



Observations of liquid lithium uptake in a porous molybdenum foam

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ABSTRACT

Recent experiments with liquid lithium in magnetically confined plasmas have demonstrated improved plasma performance. These results have led researchers working on the National Spherical Torus Experiment to consider using a porous molybdenum foam and liquid lithium composite as a future liquid lithium divertor. In order to better understand the properties of this composite material, a small experimental apparatus was constructed at the University of Illinois to test lithium wetting uptake into the porous material. We report here results of the wetting behavior of a porous molybdenum foam with liquid lithium. Based on these observations, a simple model was used to estimate the thermal properties of the lithium infused porous material. Finally, the results of water-bath cleaning tests of the porous metal after lithium exposure are shown.

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1. Introduction

Lithium is considered an attractive PFC material for several reasons. As a low Z material, small amounts of lithium entering the plasma have far less impact on Z_{eff} than carbon or tungsten. Lithium surfaces also “getter” a large number of impurities, such as carbon and oxygen, further reducing the Z_{eff} of the plasma [1]. These effects were noticed to great effect in the Tokamak Fusion Test Reactor (TFTR) during lithium pellet injection experiments which generally improved machine operation [2,3].

The CDX-U device made several important studies of the use of liquid lithium PFCs [4–6]. Initial work studied the use of a liquid lithium rail moving on to a fully toroidal limiter. In general, plasma performance improved on CDX-U with an increasing area of exposed liquid lithium. Among the results of these experiments, a reduced Z_{eff} was shown when operating with the lithium tray. Increased electron temperatures and reduced loop voltages were also observed. Strongly reduced recycling, decreased MHD instability activity and strong pumping were also notable results [6]. The pumping is consistent with measurements indicating that liquid lithium will absorb hydrogenic species until it is volumetrically converted into lithium hydride [7].

The electromagnetic environment of the nuclear fusion reactor presents magnetohydrodynamic problems for any liquid metal introduced into the chamber. Experiments on the DIII-D tokamak saw expulsion of liquid lithium from the Li-DiMES experiment [8]. One solution to this problem is the use of a porous material

which will restrain the liquid metal against macroscopic motion. This was realized recently in the Capillary-Pore System (CPS) which used layers of molybdenum mesh as a wicking material [9]. This system was first tested on the Russian T11-M tokamak [9] and, more recently, on the Italian FTU device [10].

Following the successful implementation on smaller devices, researchers working on the National Spherical Torus Experiment (NSTX) are implementing a fully Liquid Lithium Divertor (LLD) [11]. One candidate material is a porous molybdenum foam manufactured by Ultramet [12]. The foam will act as a wicking structure pulling lithium to the surface and restraining it there against MHD forces.

In order to assess this material’s thermal capabilities, operating temperatures and reusability, a small experimental device was constructed at the University of Illinois (UIUC): the Study of Lithium Infusion in Porous Media experiment (SLIP). We report here a number of observations made working with the SLIP system characterizing the interaction between liquid lithium and the porous molybdenum foam. Section 2 describes the apparatus used for these experiments. Section 3 reports the results of our tests and the data gathered. Section 4 shows the calculation of the thermal conductivity and capacity of the composite material.

2. Apparatus and approach

Fig. 1 shows a schematic diagram of the SLIP test chamber and setup. Fig. 2 shows the Ultramet molybdenum foam sample on its mounting system as well as a close-up picture of the pre-lithium exposure foam. The blackened portions of the foam sample resulted from a prior test and this is discussed in Section 3.

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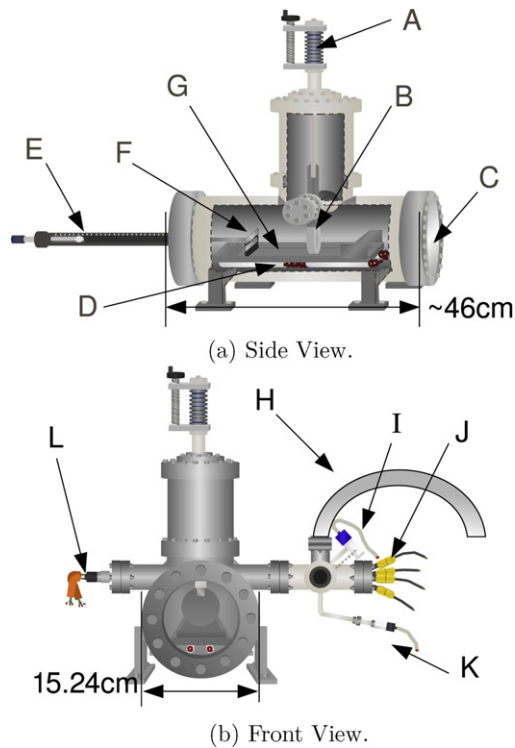


