HIGH DENSITY FUELS USING DISPERSION AND MONOLITHIC FUEL

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ABSTRACT

Fuel plates used in high-performance research reactors need to be converted to low-enrichment uranium fuel; the fuel option based on a monolithic formulation requires alloys to contain 6 – 10 wt% Mo. In this case, the fuel plates are composed of the metallic alloy U-10Mo surrounded by a thin zirconium layer encapsulated in aluminum cladding. This study reviewed the physical properties of monolithic forms. The constraints produced during the manufacturing process were analyzed and compared to those of dispersed fuel. The bonding process used for dispersion fuels differs from the techniques applied to foil bonding used for pure alloys. The quality of monolithic plates depends on the fabrication method, which usually involves hot isostatic pressing and the thermal annealing effect of residual stress, which degrades the uranium cubic phase. The preservation of the metastable phase has considerable influence on fuel performance. The physical properties of the foil fuel under irradiation are superior to those of aluminum-dispersed fuels. The fuel meat, using zirconium as the diffusion barrier, prevents the interaction layer from becoming excessively thick. The problem with dispersed fuel is breakaway swelling with a medium fission rate. It has been observed that the fuel dispersed in aluminum was minimized in monolithic forms. The pure alloys exhibited a suitable response from a rate at least twice as much as the fission rate of dispersions. The foils can support fissile material concentration combined with a reduced swelling rate.

1. INTRODUCTION

Facilities, in which radioisotopes such as Mo-99 are manufactured, are required to use low-enrichment fuels. In this regard, fuels produced with purely metallic alloys can provide the necessary structure and composition. Adherence to the enrichment level limit (20% of U-235) necessitates uranium-molybdenum (UMo) alloys to contain Mo in the range of 6 to 10 wt%. Although U-10Mo is a more effective composition, the formulation adopted for European reactors is U-7Mo. In the last decade, reactors worldwide have employed highly enriched fuels to produce over 95% of the Mo-99 used in about two-thirds of all procedures involving nuclear medicine. In these studies, U–Mo dispersions have shown elevated densities of fissile isotopes, e.g., U-235 at 8 gU/cm^3.

However, monolithic U(Mo) can provide the highest density value of up to 16 gU/cm^3. The conditions appropriate for maximizing the flux must also minimize enrichment and maintain an acceptable cycle length [1]. In Brazil, the National Nuclear Energy Commission (CNEN) has supported and supervised isotope production to satisfy public demand. The research reactor IEA-R1, which has been used to supply radioisotopes, is currently in its 60th year of operation. The operation of the facility is an attribution of the Nuclear and Energy Research Institute (IPEN) located in São Paulo. One of the tasks assumed by this reactor is radioisotope production for medical purposes to meet public health needs [2]. In this study, we compared the behavior of dispersions and monolithic forms with an enrichment value below 20%. In monolithic fuels, the meat is comprised of a single high-density fuel foil based on pure metallic uranium-molybdenum alloys. The pure form of a UMo alloy foil is surrounded by a zirconium layer and assembled in aluminum cladding. The metallic UMo showed a reduced interfacial area between the fuel and cladding coupled with reasonable swelling. Additionally, our analysis includes a discussion of the advantages and drawbacks of dispersions in contrast to monolithic fuels. The experimental tests revealed defects induced by irradiation, such as the delamination of the aluminum. The strategies for mitigating delamination are based on the composition or design of the fuel. Many diffusion barrier candidates have been researched and improved results have been observed with the addition of Zr, Nb, Si, Mo, and ZrN. However, the best results were obtained with zirconium [3].

