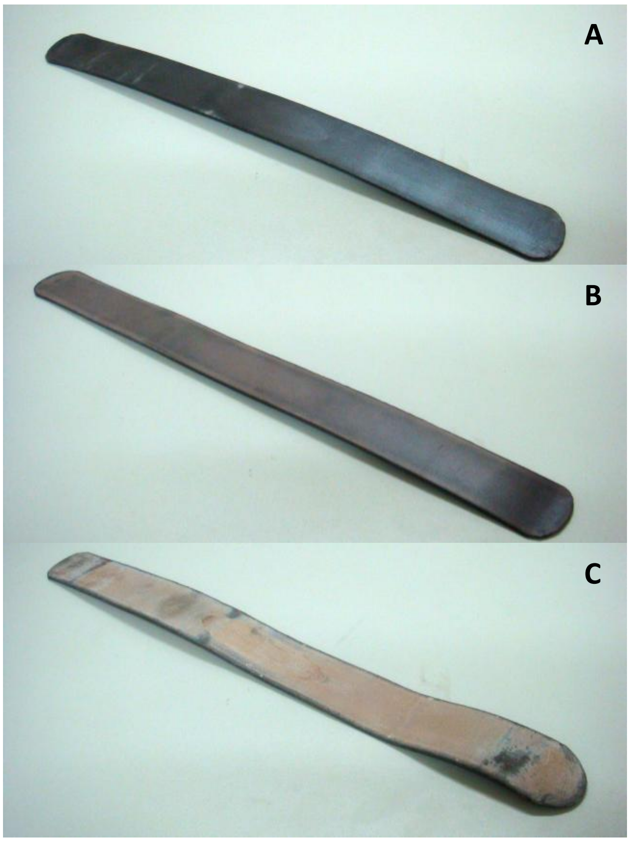

Figure 2. Monolithic fuel plates roll bonded at (A) 650 °C, (B) 800 °C and (C) 950 °C.

The samples of the plate ends, composed only by zircaloy-4, were used in the bending tests carried out in an INSTRON 5882 mechanical testing machine with 100 kN capacity. The specimens measured 150x35x1.8 mm^3 and were placed over a matrix with two supporting points separated by a distance of 75 mm. An indenter plate containing a round section bar of 8 mm in diameter at its end was placed between the supporting points of the matrix and was moved downward with a speed of 20 mm/min until reaching a sample bending angle of 90 degrees. Fig. 3 shows the assembly ready for testing. The load and displacement values of each test were registered and specimens were macroscopically evaluated in order to identify the occurrence of delaminations.

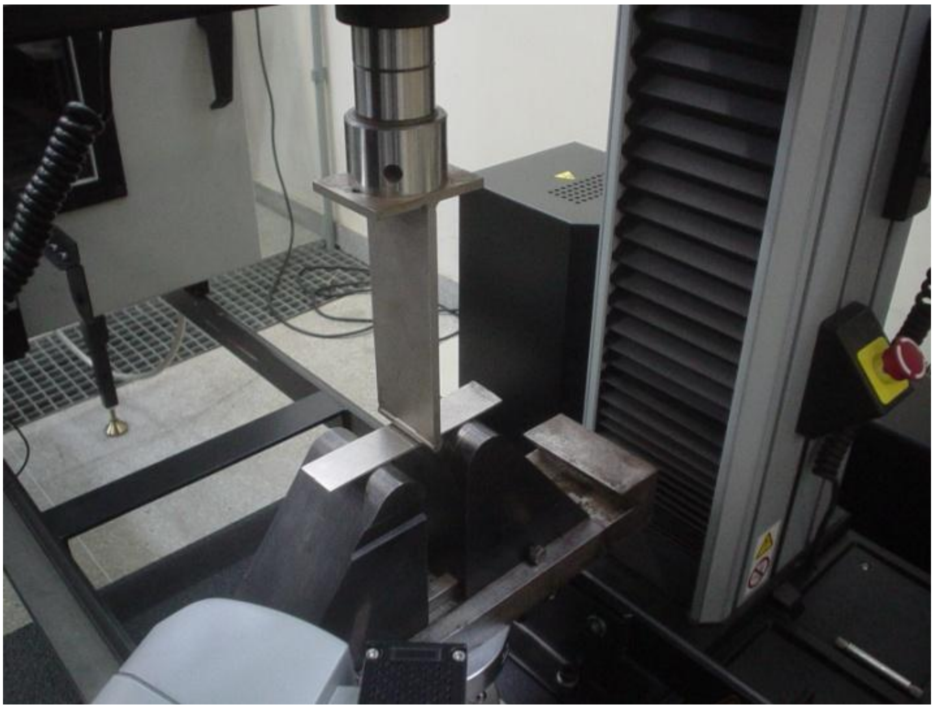

Figure 3. Bending tests assembly.