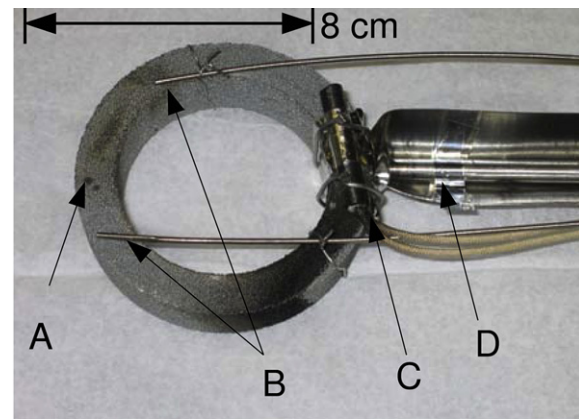
Fig. 1. Side- and front-view diagrams of the Study of Lithium Infusion in Porous media (SLIP) test stand. Labels correspond to (A) foam manipulator, (B) molybdenum foam sample, (C) window, (D) tray heaters, (E) slag tool manipulator, (F) slag tool and (G) tray. In the front view (b), labels correspond to (H) vacuum connection, (I) convectron pressure gauge, (J) thermocouple feedthru, (K) diaphragm pressure gauge, and (L) heater power feedthru. An additional sample cartridge heater is not shown in the diagram.

The system was designed for wetting tests at discrete temperatures. The major components consisted of the following items: the main vacuum vessel, a heated tray holding the lithium, a “slag tool” for cleaning the lithium once it is melted, a separate manipulator for raising and lowering the molybdenum foam sample into and out of the lithium, and pressure and temperature gauges. A digital camera was used to record the tests and viewed the system through the window.

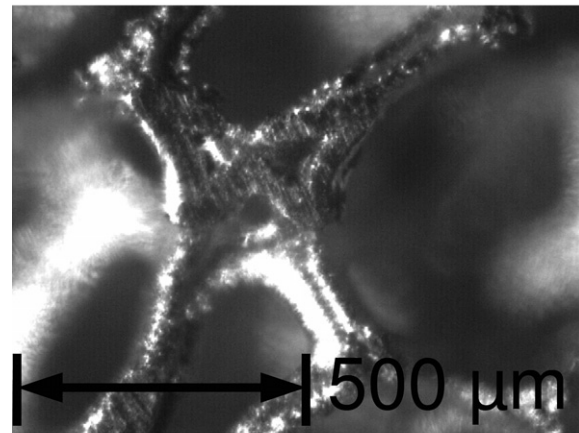
Due to the reactive nature of lithium, it was necessary to both clean the liquid surface and maintain it in a non-reactive environment. This was accomplished through the use of a slag tool and vacuum system. The vacuum chamber was made from 6” conflat chamber components enabling vacuum conditions as well as pressures slightly above atmospheric. Once lithium was loaded in the system, the chamber was repeatedly pump-purged with UHP argon and then operated with a steady argon flow maintaining 8000 Pa of pressure. Pumping was accomplished with a Welch belt-driven rough pump and a throttling valve. Pressure was monitored with a convectron pressure gauge which is accurate from the 0.1 Pa level to 700 Pa of pressure and a diaphragm pressure gauge which is accurate from atmospheric to about 133 Pa.

The slag tool consisted of a stainless steel metal screen affixed to the end of a linear motion feed-thru. As the slag tool was pushed through the molten lithium, the slag on the surface of the lithium was pushed forward and into a catch at the end of the tray. Due to the coarseness of the screen, the liquid lithium was able to flow beneath the slag and through the screen remaining in the tray. The result was a clean lithium surface ready for testing.

The foam sample was a right circular cylinder. The outer and inner diameters were 7.7 and 5.5 cm respectively. The height of the



(a) Foam Sample.



(b) Clean Foam.