2. BACKGROUND

2.1. U-MO Fuel Overview

In the 1950s, research towards developing fuel based on uranium-molybdenum (U-Mo) alloys focused on research reactors in the U.S. and Russia. Beginning in 1960, five pulsed reactors, such as Godiva IV, used U-Mo alloys with Mo content higher than 10 wt%. During 1978, the Reduced Enrichment for Research and Test Reactors (RERTR) was initiated by the U.S. Department of Energy (DOE). The objective of RERTR is to create technology for reducing the use of highly enriched uranium (HEU) to mitigate nuclear risk. Myriads of tested U-Mo alloy specimens have shown that they can convert high-performance research reactors (HPRR) [1]. Currently, an effort sponsored by global plans to reduce the amount of fuel enrichment has also led to monolithic fuels being considered. A strategic plan signed in 1993 requires radioisotope facilities to employ low-enrichment fuels for Mo-99 production. Fuels produced with pure metallic alloys became an option capable of offering the structure necessary to manufacture Mo-99. The enrichment level, limited to 20% of U-235, requires the Mo content of U-Mo alloys to remain between 6 to 10 wt%. A more effective configuration would be U-10Mo; however, in European reactors formulations have been adopted based on U-7Mo. Currently, around seven research reactors are using HEU fuels to supply the Mo-99 demanded globally.

2.2. Dispersion Fuel Elements

The production of UMo fuels dispersed throughout an Al-matrix is limited because a rolling process is employed. Currently, the uranium content of the fuel is restricted to a volume fraction of less than 55% in the meat. The classes of fuel dispersion based on UMo (7–10 wt% Mo) continued to be proposed for the conversion of HEU to LEU. The use of dispersion is not adequate for converting an HPRR to use LEU. This is because during normal operation,

the fuel plate may swell due to the build-up of fission gas at the grain boundaries of fuel dispersion. The interaction layer (IL) causes performance loss to fuel dispersion with low thermal conductivity. The major problem is the growth of the IL, which depends on a few conditions, such as the fission rate and temperature, and is influenced by the irradiation time. The preferred material for cladding manufacturing is Al6061 and in some cases, zircaloy. The conversion to medium-power research reactors has been successful using dispersions of LEU fuels, such as the silicide U_3Si_2 and U_3Si. In practice, the produced power exceeds 10 MW in HPRR reactors. The LEU specifications can support a peak fission density of 7×10^{21} fission/cm^3 and power density of 40 kW/cm^3 [4].

2.3. High-Density Fuels

High-density uranium is essential to identifying monolithic fuels. HPRRs need a minimum uranium density to convert fuels with a density exceeding 8.5 gU/cm^3 without severe loss of their scopes [5]. In practice, high-density fuels, such as LEU, imply that the fissile density must exceed 8 gU/cm^3. In contrast, pure metallic alloys, which have a uranium density of 15.6 gU/cm^3, can convert any HPRR to LEU, at the same capacity. The concentration adopted for molybdenum varies from 7 to 10 wt%. Molybdenum should retain the gamma phase at low temperatures; however, it is necessary to increase the concentration of the Mo-95 isotope, which has a significant capture cross-section, resulting in a decrease in the fission rate.

2.4. Interaction layer

In dispersed fuels, silicon is included in the aluminum matrix in an attempt to avoid swelling. In a UMo/Al dispersion, inserting silicon or niobium is necessary in order to reduce the size of the IL [6]. However, the use of a diffusion barrier is a more effective method to mitigate irradiation effects. The barrier applied to U-Mo/Zr is a thin layer of zirconium intended to eliminate the interactive area between the fuel and Al matrix [7]. The growth of the IL is correlated with fuel dispersion such as U_3Si and U_3Si_2. Both Eq. (1) and Eq. (2) are used to express the IL thickness.

$$Y^2_{(U3Si)} = 4.3 \times 10^{-6} F^{0.5} \exp\left(-\frac{55700}{RT}\right)t \qquad (1)$$

where Y is the thickness of the layer in μm; F is the fission rate (f/cm^3-s); T is the temperature in K, t is the irradiation time in s, and R = 8.314 J/(mol-K).

$$Y^2_{(U3Si2)} = 2.2 \times 10^{-8} F^{0.5} \exp\left(-\frac{40600}{RT}\right)t \qquad (2)$$

Alternatively, niobium or molybdenum, in addition to zirconium, have been used in the barrier. The thickness of the Zr/U–Mo diffusion barrier increases by Arrhenius' law as a function of temperature. Eq. (3) provides the numeric fit for the IL thickness defined as U10Mo.

$$Y^2_{(U10Mo)} = 2.6 \times 10^{-8} F^{0.5} \exp\left(\frac{3850}{T}\right)t \qquad (3)$$