3. RESULTS AND DISCUSSION

The load versus displacement curves obtained through bending tests are shown in Fig. 4. The fragile behavior was noticed for the roll bonding temperatures of 650 ^{o}C and 800 ^{o}C. The fracture of the specimens at those temperatures caused the premature test interruption. Delamination between cover and frame plates were observed only for the 650 ^{o}C roll bonding temperature. Additionally, it was noticed that the specimens roll bonded at 800 ^{o}C presented higher loads and fractured before the ones roll bonded at 650 ^{o}C. This behavior is supposed to be a consequence of the fact that the latter offered lower resistance to the bending loading due to occurrence of delamination. The bending tests for the specimens roll bonded at 950 ^{o}C did not show any fractures. Furthermore, no delamination was observed in the test.

Cross-section optical micrographs of the cold rolled plates for the roll bonding temperatures of 650 ^{o}C, 800 ^{o}C and 950 ^{o}C are presented in Figs. 5, 6 and 7, respectively. A thickness constraint was observed in all the plates and it is related to the ingot central porosity (see Fig. 1). Delamination between the zircaloy cover plate and the monolithic fuel meat was noticed for the plate roll bonded at 650 ^{o}C (letter C in Fig. 5). The presence of a perpendicular discontinuity between the fuel meat and the zircaloy-4 frame was found in the roll bonding temperature of 950 ^{o}C (Fig. 7D). No delamination was found for the plate roll bonded at 800 ^{o}C.