Fig. 2. Pictures showing foam sample (a) mounting details and (b) a closeup of the molybdenum foam under optical microscope before lithium exposure. In (a), labels correspond to (A) the sample ring itself, (B) stainless steel sheathed K-type thermocouples, (C) cartridge heater element and (D) mounting support. The foam sample diameter is 7.7 cm. The width of the close-up picture is 1 mm.

cylinder was 2.1 cm and the sample weighs 60.7 gm total. The sample was reported to be 80% porous (void fraction) by the manufacturer.

The tray and foam sample were heated independently using resistive heaters. The tray was constructed from 0.25 mm thick stainless steel shimstock which minimized thermal conduction to the vacuum vessel. Resistive heaters were created by helically winding stainless steel wire (1 mm diameter) which is inserted into an alumina tube to provide electrical isolation. These heaters proved more robust in the lithium environment than commercially available options and operated with up to 10 A of input current (the maximum available). The heaters were resistant to lithium chemical attack and could attain high operating temperatures before failure. Heating power was controlled with variacs.

Thermocouples were attached in three main locations in the system. The first was positioned inside the tray along the bottom of the lithium pool such that the junction bead is directly beneath the foam sample. The second and third were both attached externally to the sample. This method was taken, as opposed to drilling into the foam, to avoid damaging the foam exterior as the interior was made of carbon which could react with the lithium.

Initially, the lithium and molybdenum foam were both brought to a steady temperature. Following temperature equilibration, the slag tool was used to remove any impurities from the surface of the lithium. The foam sample was then brought into contact with

the lithium and observations were made to determine if wetting was occurring. When no wetting was observed, the foam was retracted and then heated to a new, higher temperature.

In another set of tests following heater failure on the foam sample, the foam sample was left in contact with the lithium. Temperature was controlled with the tray heaters alone, though all thermocouples continued monitoring throughout the tests.

Once wetting had been achieved and the tests completed, the sample was allowed to cool and solidify and was removed from the system for inspection. A standard cleaning method developed at UIUC is to use a large water bath to react away lithium from metal parts. This method consisted of submerging the sample under water and waiting until evidence of chemical reaction (vigorous bubbling) ceased. The samples were examined under an optical microscope before and after cleaning.

3. Results

Fig. 3 shows a typical operating graph. Dip tests were carried out over the course of several experimental runs and showed a lack of wetting from 200 °C up to 300 °C. A dip test at 350 °C did show wetting, however during this run, a visible layer of lithium had been evaporated onto the molybdenum foam sample. Repeated dip tests up to 320 °C at a higher operating pressure to reduce evaporation also showed no wetting.

The second test method utilized (continuous contact with the lithium) was carried out with the lithium temperature ranging from 300–440 °C. While visible observations were made continuously, no sudden change in wetting was observed at any point. At 440 °C the lithium was removed and it was seen to have wicked into the material. This was exhibited by the ability to pull a liquid “bridge” of lithium out with the foam above the surface. When brought back into contact with the liquid pool below, the lithium instantly wetted the foam pulling itself into a “bridge” once again.

After allowing the sample to cool and any remaining lithium to solidify, the sample was removed for inspection. Distinct regions were visible on the sample. The bottom of the sample in direct contact with the lithium had a layer of lithium conforming to it. Tests on the first experiment run were conducted in a carbon rich environment (as a result of outgassing polymers) and resulted in a large blackened portion of the ring. On other runs, the portions that had been wetted had a visible change in lustre and also included a blackened portion, though not as significant as in the first run.

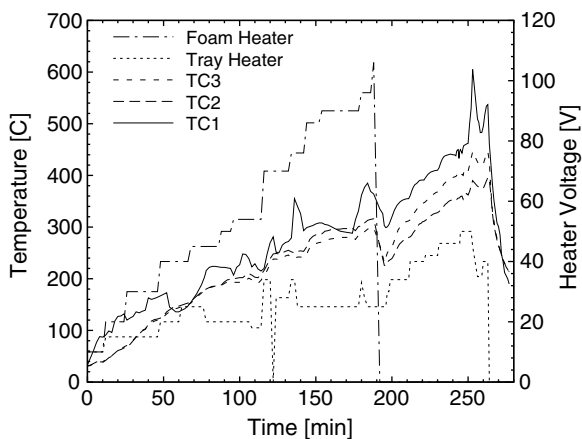


Fig. 3. Figure showing the heater voltages and thermocouple temperatures for the second run test. The drop in Foam Heater power at 190 min is due to foam heater failure. Time derivative method is applied with data between 195 and 245 min. TC1 corresponds to the liquid lithium and tray temperature, TC2 and TC3 are foam temperature measurements with different TC-liquid distances.

