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Fabrication and Characterization of a Conduction Cooled Thermal Neutron Filter

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Abstract – *Installation of a conduction cooled thermal (low-energy) neutron filter in an existing domestic test reactor would provide the U.S. the capability to test new reactor fuels and materials for advanced fast (high-energy) reactor concepts. A composite consisting of Al_3Hf-Al has been proposed for the neutron filter due to both the neutron filtering properties of hafnium and the conducting capabilities of aluminum. Knowledge of the thermal conductivity of the Al_3Hf-Al composite is essential for the design of the filtering system. The present objectives are to identify a suitable fabrication technique and to measure the thermophysical properties of the Al_3Hf intermetallic, which has not been done previous to this study. A centrifugal casting method was used to prepare samples of Al_3Hf. X-ray diffraction and Rietveld analysis were conducted to determine the structural make-up of each of the samples. Thermophysical properties were measured as follows: specific heat by a differential scanning calorimeter, thermal diffusivity by a laser flash thermal diffusivity measuring system, thermal expansion by a dilatometer, and thermal conductivity was calculated based on the previous measurements. All measurements were acquired over a temperature range of 90°C - 375°C with some measurements outside these bounds. The average thermal conductivity of the intermetallic Al_3Hf (~7 at.% Hf) was found to be ~ 41 W/m-K for the given temperature range. This information fills a knowledge gap in the thermophysical properties of the intermetallic Al_3Hf with the specified percentage of hafnium. A model designed to predict composite properties was used to calculate a thermal conductivity of ~177 W/m-K for an Al_3Hf-Al composite with 23 vol% Al_3Hf. This calculation was based upon the average thermal conductivity of Al_3Hf over the specified temperature range.*

I. INTRODUCTION

The capability to conduct fast (high-energy) neutron irradiation tests in a domestic facility is needed to meet anticipated future nuclear fuel and materials testing needs.[1] A thermal (low-energy) neutron filtering test system installed in an existing thermal reactor provides a more affordable and near-term option than design and construction of a new fast-flux test reactor.[2]

Design requirements for the testing system included a fast (E > 0.1 MeV) flux of at least 1 x 10^{15} neutrons/(cm^2-s) with a fast-to-thermal flux ratio in the vicinity of 40.[3] It was assumed that typical experiments would consist of fuel specimens (~1 cm diameter) with axial heating rates up to 2.3 kW/cm and a total heat load from all specimens as high as 200 kW.

A detailed review of domestic and foreign reactors and accelerators resulted in the recommendation of Idaho National Laboratory's (INL's) Advanced Test Reactor (ATR) as the best host for such a system. The original design employed a gas cooling system at a cost of $80-100 million whose major cost driver was the pressurized helium used as the coolant. To reduce this cost, a novel concept was proposed that relies upon a neutron filtering material capable of both high thermal neutron absorption and high thermal conductivity. This solution is necessary

because while water is a cheap and effective coolant that reactors are already built to handle, it also thermalizes neutrons. Having a high thermal conductivity would allow the material to conduct heat farther away from the experiment to pressurized water flowing through small cooling channels.[4]

A composite material consisting of hafnium aluminide (Al_3Hf) particles in an aluminum matrix could both absorb thermal neutrons and conduct heat away from the test position. Neutronic calculations performed to determine the fast-to-thermal ratios and fast flux attainable with the thermal neutron absorber indicate that an Al_3Hf intermetallic-Al composite with ~7 at% Hf is optimum to meet design requirements.[4] Therefore, this study will be limited to an Al_3Hf intermetallic-Al composite of ~7 at% Hf. Assuming this atomic fraction is represented in the form of the Al_3Hf intermetallic particles dispersed in an aluminum matrix, the required volume percentage of the Al_3Hf intermetallic is about 23%.