Fig. 1, shows a comparison between the IL thickness of dispersed fuels and pure metallic alloys as a function of the fission density for the alloys U_3Si-Al, U_3Si_2-Al, and U10Mo-Al.
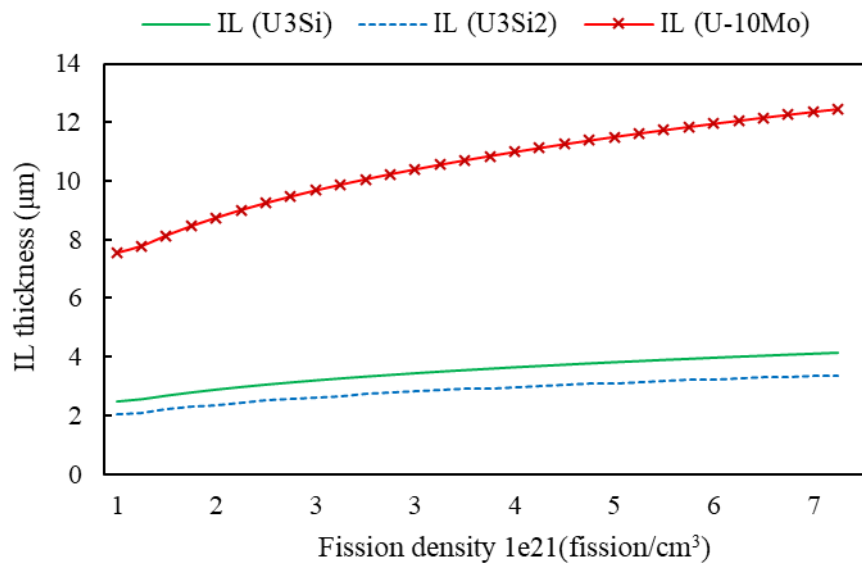

Figure 1: Calculated IL thickness for U_3Si, U_3Si_2, and U10Mo [6], [7]

3. U–Mo SYSTEM DESCRIPTION

At temperatures below 660 °C, uranium is highly anisotropic due to its low-symmetry orthorhombic crystal structure (α-phase). The tetragonal (β-phase) is stable between 668 and 776 °C, and the cubic (γ-phase) is stable above 776 °C. However, transition metals such as uranium allow the metastable retention of the cubic gamma phase below the equilibrium isotherm. The stability of the metallic alloy increases as a function of the weight fraction of Mo. The monolithic UMo meat can yield a fissile density of up to 16 gU/cm^3. Currently, research efforts are being focused on enhancing the outcome obtained from programs such as the RERTR..

3.1. Fabrication of Monolithic Fuel Plates

The first method to obtain monolithic plates consists of applying hot rolling to an U10Mo ingot. This reduces the thickness of the fuel alloy by approximately ten times, because rolling occurs at an elevated temperature. The disadvantages of these procedures are that they are expensive [8] and time intensive and generate substantial quantities of waste product. Experiments performed with fuel plates have shown that a few key factors need to be considered to improve the standard roll-bonding methods for suitable conditions to be reached. Mechanical stability is an essential aspect of a metallic alloy created for a fuel plate. The manufacturing process of a small alloy may involve four stages. First, uranium and molybdenum may be heated to a temperature of 1300 to 1500 °C. Obtaining the required homogeneous alloy with molybdenum is not a simple task because of the high melting point of molybdenum (2,623 °C). Casting is performed to reduce the size of the foil via hot and cold rolling. The second stage consists of the foil rolling process, which allows the material to be heated to 650 °C in the air without oxidization. The third phase occurs after decanting;

during this stage, the fuel foil is bonded with two layers of zirconium and is sandwiched between two sheets of aluminum [9].

3.2. Hot Isostatic Pressing

Metallurgical technology suggests using two similar practices to fabricate the plates: friction stir welding (FSW) and hot isostatic pressing (HIP). Initially, the fuel plates were produced by friction bonding of the uranium–molybdenum foil being positioned between two aluminum plates. Friction bonding is part of the natural evolution of the friction stir welding process. The diffusion bonding process, occurring at the atomic level, takes place when two objects are heated under high pressure. Bonding and diffusion take place at the interface between the fuel and the cladding [10].