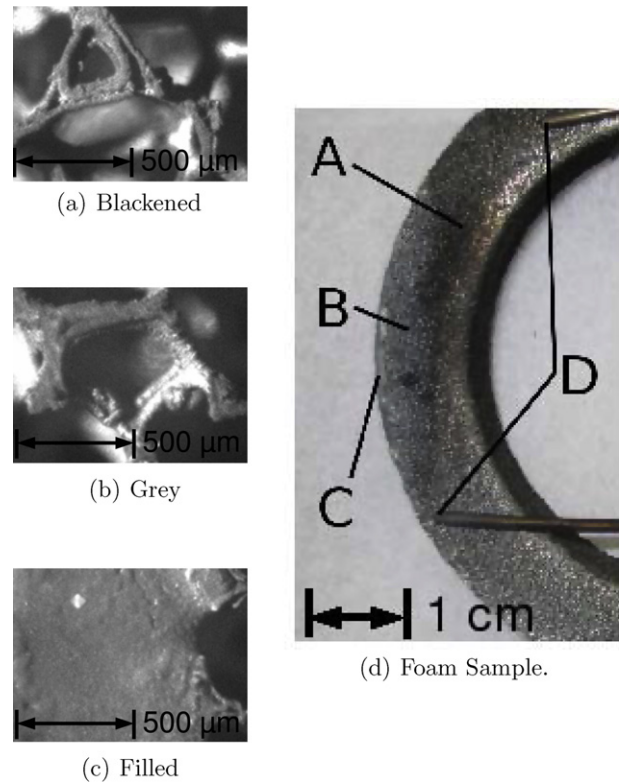


Fig. 4. Photographs of foam sample after liquid lithium exposure. Foam sample locations correspond to (A) blackened portion, (B) grey areas, and (C) bulk lithium/filled foam. (D) corresponds to thermocouples. Each microscope image (a–c) is 1 mm wide.

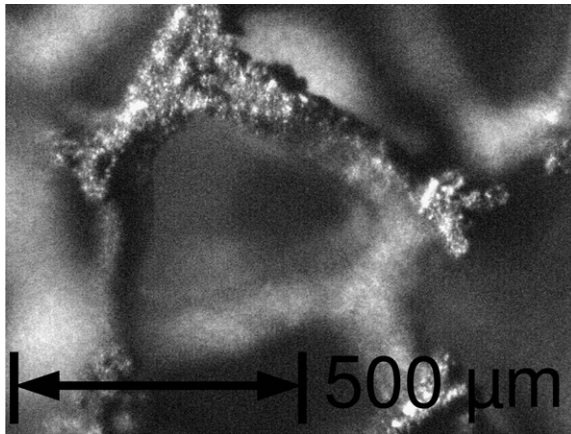
These regions along with optical microscope images are shown in Fig. 4.

The samples were subjected to a standard cleaning method which consisted of allowing the lithium to react away in liquid water. The samples showed the typical reactivity when placed into the water and no violent reactions were observed. After the reactions ceased, the samples were removed and inspected. All of the areas of the foam which were exposed to the lithium were returned to their original condition except for those which were blackened. These, instead, remained discolored though light mechanical rubbing improved the color a small amount. Upon examination under microscope, it was discovered that the blackened areas had a dark color, though raised areas of the foam had exposed metal. This is shown in Fig. 5.

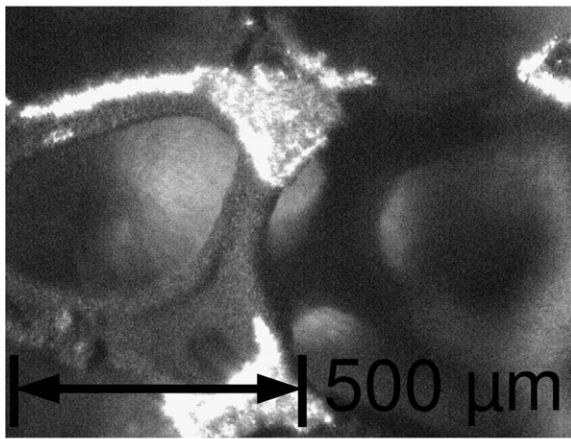
4. Theory and discussion

When visual inspection of the material failed to show when wetting occurred, another method was required. The level of saturation of a wicked material in a porous media has been determined by examining the bulk material properties [13]. This method has been performed with electrical conductivity measurements on columns of salt water solution in packed beds. We have made use of thermal property changes to determine when this transition took place.