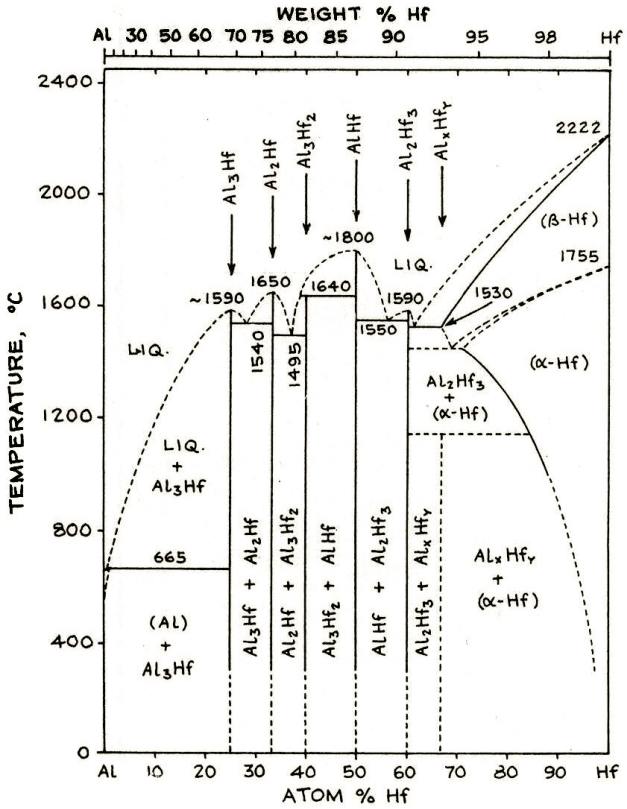

Fig. 1. Aluminum-Hafnium phase diagram. [5]

I.A. Hf-Al Mixtures

The binary phase diagram (see Figure 1) indicates that there is very little or no solubility of hafnium in solid aluminum; intermetallics form across nearly the entire binary diagram. Since the composite will contain excess aluminum, the intermetallic Al_3Hf which contains the least hafnium was chosen as likely to be the most stable. The phase diagram indicates that a 5-10 at% Hf mixture should consist of Al_3Hf in aluminum, the Al_3Hf being stable.

The phase diagram illustrates that a simple mixture of hafnium and aluminum would not be stable, and many intermetallics could form. Using an Al matrix, the diagram suggests that adding Al_3Hf, the intermetallic with the highest Al concentration, would form the most stable mixture. The phase diagram also indicates that a mixture of Al_3Hf and aluminum is stable up to the melting temperature of aluminum (660°C), so the operating temperature should be held below 660°C. The Al_3Hf has a density of 6.32 g/cm^3. [6]

The present objective is to determine a fabrication technique and thermophysical properties of the Al_3Hf intermetallic. The desired thermophysical properties, which were previously unknown, include the following: specific heat, thermal diffusivity, thermal expansion, and thermal conductivity. These thermophysical properties were obtained over a temperature range of 90°C to 375°C. A centrifugal casting technique was used for producing the intermetallic. The thermophysical properties were measured in order to confirm that the thermal conductivity of the material is adequate for use in the thermal (low-energy) neutron filter design. This information will provide new knowledge about the Al_3Hf intermetallic.

II. EXPERIMENTAL

II.A. Fabrication

Al_3Hf samples were fabricated using a centrifugal casting method. Based on preliminary studies, 69 wt.% hafnium bar stock and 31 wt.% of laser-welded aluminum granules were placed together in the crucible for casting. The samples were then cast in an RDO Induction L.L.C. centrifugal caster.

Three samples were produced, which hereafter will be referred to as #53, #54, and #55. All samples were cast under the following conditions: pressure of 20.7 to 48.3 kPa (3 to 7 psi), 3 Argon manual wash cycles, 10 on/off induction current cycles for mixing, speed of 500 rpm, pyrometer casting temperature of ~1450°C, and water quenching directly after casting. However, while sample #55 was cast at ~1450°C it was exposed to more heating during the process with a peak pyrometer temperature of ~1530°C. The possible results of this extra heating will be discussed later in the paper.

Pieces of each of the three samples were crushed using a mortar and pestle. Powder X-ray diffraction (XRD) was performed to confirm the phase produced by the fabrication process. The patterns of the three samples in the binary system Al-Hf were collected at the Idaho National Laboratory (INL) on a Sintag (theta-theta) diffractometer with $k\alpha 1/2$ emission. Instrument conditions

were as follows: slits – 3, 4, 0.5, and 0.2; step size – 0.02, rate – 1° per minute; range – 18 to 85° 2theta. Rietveld structure analysis (Bruker TOPAS3) was then applied to the data using a general parameter approach to determine the percentage of each phase present.

A composite of the Al_3Hf intermetallic in an aluminum matrix was formed using cast Al_3Hf samples. A mortar and pestle were used to grind solid chunks of the Al_3Hf sample into powder. The material was found to be very brittle and friable. The resulting powder was ground and sieved through a 212µm mesh. A sonic sifter was then used to sieve the Al_3Hf powder into size ranges (e.g. 75µm - 105µm and 149µm - 212µm). The tower held sieve sizes of 38µm, 53µm, 75µm, 105µm, 149µm, and 212µm. All sonic sieving was completed in an Argon atmosphere glovebox.