3.3. Transient Liquid Phase Bonding

After a few thermic cycles, the temperature reaches 425 °C, at which the hot co-rolling process used to fabricate monolithic fuel plates can be expected to yield fractures. The use of a bonding method prevents cracks between the fuel and cladding. These cracks are localized at the center of the fuel plate and in the interface between Al6061 and the Zr barrier. To overcome the discontinuous bonding surfaces associated with diffusion bonding, the friction stir welding method was employed during the transient liquid phase bonding (TLPB). TLPB creates a fluidic phase, which spreads across the interface and relies on the interlayer material to diffuse into the bonding interfaces and join the elements together. TLPB uses a eutectic mixture of silicon and aluminum at 573 °C, whose temperature is lower than the melting point of aluminum at 660 °C. Monolithic alloys are more expensive than dispersed fuels due to the manufacturing process [11].

3.4. Irradiation Behavior of Monolithic Alloy

In the fuel plates composed of UMo alloy dispersed in aluminum, considerable swelling was detected at half irradiation cycles, or over 1×1021 fission/cm^3. The breakaway swelling developed a span due to the bubble released after the highest swelling rate. The reactions produced in the fuel and cladding created undesirable metallic composites in the IL. Radiation-induced sintering is one of the factors that can reduce porosity and swelling [12]. In monolithic fuel, UMo foils form a laminate structure and bond with aluminum cladding in order to reduce the inherent problems of dispersion. Several irradiation tests, conducted with pure alloys such as UMo, could achieve a long burn cycle with stable swelling at typical operating temperatures below 250 °C. The concentration of fuel such as LEU should be limited to a composition of 10 wt% Mo. Metallic fuel in the uranium gamma phase may increase the irradiation stability, mechanical properties, and corrosion resistance [13]. Regardless, the radiation may induce the decomposition of the gamma phase by transformation ($\gamma \rightarrow \alpha + \gamma'$).

3.5. Recrystallization Kinetics

Recrystallization develops on long burn cycles, when the UMo meat exhibits the effect of grain subdivisions enhancing fission gas bubble (FGB) swelling. FGB depends on a few

features, such as the fuel composition, fuel type, manufacturing process, and fission rate. The Avrami equation describes the transformation reaction; phase alterations might explain the recrystallization kinetics model for the UMo alloys during irradiation. Transformation kinetics define the crystal changes of the U-Mo system in the uranium-rich region of the phase diagram. Both dispersion and foil fuels may exhibit recrystallization ability, based on the fuel grains being subdivided. Fig. 2, presents the recrystallized volume proportion [14].
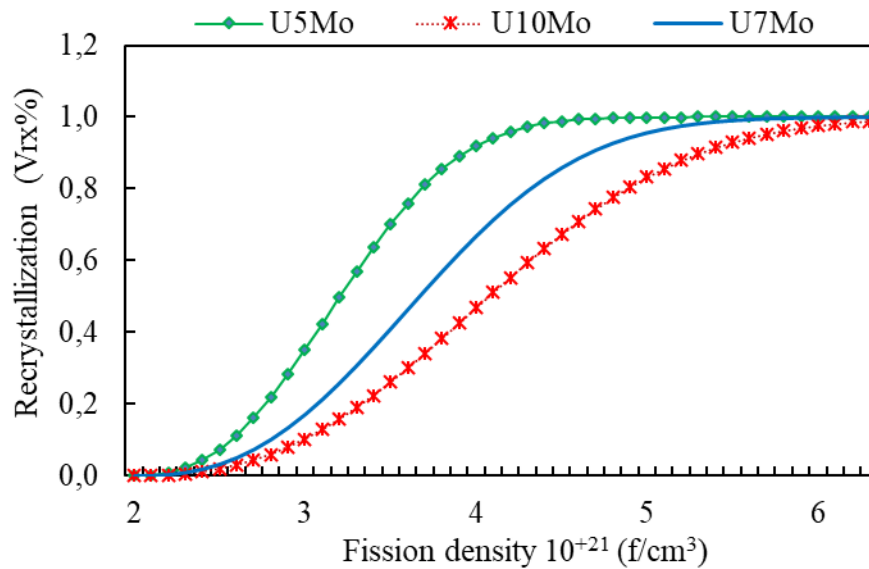