A parallel thermal conduction model was used for the porous material. In this model, the effective cross-sectional area of the porous metal was the fractional percentage based on the porosity percentage. For the porous material tested, this yielded a 20% fraction filled with molybdenum. In the zeroth order model, we assumed that the lithium completely filled the pores taking up the



(a) Before Cleaning.



(b) After Cleaning.

Fig. 5. Optical microscope images of molybdenum foam blackened regions before (a) and after (b) water bath cleaning. Raised areas in the after image are clearly seen by the exposed metal (c.f. Fig. 2) Each image is 1 mm wide.

remaining 80% of the total conduction area. Using the method of thermal resistances [14] one we derived the change in bulk material properties between an empty state and a filled state. For the case of the thermal conductivity, k , the following calculation was made:

$$\frac{1}{R_{\text{eff}}} = \frac{1}{R_{\text{Mo}}} + \frac{1}{R_{\text{Li}}} \quad (1)$$

$$\frac{k_{\text{eff}}A_{\text{tot}}}{L} = \frac{k_{\text{Li}}fA_{\text{tot}}}{L} + \frac{k_{\text{Mo}}(1-f)A_{\text{tot}}}{L} \quad (2)$$

$$k_{\text{eff}} = fk_{\text{Li}} + (1-f)k_{\text{Mo}} \quad (3)$$

where subscripts eff, Li and Mo refer to the effective property and those of lithium and molybdenum respectively. The thermal resistivity is denoted R , A_{tot} is the total area, L is the thickness through which heat is conducted and f is the void fraction of the porous material. A similar method was utilized for the thermal capacity property. The results of this analysis are shown in Table 1.

The time dependent response of a plane-slab thermal conductor was described by the following equation:

Table 1
Table summarizing material properties used for thermal response estimates

Material	$\rho \frac{\text{kg}}{\text{m}^3}$	$C \text{ (J/kg-K)}$	$k \text{ (W/m-K)}$	$\alpha \text{ (m}^2/\text{s} \times 10^6)$	Ref.
Molybdenum	10240	251	138	53.6	[14]
Liquid lithium	508	4350	45	20.4	[15]
Porous Mo + Li	2454	930	64	28	Eq. (3)

$$\rho C_p A_{\text{tot}} L \frac{dT_1}{dt} = Q_{\text{tot}} = \frac{T_1 - T_0}{R_{\text{eff}}} \quad (4)$$

where ρ is the density, C_p is the thermal capacity, T_1 is the temperature of the slab and T_0 is a reference temperature. Eq. (4) again relies on the resistance model for heat transfer. The solution of this equation is elementary and can be stated in the following way:

$$T_1 - T_0 = C_1 \exp\left(\frac{t}{\rho C_p R_{\text{eff}} A_{\text{tot}} L}\right) \quad (5)$$

where $\rho C_p R_{\text{eff}} A_{\text{tot}} L$ is an effective time constant, τ for the thermal response of the system and C_1 is a constant of integration. An effective method of elucidating this behavior was to look at the logarithm of the temperature response and simplify the τ term and obtain a linear dependence on time:

$$\ln(T_1 - T_0) = \frac{\alpha}{L^2} t + \text{const.} \quad (6)$$

Where $\alpha = \frac{k}{\rho C_p}$ is the thermal diffusivity of the material. A change in the thermal properties of the material resulted in a change in this time constant. The expected difference between the empty foam and the lithium infused foam is calculated to be the following:

$$\frac{\tau_{\text{Mo}}}{\tau_{\text{Li+Mo}}} = \frac{\alpha_{\text{Mo}}}{\alpha_{\text{Li+Mo}}} = \frac{53.6 \times 10^{-6}}{28 \times 10^{-6}} = 1.9 \quad (7)$$

Examining the thermal data shown in Fig. 3, a region of applicability was seen from 195–245 min. During this time, there were several step changes in heater voltage with an accompanying exponential rise in temperature, as predicted by Eq. (5). Applying Eq. (6) to the region of interest several regions of linearity were shown in Fig. 6. The shift in value prior to each linear region was due to a brief period of temperature transition which was dominated by \sqrt{t} behavior in the liquid and foam separately, though was neglected in the present analysis.