Consideration was given to the optimum particle size of the Al_3Hf intermetallic dispersed within the aluminum matrix. Using the Born approximation to find the maximum macroscopic transport cross section, the optimum particle size for a nanodispersed moderator can be found.[7] Based on that approximation there appears to be no appreciable difference in the macroscopic transport cross section for differing particle sizes above 1 µm. Therefore particle size for this study is not limited by this consideration.

Since the amount of prepared Al_3Hf was limited, a surrogate powder was chosen to explore concerns of uniform mixing and packing density of the composite. Vanadium was chosen as the surrogate powder for two main reasons. First, because the darker grey color made it easy to distinguish from aluminum when checking for even distribution of particles; second, because the respective densities of vanadium and Al_3Hf (e.g., 6.1 g/cm^3 and 6.28 g/cm^3) are similar. This density similarity is important because it was the large difference between Al_3Hf's and Al's densities that led to the concern over even particle distribution after mixing. One of the differences between the surrogate and Al_3Hf is vanadium's larger grain size. However, it is believed that Al_3Hf's smaller grain size would improve packing density so the difference was not concerning.

The results of the mock-up composites were examined optically and appear to have fairly even particle distribution. The density of five, pressed, half-inch diameter samples were also measured and their mean density was calculated to be 1.01 times the theoretical density. These two factors indicated that the mixing and pressing procedures should be adequate to use for the Al_3Hf powders. Powder from each size range was used to create laser flash sized compacts.

II.B. Characterization

Thermophysical properties of the Al_3Hf intermetallic have not previously been measured or obtained. Specific heat (constant pressure), thermal diffusivity, and thermal expansion were investigated for design of the neutron filter. Thermal conductivity is an important property for the design of the filter that can be determined from the product of density, specific heat (constant pressure), and thermal diffusivity measurements

$$k=\rho c_p \alpha \quad (1)$$

where

α = thermal diffusivity
c_p = specific heat
k = thermal conductivity
ρ = density

Constant values of density were used in order to determine the thermal conductivity. Using the available data from the Rietveld analysis and known density values for various intermetallics in the Al-Hf binary system, the density values were calculated.[8] The resulting densities of samples #53, #54, and #55 were 6.32 g/cm^3, 6.32 g/cm^3, and 6.29 g/cm^3 respectively.

The specific heat of the samples was measured with a Netzsch DSC404 differential scanning calorimeter (DSC). Two sets of heating and cooling measurements were made within a temperature band of 40°C-435°C using dynamic heating and cooling rates of 10ºC/min. The thermal diffusivity measurements were collected using a Netzsch LFA 427 laser flash thermal diffusivity measuring system. Data was collected on the samples from 40°C-475°C at approximately 100ºC intervals upon heating and cooling. The samples were allowed to reach thermal equilibrium prior to the collection of data at each temperature step. Thermal expansion data was measured using a Netzsch DIL402E pushrod dilatometer system. Data was collected upon heating and cooling at 5ºC/min from 20ºC-400ºC. Thermal conductivity of the Al_3Hf samples was calculated using Equation 1 with these measured thermophysical properties.

III. RESULTS AND DISCUSSION

III.A. Structural Data

The Rietveld analysis, shown in Figure 2 and Table I, indicates that the fabrication technique produces an acceptable amount of Al_3Hf in the material. Samples #53 and #54 were comprised mainly of the low-temperature phase of Al_3Hf. Sample #55 contained a much higher percentage of the high-temperature phase which is believed