Figure. 2: Volume fraction of recrystallized grains

4. ANALYSIS OF THERMOMECHANICAL PROPERTIES

The Neumann–Kopp formulation allows the calculation of the specific heat capacity for fuel dispersion by using the law based on fraction contents. The U10Mo monolithic alloys, using the Al6061-T6 as cladding, add a zirconium barrier. Dispersion alloys have high heat capacity due to their chemical composition. The dispersion-fuel concept contains a significant fraction of aluminum. The specific heat of the fuel meat U10Mo, was represented as Cp, (J/Kg-K) despising irradiation effects, as a function of temperature in K, where, Xi (i = Al, Zr, UMo) are the atomic contributions produced by all the phases of the fuel alloy. Equation (4) exhibits the Neumann–Kopp law applied to the heat capacities of fuel meat. The polynomial best fit produced correlations given as a function of temperature, for U10Mo in Eq. (5) and U_3Si/Al in Eq. (6). Equation (7) shows the heat capacity of Al6061-T6 [15-18].

$$Cp = Cp_{Al}\chi_{Al} + Cp_{Zr}\chi_{Zr} + Cp_{(UMo)}\chi_{(UMo)} \tag{4}$$

$$Cp(U10Mo) = 13.75 \times 10^{-5}T^2 - 10.18T + 161.3 \tag{5}$$

$$Cp(U_3Si_2) = 10.48 \times 10^{-2}T + 170.4 \tag{6}$$

$$Cp(Al6061) = 3.28 \times 10^{-5}T^2 + 0.4169T + 721.4 \tag{7}$$

4.2. Thermal Conductivity

The thermal conductivity of the meat of the fuel may be calculated by applying the Neumann–Kopp model. The thermal conductivity increases as a function of the dispersion alloy temperature [16]. Equation (8) represents the thermal conductivity of uranium for the range $255 < T \leq 1173$ K. Equation (9) reproduces the conductivity of Mo, where T is in the range of $300 < T \leq 800$ K. Equation (10) represents the thermal conductivity of Al6061-T6 and is valid for $300 < T \leq 873$ K.

$$k(U)_{(U)} = 5.907 \times 10^{-6} T^2 + 1.591 \times 10^{-3} T + 21.73 \tag{8}$$

$$k(Mo) = -4.0 \times 10^{-2} T + 150.0 \tag{9}$$

$$K_{(Al6061)} = 0.01972 T + 16.739 \tag{10}$$

The formulation considers the effects of aluminum content on fuel meat and computes each contribution individually. The fuels are composed of different atom fractions of molybdenum in the fuel plate, which leads to the individual results. The thermal conductivity must increase as the temperature rises. The fitted thermal conductivity curves are plotted in Fig. 3, as a function of the temperature of the UMo alloy.
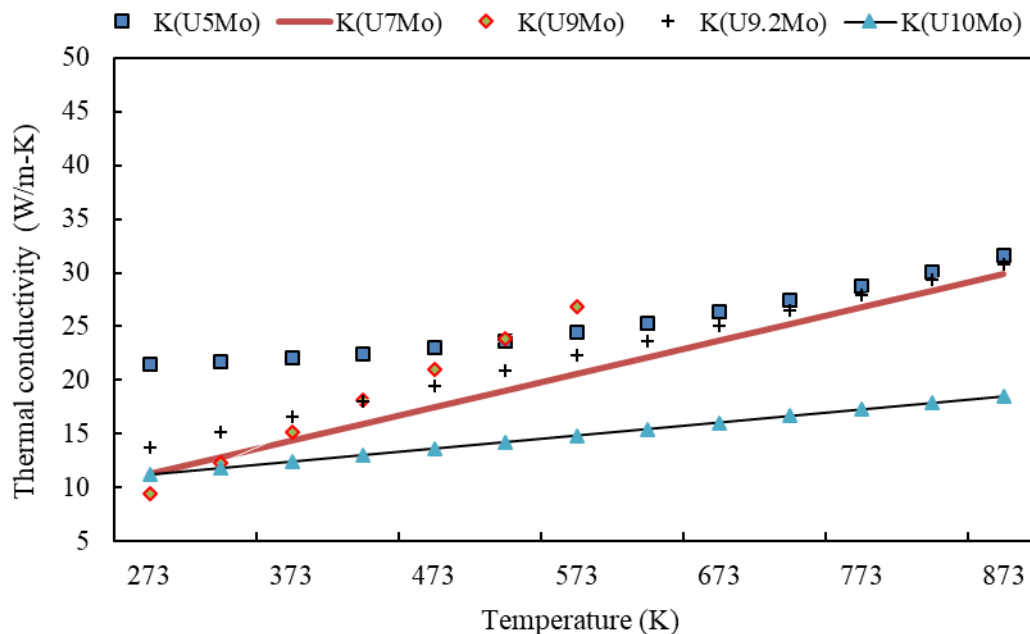