As seen in Fig. 6, there were several regions of linear behavior. The arrow marks a location where the slope (proportional to α) changed without an accompanying change in heater voltage. Fig. 7 shows the values of these slopes and estimated error for comparison. There was significant uncertainty in the calculation, however the expected value for the ratio, 1.9 as given by Eq. (7), was within the error limits of all the measurements. With this in mind, we conclude that wetting and wicking occurred at the point indicated by the arrow in Fig. 6 and that the parallel conduction paths model discussed above was consistent with these test results.

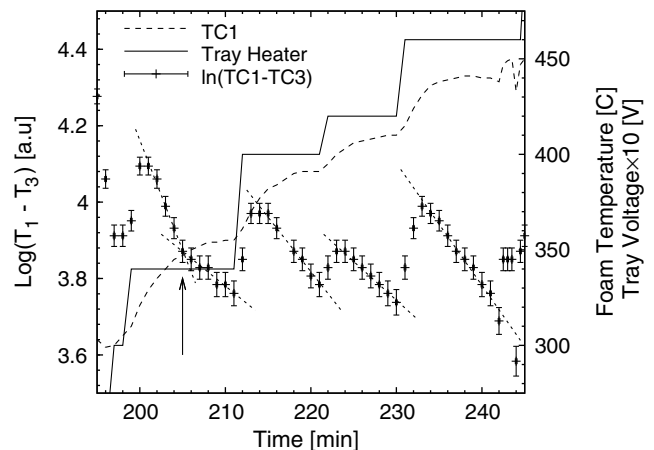


Fig. 6. Figure showing the logarithm of the temperature response and regions of linearity with time. Tray heater voltages and temperature of the foam are shown as well. Dashed lines indicate linear regression fits to the data. Arrow shows apparent change in slope absent heater alteration.

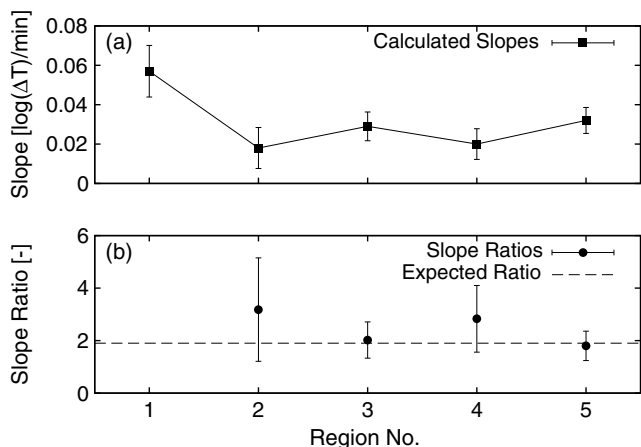


Fig. 7. Figure showing (a) calculated slopes for the five test regions as well as (b) the calculated and expected slope ratios. Expected value is calculated by Eq. (7) and falls within error limits for all ratios taken.

The wetting point corresponds to 205 min into the test with 347 °C and 300 °C in the lithium and foam measurement points respectively which form upper and lower bounds for when wetting occurs. While the dip test showed negative results at 320 °C, we note that the wetting angle in alkali liquid metals can be a time-varying quantity [16] and the second test method utilized continuous contact between the molybdenum and liquid lithium.

It was believed that the black material was from an external source and not leached from the foam. This seemed the most reasonable conclusion considering that clean metal had been exposed with light rubbing and there were external sources of carbon in the system during these tests. The bulk of the material was returned to its original state so we concluded that the water-bath method of cleaning was compatible with the porous molybdenum foam and can see no impediment to its use. The ramifications of such residues in the LLD application require further study.

5. Conclusion

An experimental apparatus has been constructed to examine the wetting behavior of a porous molybdenum foam made by Ultr-

amet. Two test methods were utilized: dip tests at discrete temperatures and a continuous test with increasing temperature. Visual inspection failed to yield the wetting temperature of the molybdenum foam, however analysis of the temperature data recorded during the test yields a wetting point between 300 °C and 347 °C. The thermal data was consistent with a parallel conductor thermal model for the lithium-infused porous metal. Finally, the water-bath cleaning method for removing lithium was shown to be successful with the exception of a mechanically-removable, black “soot” on the surface of the metal.

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