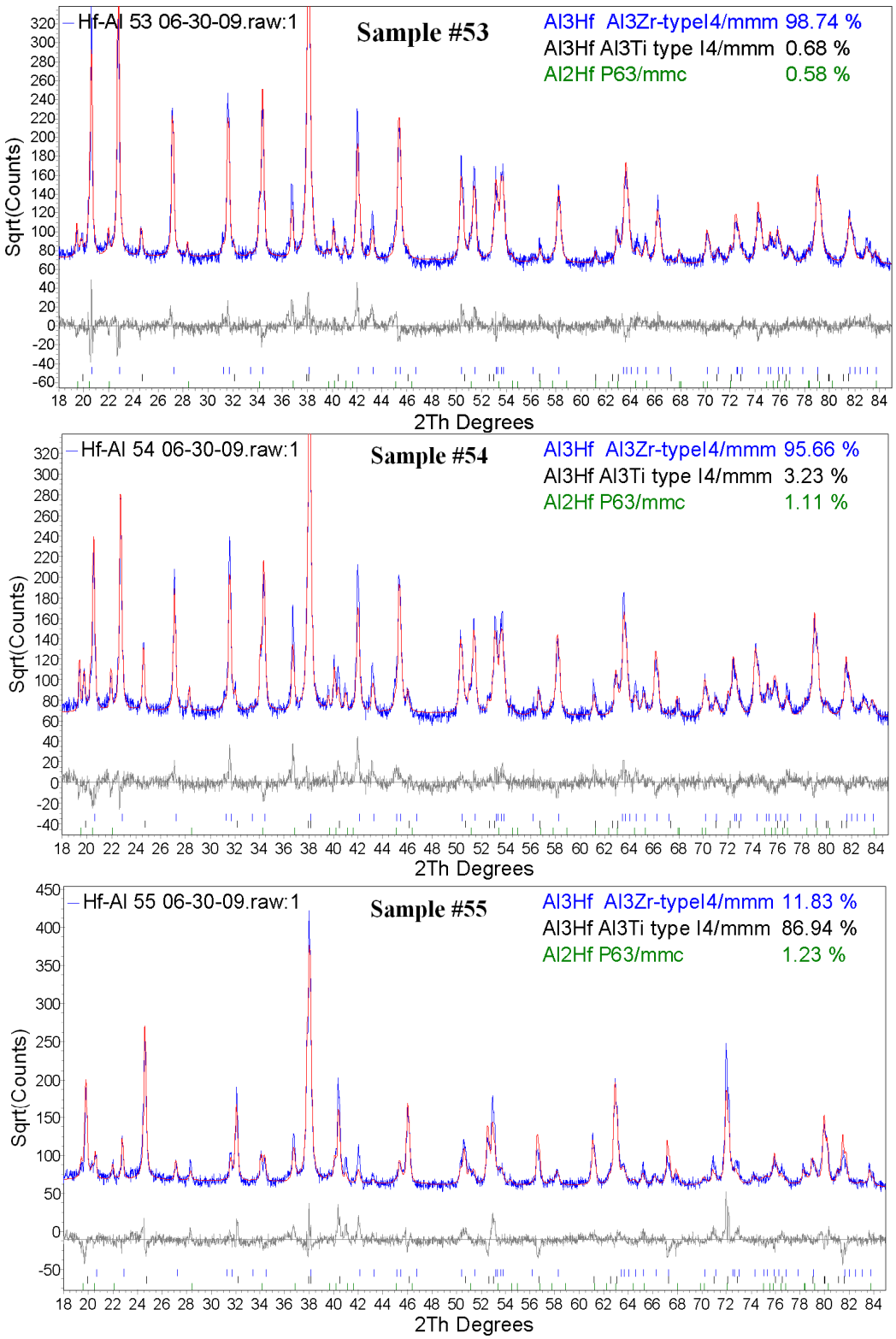

Fig. 2. Rietveld Profile Refinement for Samples #53, #54, and #55. Blue pattern: measured intensities, red pattern: calculated intensities ($F_{hkl}2$), grey pattern: intensity differences.

to be a result of being heated to a higher peak temperature before casting, as was described in the Fabrication section.

Table I outlines the composition of each sample based on the weight percent of the manifesting intermetallics. Table II provides information on the unit cell configuration for each of the intermetallics contained in the samples. Note also that there were no oxides or excess aluminum detected in any significant amounts.

TABLE I

Phase Analysis of Samples #53, #54, and #55

	Quantitative Phase Analysis [wt.%]		
Sample	Al_3Hf (Al_3Zr – type) I4/mmm	Al_3Hf (Al_3Ti - type) I4/mmm	Al_2Hf P63/mmc
#53	98.7 ± 3.4	0.7 ± 3.4	0.58 ± 0.31
#54	95.7 ± 3.0	3.2 ± 3.1	1.11 ± 38
#55	11.8 ± 1.3	86.9 ± 1.4	1.23 ± 0.42

TABLE II

Refined Lattice Parameter (no internal standard added)