Figure 3: Thermal conductivities of U5Mo, U7Mo, U9Mo, and U10Mo [15-18]

The UMo fuel dispersions with lower molybdenum content, of up to approximately 5 wt%, exhibit higher thermal conductivity compared to contents ranging from 10 to 14 wt%. The U_3Si_2 and U_3Si alloys show similar values of approximately 15 W/m-K, regarding thermal conductivity. Equation (11) shows an empirical correlation representing the thermal conductivities of dispersion. Equation (12) accounts for the case of the U10Mo monolithic

fuel, where K is the conductivity in W/m-K, and the temperature T in K is valid in the range of $300 < T \leq 873$ K.

$$k(U7Mo) = -1.213 \times 10^{-7} T^2 + 0.03116 T + 2.78 \qquad (11)$$

$$k(U10Mo) = -9.893 \times 10^{-6} T^2 + 0.05373 T - 4.116 \qquad (12)$$

4.3. Thermal Expansion

The fuel plates exhibit a high coefficient of linear expansion compared to UO_2 represented for λ in (µm/m-K). The thermal expansion produces volumetric distortions in the fuel plates and partially blocks the coolant channels. Equation (13) represents the linear coefficient of dilatation for the metastable phase γ-(UMo). Equation (14) shows the numerical fitting of the thermal expansion for dispersion, U7Mo/Al. Equation (15) represents the linear expansion of the U10Mo metallic alloy, which is valid in the temperature range of $300 < T \leq 873$ K. The thermal expansion of different fuels, given as a function of temperature [15-18], are plotted in Fig. 4.

$$\lambda_{(\gamma-UMo)} = 7.91T + 1.21 \times 10^{-2} \qquad (13)$$

$$\lambda_{(U7Mo\text{-}Al)} = 3.317 \times 10^{-8} T^2 + 6.351 \times 10^{-3} T + 16.3 \qquad (14)$$

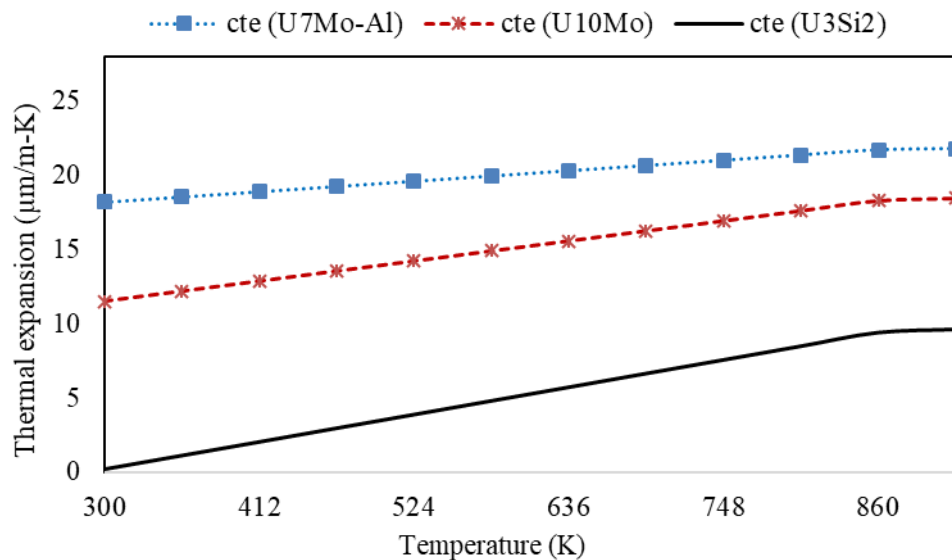$$\lambda_{(U10Mo)} = -1.595 \times 10^{-5} T^2 + 30.41 \times 10^{-3} T + 2.299 \qquad (15)$$