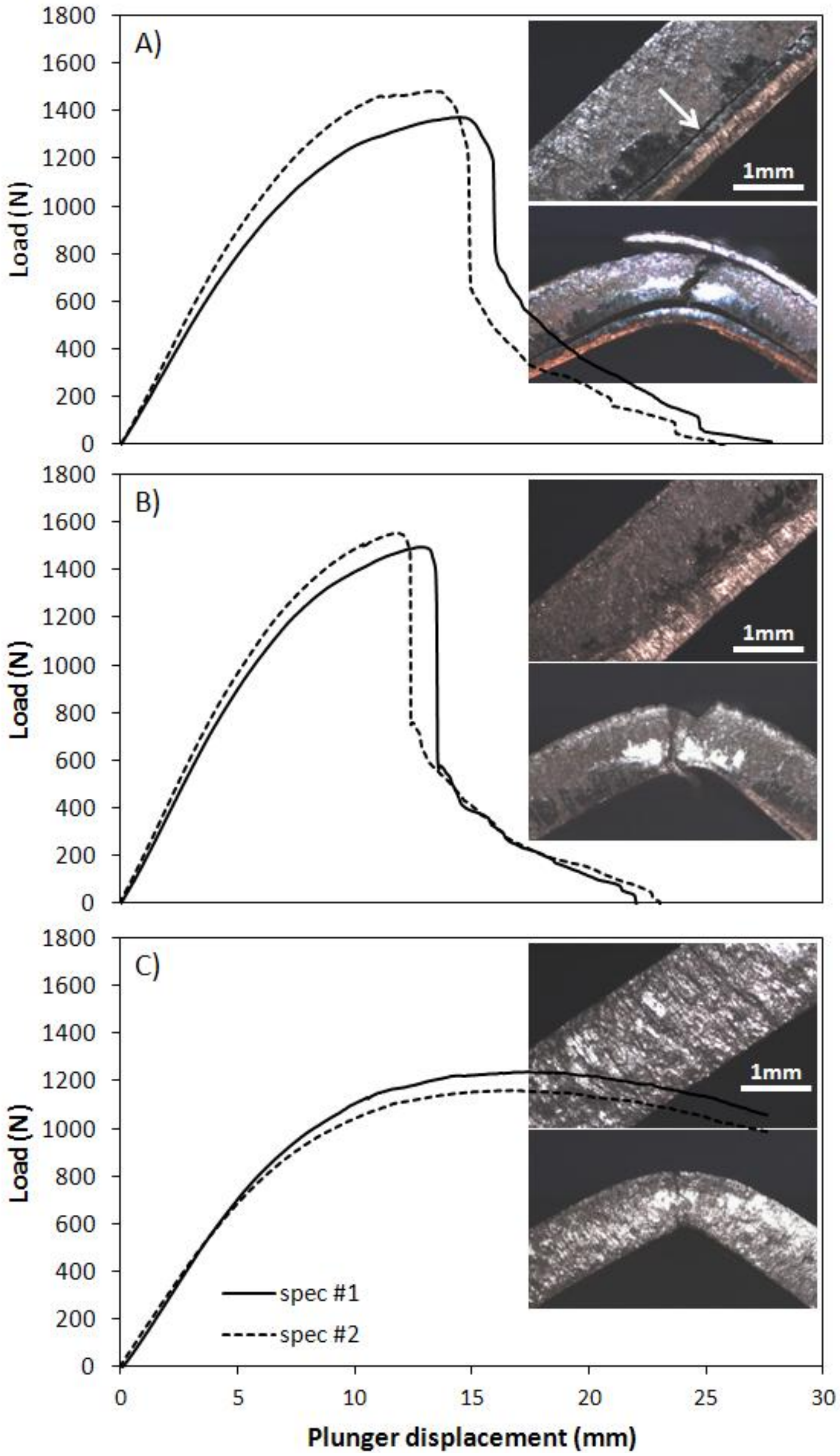

Figure 4. Load versus displacement curves for samples roll bonded at (A) 650 °C, (B) 800 °C and (C) 950 °C. The white arrow at the top indicates delamination occurrence.

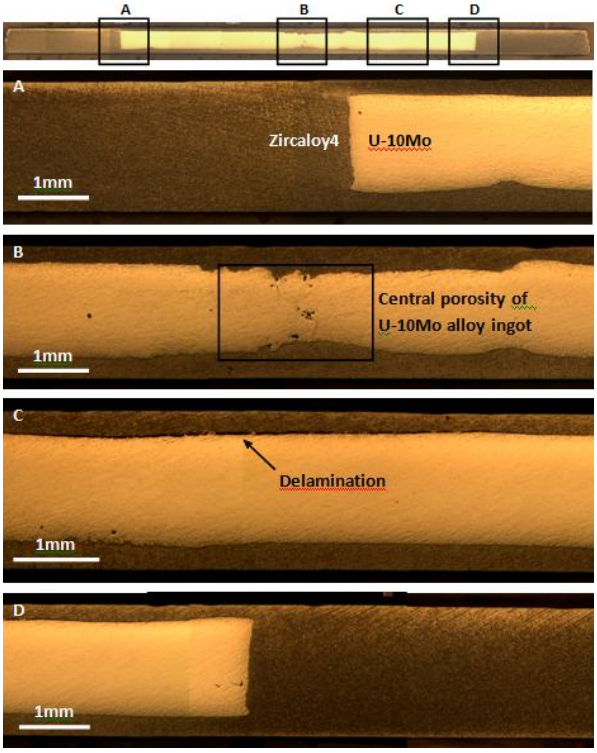

Figure 5. Cross-section of the roll bonding plate at 650 °C.

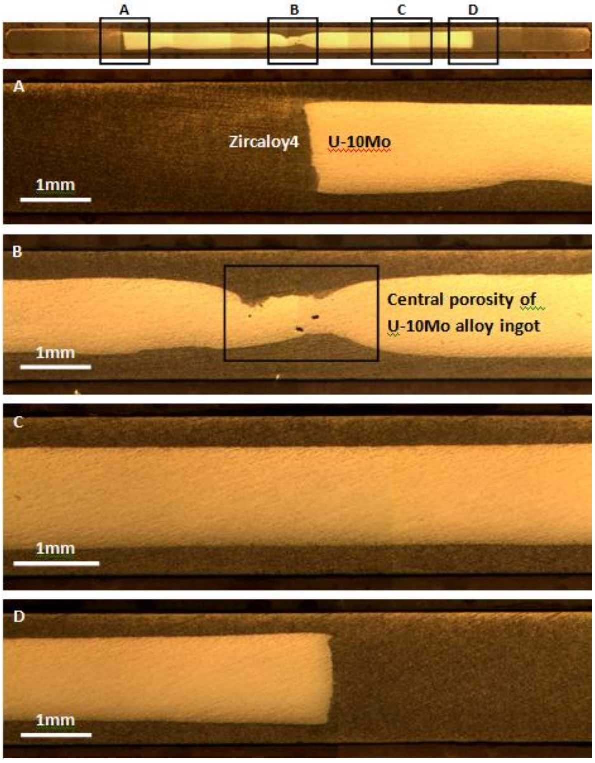

Figure 6. Cross-section of the roll bonding plate at 800 °C.