	Refined Lattice Parameter [Å]		
Sample	Al_3Hf (Al_3Zr – type) I4/mmm	Al_3Hf (Al_3Ti - type) I4/mmm	Al_2Hf $P6_3$/mmc
#53	a = 3.98883(97) c = 17.1568(43)	a = 3.9348(18) c = 8.9022(70)	a = 5.2397(15) c = 8.6741(26)
#54	a = 3.9876(10) c = 17.1566(44)	a = 3.9312(12) c = 8.8965(31)	a = 5.2372(15) c = 8.6733(25)
#55	a = 3.9868(13) c = 17.1560(68)	a = 3.9319(10) c = 8.9019(23)	a = 5.2388(17) c = 8.6696(32)

The low-temperature phase prevails in both samples #53 and #54. The low-temperature phase is associated with an Al_3Zr type. The high-temperature phase that is associated with sample #55 relates to an Al_3Ti type. It is suspected that the thermophysical properties of samples #53 and #54 should be close to those of Al_3Zr. Al_3Zr has an average thermal conductivity of 42.0 W/m-K.

III.B. Thermophysical Properties

The specific heat of the samples was measured as a function of temperature as the material was heated and cooled (Figure 3). The precision uncertainty maximum to achieve 95% confidence for samples #53, #54, and #55 are 10.3%, 9.5%, and 5.0%, respectively. This was due mainly to the small number of samples taken. Future work includes performing measurements on additional samples.

There is no significant difference between the specific heat of the three samples. The high-temperature phase present in sample #55 does not appear to affect the specific heat of the material. However, sample #55 did display a more pronounced difference between the heating and cooling curves than the predominately low-temperature phase samples. This difference is possibly a result of relaxation phenomena in the measurement, or perhaps the high-temperature phase is not stable and reverted to the low-temperature phase upon cooling – the cooling curve is more similar to that of #53 and #54 than was the heating curve. At least some of the hysteresis between the heating and cooling curves found in all of the data is likely due to grain coarsening. Further research is necessary to determine if this is a function of the measurement or of the material.

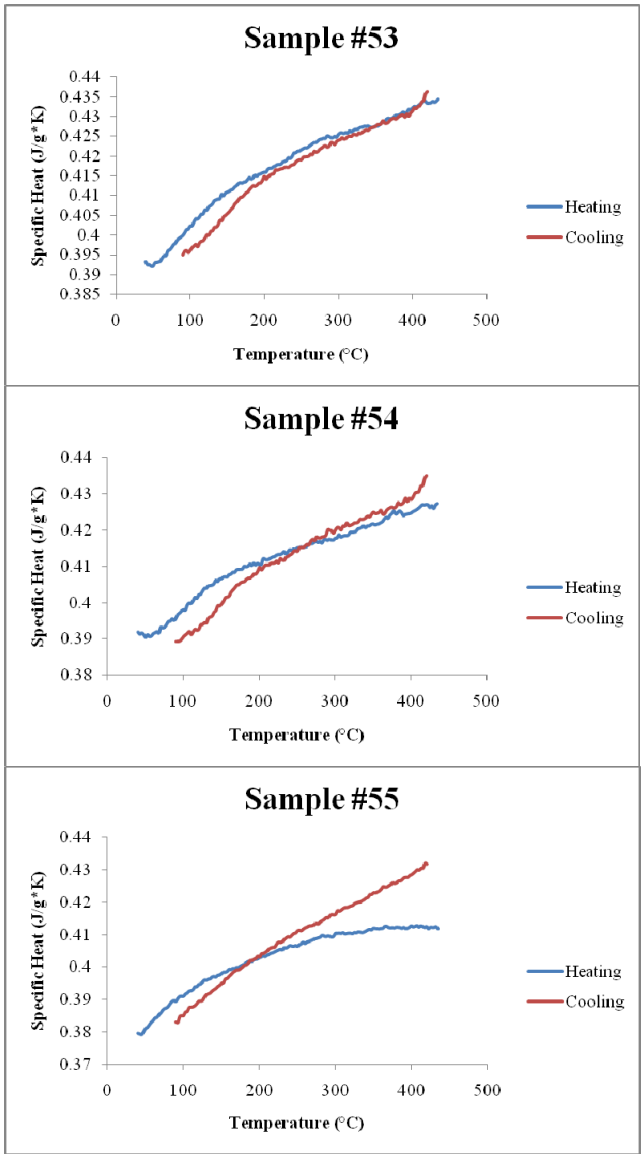

Fig. 3. Heat capacity varied with temperature for samples #53, #54, and #55 (heating and cooling).