Figure 4: Thermal expansion of U10Mo, U3Si2 monolithic, and U7Mo/Al.

Pure metallic alloys such as U10Mo exhibit a coefficient of thermal expansion of 12 µm/m-K, due to the absence of Al. However, the Al6161 matrix indicates a superior ratio of approximately 27.5 µm/m-K, both at 300 K. The dispersion is approximately 18 µm/m-K due to the high coefficient of aluminum.

4.4. Density

The density and melting point of uranium are 19 g/cm^3 and 1132 °C, respectively, and the melting point of pure molybdenum is 2,623 °C. Using the Koop rule, we can calculate the thermal properties of the monolithic fuels' containing phases, such as (Zr+UMo) and (UMo). Equation (16) defines the empirical correlation based on the fitted line and represents the density of the metallic alloys. Equation (17) provides the numerical melting curve of the UMo alloys.

$$\rho_{(UMo)} = -0.085 \times \chi_{(Mo)} + 19.1 \qquad (18)$$

$$Ys_{(U7Mo-Al)} = 3.491 \times 10^{-4} T^2 - 7.715 \times 10^{-2} T + 517.2 \qquad (19)$$

$$Ys_{(U10Mo)} = 4.59 \times 10^{-6} T^3 - 7.177 \times 10^{-3} T^2 + 2.736T + 485.3 \qquad (20)$$

The meat of the fuel plates has a diamond pyramid hardness (DPH) in the range of 350 to 50 DPH. Hot-rolled dispersion plates for 1.5 h at 500 °C, fabricated with atomized powder, exhibit hardness of 290 DPH, with 17% uncertainty. Plates made from the machined powder process showed 350 DPH, with a deviation of 15%. However, as-rolled foils for 1 h at 650 °C must demonstrate the lowest hardness value. Equation (22) exhibits the hardness of solid uranium solution, given in GPa, as a function of molybdenum concentration ranging between 0 and 14 wt% [19].

$$Hd_{(\chi_{Mo})} = -3.748 \times 10^{-4} \chi_{Mo}^3 + 4.079 \times 10^{-2} \chi_{Mo}^2 - 0.6481 \chi_{Mo} + 6.206 \qquad (21)$$

The hardness of UMo alloys decreases because of the increasing temperature but tends to rise for molybdenum content within the range of 7-14 wt%. Fig. 5, presents the modulus of the elasticity curve, in GPa, as a function of the molybdenum weight concentration [19].
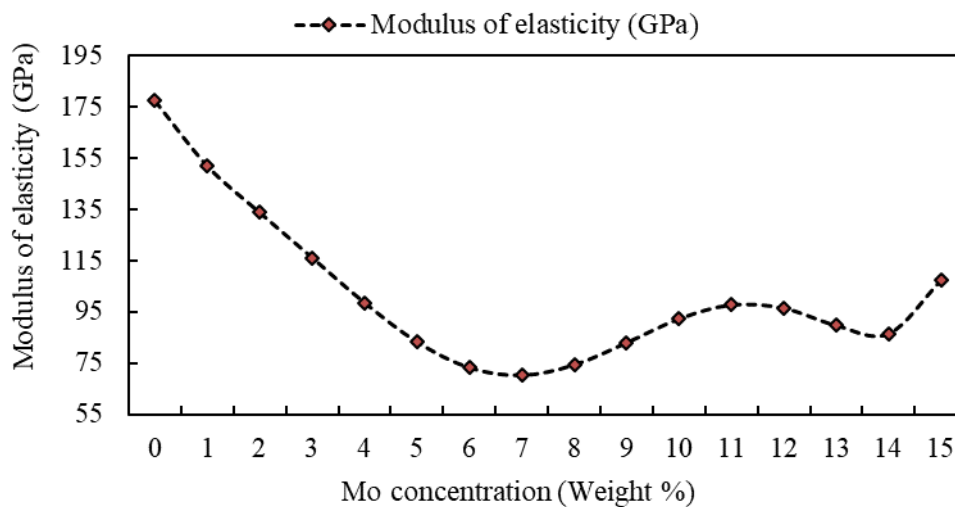