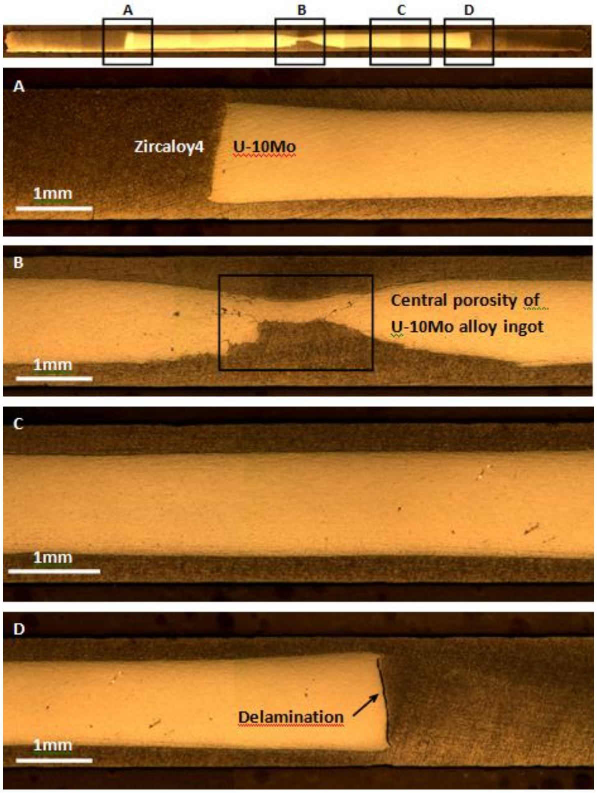

Figure 7. Cross-section of the roll bonding plate at 950 °C.

Despite the surface cleaning with pickling solution before mounting, the delamination occurrence may not be attributed only to the roll bonding temperature. Other factors can also be related to the lack of metallurgical adhesion. Oxidation during TIG welding or reheating may be cited as example and need to be investigated.

Examination of longitudinal section of the plates revealed the development of the “dog bone” defect in the monolithic fuel meat interfaces, as shown in Fig. 8. The same phenomena, but with a much lesser intensity, can be noticed at the cross-sections showed in Figs. 5, 6 and 7. This occurrence is associated with the mechanical strength differences between fuel meat and cladding, being the fuel meat in the monolithic form [22]. For 650 °C and 950 °C roll bonding temperatures, the “dog bone” build up was more intensive. This phenomenon can be associated to the lower reaction force exerted by the adjacent softer cladding material, which led to the meat thickening in its ends. In all cases, the U-10Mo meat was exposed in the plate surface. Additionally, delamination was noticed in the longitudinal sections for the three temperatures, unlike the cross-section evaluation, where the 800 °C roll bonding temperature presented no delamination occurrence.

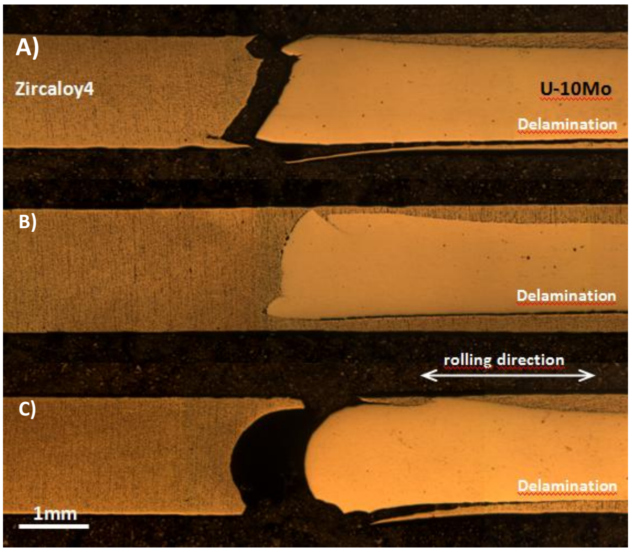

Figure 8. Micrographs for longitudinal direction of the roll bonded plates at (A) 650 °C, (B) 800 °C and (C) 950 °C.

Thickness measurements of the plates were performed in a light microscope. In the Fig. 9 is presented an example of the plate roll bonded at 800 °C. Fig. 10 summarizes the total thickness reduction in roll bonding and cold rolling processes as well as the specific values achieved in the cover plates and the fuel meat, for the three roll bonding temperatures investigated. It is clearly noticed that roll bonding at 650 °C resulted in a higher difference in the thickness reduction imposed to the cladding and fuel meat in relation to the total thickness reduction of the plate, whereas the roll bonding temperature of 800 °C presented the smallest difference. The difference increased again when the roll bonding temperature was set up in 950 °C.