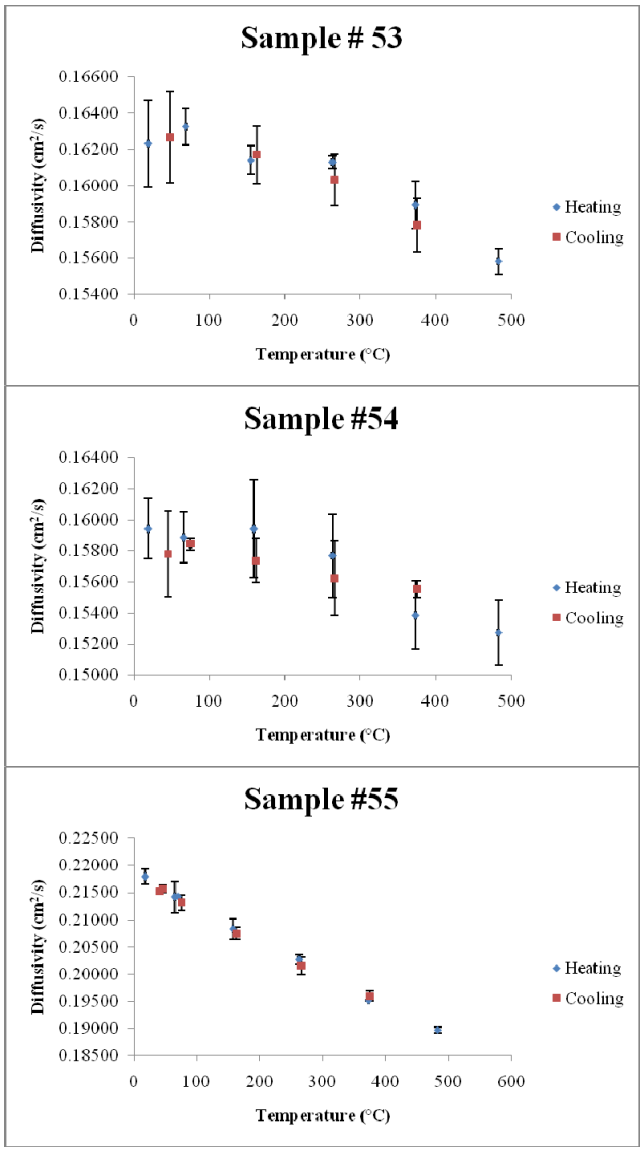

Fig. 4. Thermal diffusivity measurements versus temperature for samples #53, #54, and #55 (heating and cooling).

The thermal diffusivity measurements upon heating and cooling for each sample are shown in Figure 4. Error bars indicate the precision uncertainty for a 95% confidence interval.

Samples #53 and #54 have similar thermal diffusivity values as expected due to their similar phase compositions. The thermal diffusivity values for sample #55 are ~25% greater than those of the other samples for a given temperature. This difference could be due to the larger manifestation of the high-temperature phase of Al_3Hf contained in the sample. The effect of this will be propagated into the thermal conductivity calculation.

Thermal expansion data upon heating and cooling for samples #53 and #55 are given in Figure 5. The samples both show linear expansion with temperature indicating no obvious phase transitions. However, if some of the high-T phase in #55 had transitioned to the low-T phase during the cycle it may not be indicated by thermal expansion as the crystal structures are so similar.

Figure 6 shows the variation of the calculated thermal conductivity as a function of temperature for the three samples. Results for the two low-temperature phase samples are noticeably lower than that of the high-temperature phase sample. The thermal conductivity does not vary appreciably with temperature. Uncertainties from specific heat and thermal diffusivity measurements were propagated to determine the uncertainty of the thermal conductivity values. Each curve has a maximum uncertainty of 5%.

The thermal conductivity values for sample #55 tend to be ~25% greater than those of samples #53 and #54 as a result of its higher thermal diffusivity values. A larger portion of the high-temperature phase of the Al_3Hf intermetallic seems to increase the thermal conductivity of the material.

The thermal conductivity values obtained for samples #53 and #54 are close to those of Al_3Zr[9], confirming the hypothesis that a similar crystal structure would yield similar thermophysical properties.