Figure 5: Elastic modulus of (U-Mo) as a function of the Mo concentration

4.6. Swelling

The swelling level is a function of temperature, fuel density irradiation, and concentration of fissile uranium. In experiments, uranium silicide fuels with a density of 4.8 gU/cm^3 and 6% swelling to a fission density of 10^{21} f/cm^3, burn 50% to 80% of atoms. The increase in the thickness of the plate is around 3%. Equations (20) and (21) display an empirical correlation describing the swelling given as a function of the fission rate. In the diffusion type, U$_3$Si$_2$/Al shows cumulative swelling because of the burn cycle. Eq. 21 gives the empirical correlation, which is the swelling as a function of the burnup Bu. The swelling of the dispersion is double the swelling of uranium dioxide.

$$Sw_{(U3Si2-Al)} = 7.111 \times 10^{-6} Bu^2 + 7.022 \times 10^{-4} Bu + 11.11 \times 10^{-4} \qquad (23)$$

$$Sw_{(UO2)} = -2.839 \times 10^{-6} Bu^2 + 7.443 \times 10^{-4} Bu + 6.642 \times 10^{-4} \qquad (24)$$

The swelling is a function of the fission densities; in general, at 3×10^{21} f/cm^3, the swelling is ~20%, and at 7×10^{21} f/cm^3, the swelling is 44%. The manufacturing process of monolithic fuel plates induces residual stress. The thermal cycle model implements heat-treatment, hot rolling, and bonding. The metallic alloys are maintained at approximately 500 °C for 1 h, and then cooled at approximately 20 °C. The recrystallization indicated a transition to 2.0×10^{21} f/cm^3 to U7Mo and presumed the shift toward 3.0×10^{21} f/cm^3 to U10Mo defined in the experiments.

5. CONCLUSION

The analysis of the characteristics of fuel types demonstrated the physical properties required to build a new fuel plate system based on uranium–molybdenum alloys. The aluminum fraction in the fuel meat causes a few differences between foil-based fuels and dispersions. The correlations presented in this paper can predict the thermal properties of unirradiated plates. The plastic properties of the metallic alloys are superior to those of dispersions. At typical operating temperatures, the severe disadvantage of UO$_2$ is a lower thermal conductivity ranging between 5-2 W/m-K compared to that of U$_3$Si$_2$, which ranges between 9-20 W/m-K. The U$_3$Si$_2$ pellets have a uranium density of 11.3 g/cm^3 compared to 9.68 g/cm^3 of UO$_2$.

Some of the problems presented by high-density fuels, i.e., a swelling knee and diffusion layer growth, are expected to be addressed shortly. The diffusion layer that is produced is avoided by using zirconium or silicon as a barrier, which should theoretically serve to reduce the undesirable effects. The pure metallic alloys can convert any fuel used in high-flux reactors to LEU enriched below 20%. Investigations have revealed a few candidates as substitution for aluminum cladding, notably zircaloy-4 and silicon carbide. A monolithic plate based on UMo alloys, manufactured by the HIP method, can consolidate the fuel quality and reduce the fabrication cost. Fuel performance is partially correlated with the metastable gamma phase, which is degraded by the high temperature and stress induced by the manufacturing process during fabrication. These shortcomings could be overcome by developing an efficient bonding process for the fuel plate and an innovative solution for the swelling rate.

ACKNOWLEDGMENTS

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