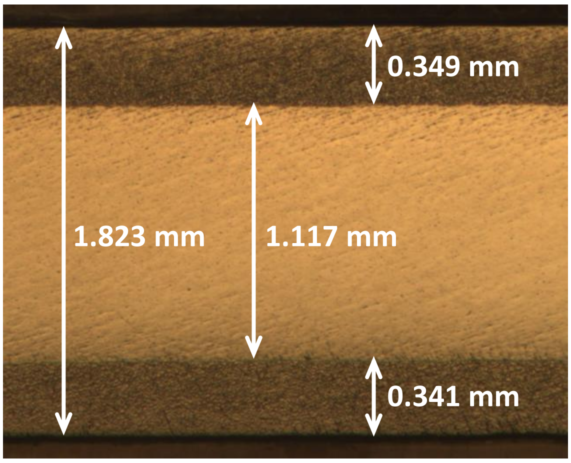

Figure 9. Thickness measurements in the cross-section of the monolithic fuel plate roll bonded at 800 °C.

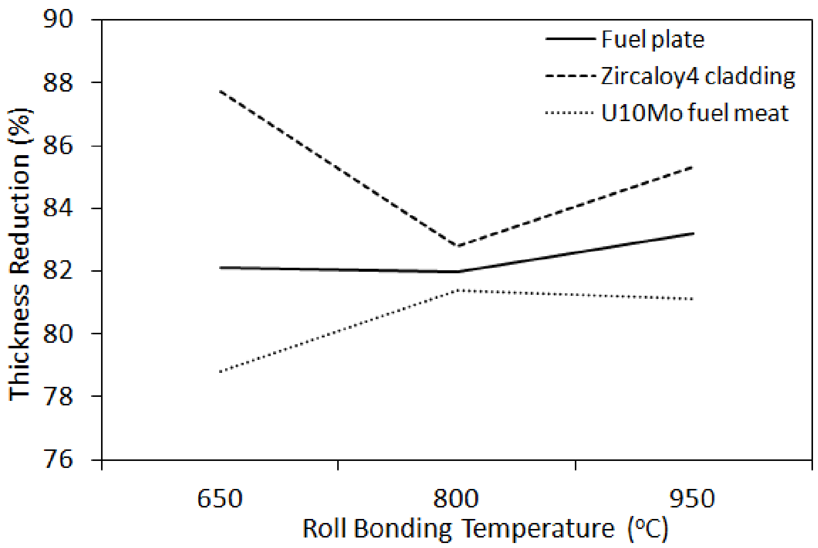

Figure 10. Thickness measurements in the cross-section of the monolithic fuel plate.

The mechanical properties discrepancy among the materials employed in the plate processing is the main reason for this behavior in the thickness reduction of the cladding and the fuel meat. It can be proposed that at 650 ^{o}C, the U-10Mo meat offered a much higher resistance against plastic deformation in comparison with the zircaloy4 cladding, which led to the higher contribution to the latter in the total thickness reduction. At 950 ^{o}C, the zircaloy4 cladding presented a remarkable decrease in its mechanical strength, offering a weaker resistance to plastic deformation.

The increasing difference in the thickness reduction of the cladding and the fuel meat, in comparison with to the total thickness reduction, for the lowest and the highest roll bonding temperatures suggests the existence of an optimal temperature for roll bonding process which is close to 800 ^{o}C. The determination of the optimal temperature range for the roll bonding process is essential for finding the initial dimensional parameters of the plate elements, which reduces the need of an increased amount of preliminary tests to define the initial dimensions, if a change in the product specification is proposed.

4. CONCLUSIONS

Roll bonding at 800 ^{o}C showed to be the best option for the continuity in the fabrication process development of monolithic nuclear fuel plates based on U-10Mo fuel meat and zircaloy4 cladding.

No delamination was noticed in the bending tests specimens of the plate roll bonded at that temperature, as well as in the metallographic investigation of the plate cross-section. The thickness reduction of the cladding and fuel meat was closer to the total thickness reduction among the roll bonding temperatures investigated.

Despite these better results, the “dog bone” defect and delamination were noticed at the longitudinal end of the U-10Mo fuel meat, leading it to be exposed at the plate surface.

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