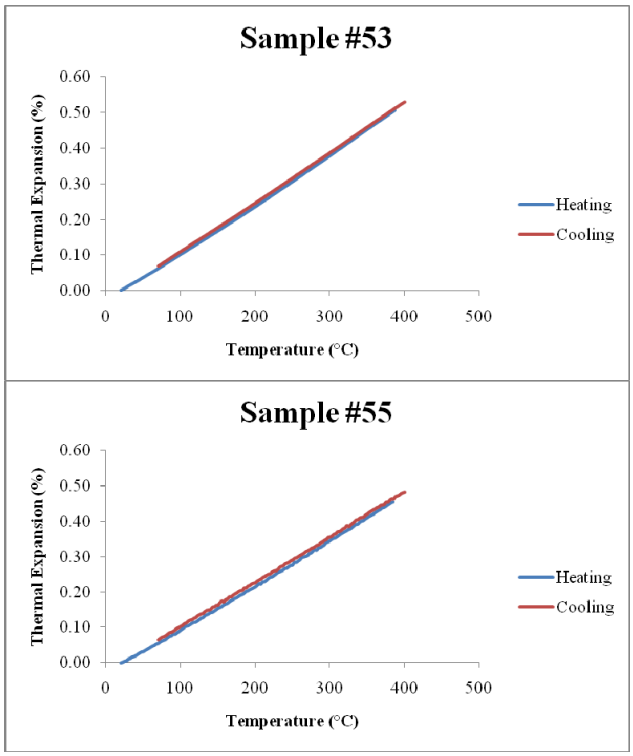

Fig. 5 Thermal expansion data verses temperature for samples #53 and #55 (heating and cooling).

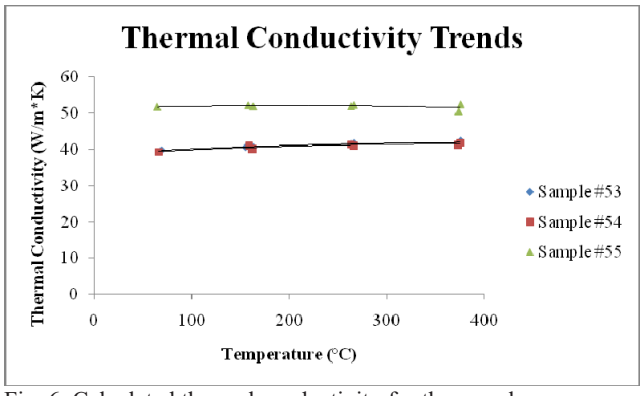

Fig. 6. Calculated thermal conductivity for the samples.

III.C. Design for Al_3Hf-Al Composite

To determine the force required to form the composite, aluminum compacts were pressed to determine how much pressure was required to achieve 99% of the theoretical density. Figure 7 shows the variation of aluminum compact density with die pressure. The associated micrographs showing the particle packing that supports this plot. Figure 8 shows optical microscopy images of the surface of the as-pressed samples.

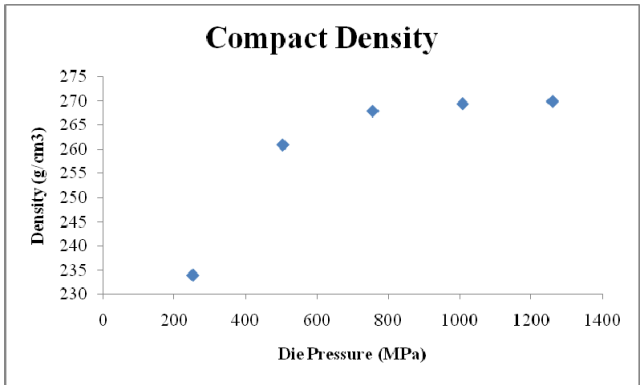

Fig. 7. Variation of aluminum compact density with die pressure. (data courtesy of Pavel Medvedev, Idaho National Laboratory)

Based on the die pressure data, a pressure of ~702 MPa was used to press the powder into pellets. This pressure allowed for 99% compact density to be achieved. This fabrication process used to produce the Al_3Hf-Al composite must still be validated by microstructure analysis.

A model to determine the conductivity for the Al_3Hf-Al composite was initially developed based on previous studies.[10] Results of this model are shown in Figure 9 and indicate the expected value for thermal conductivity at various volume fractions of the Al_3Hf intermetallic. Only samples #53 and #55 were used since sample #54 agrees closely with #53. The average thermal conductivity of the intermetallic for samples #53 and #55 is 41.2 W/m-K and 51.9 W/m-K, respectively.

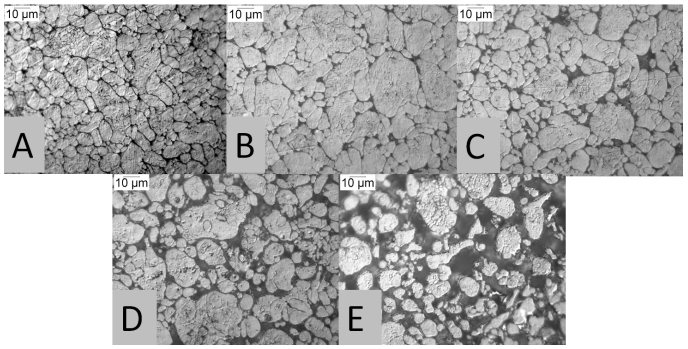

Fig. 8. Effect of die pressure on the microstructure of the Al powder compacts. Die pressures: A) 1260 MPa, B) 1008 MPa, C) 756 MPa, D) 504 MPa, E) 252 MPa. (photo courtesy of Pavel Medvedev, Idaho National Laboratory)

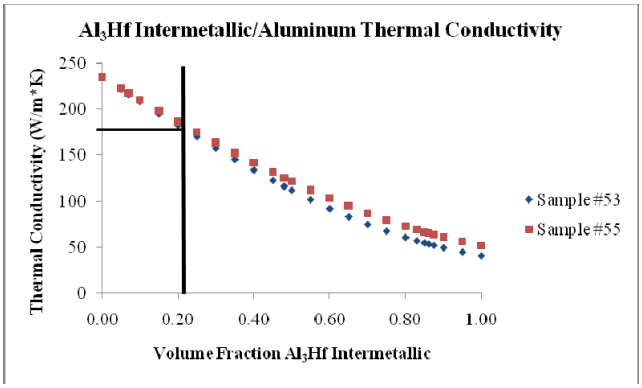

Fig. 9. Model representing Al_3Hf-Al composite thermal conductivity.

The model was produced assuming no matrix porosity and an aluminum thermal conductivity value of 235 W/m-K. The vertical line in Figure 9 indicates the design condition (23 vol% intermetallic) for the conduction cooled thermal (low-energy) neutron filter.

In the model, there is not a significant difference between the composite containing sample #53 and the composite containing sample #55. The thermal conductivity for samples #53 and #55 placed in an aluminum matrix are 177 W/m-K and 182 W/m-K, respectively. There is less than 3% difference between the thermal conductivities of the two samples based upon the model prediction. Therefore, the casting temperature can be chosen by convenience. These values also indicate that the composite material will be able to adequately conduct heat away from the experiment.

Designing the composite using an intermetallic of Al_3Hf with mostly high temperature phase has limitations. Under certain temperature conditions a phase transformation can occur reducing a majority of the high temperature phase intermetallic to the low temperature

phase. This was discussed in Section III.B. in relation to the specific heat measurement of sample #55. As a result, the fabrication technique will be to produce an intermetallic similar to sample #53 so that the composition consists of more low-temperature phase Al_3Hf.

IV. CONCLUSIONS

The development of a conduction cooled thermal (low-energy) neutron filter relies on the understanding of the fabrication technique and thermophysical properties of the Al_3Hf intermetallic. Knowledge of thermophysical properties is necessary to perform thermal analyses to determine the ability of the proposed neutron filter to conduct or dissipate a large quantity of locally generated heat. Conclusions of this study:

1. Centrifugal casting of hafnium (69 wt.%) and aluminum (31 wt.%) at pyrometer temperature ~1450°C yields a material containing > 95% low-phase Al_3Hf intermetallic. Casting temperature does not have a significant effect on thermal conductivity of the composite.
2. Specific heat of the intermetallic ranges from 0.39 J/g-K to 0.43 J/g-K over a temperature range of 90°C - 420°C.
3. Thermal diffusivity of the intermetallic ranges from 0.162 cm^2/s to 0.154 cm^2/s over a temperature range of 0°C - 500°C.
4. Thermal expansion measured, and as expected shows no phase transformations.
5. Thermal conductivity of the intermetallic ranges from 39 W/m-K to 42 W/m-K over a temperature range of 70°C - 375°C. A larger portion of the high-temperature phase of the Al_3Hf intermetallic appears to increase the thermal conductivity.
6. A model based on the average value for the thermal conductivity of the intermetallic predicts the Al_3Hf-Al composite has an average thermal conductivity value of 177 W/m-K.

Further research remains to confirm that the composite will have thermophysical properties that agree with the model presented. Future work includes:

1. Fabrication and characterization of an Al_3Hf-Al composite.
2. Post-irradiation characterization of the composite material.
3. Determination of high-temperature phase relaxation phenomena.

ACKNOWLEDGMENTS

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NOMENCLATURE

α	thermal diffusivity
c_p	specific heat
k	thermal conductivity
ρ	density

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