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ABSTRACT

This report provides an overview of thermophysical properties of unirradiated uranium alloyed with ten weight percent molybdenum (U-10Mo), with particular focus on those material properties needed for modeling of new fuels for HPRRs (High Performance Research Reactors). The report contains both historical data available in the literature on U-10Mo, as well as more recent results conducted by the Global Threat Reduction Initiative fuel development program. The main use of the report is intended as a standard U-10Mo alloy properties reference for reactor models and simulations.

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ACRONYMS

AM	arc-melting
Cp	specific heat capacity
CTE	coefficient of thermal expansion
DU	depleted uranium
GTRI	Global Threat Reduction Initiative
HPRR	High Performance Research Reactor
LEU	low enriched uranium
Mo	molybdenum
ppm	parts per million, also expressed as $\mu g \bullet g^{-1}$
U	uranium
U-10Mo	uranium alloyed with ten weight percent (nominally) molybdenum
VIM	vacuum induction melting
wt%	weight percent

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1. INTRODUCTION

Low enriched uranium (LEU)-molybdenum fuel alloys are a key element of the Global Threat Reduction Initiative (GTRI) fuel development program for three main reasons: (1) the high uranium density in LEU-molybdenum alloy fuel makes it a potential replacement fuel for research reactors currently operating with highly enriched uranium, (2) the dimensional stability characteristic of the body-centered cubic gamma phase can persist during irradiation as body-centered cubic metal, permitting higher central metal temperatures for a given amount of swelling, and (3) the metallurgical properties that ensure integrity and corrosion resistance to high-temperature water are far greater than that of unalloyed uranium.

A compilation of relevant thermophysical properties of the LEU alloyed with 10 wt% (nominally) molybdenum, hereinafter referred to as U-10Mo, is presented in this report. The report is intended to be used as a common source of data for work related to the High Performance Research Reactor (HPRR) conversions. Material properties summarized in this report were primarily selected based on the analysis needs of reactor operators, modelers, researchers, fabricators, and regulators. Additional properties and supporting analysis can be found in the references cited.

Material properties included in this report are meant to act as a catalyst to bring standardization to models and simulations employing the U-10Mo alloy. Data summarized in this report represents the best information available obtained from both historical and current programmatic studies. In some cases, there is a large spread in the data, particularly for properties related to fabrication and structural integrity of the fuel alloy. New and additional research to further validate, modernize, and complement the best available data is underway. This report will be updated as the new and additional research is completed.

Additional documents are being prepared to fully characterize monolithic U-10Mo, as well as cladding and structural materials. It is critical to begin new research and continue efforts already underway to validate or modernize the existing research available, since the available references on U-10Mo cited in this presentation are approaching a half century since publication. This report provides data that will be revised to include new data as measurements and characterizations are completed.

2. THERMOPHYSICAL PROPERTIES

2.1 Specific Heat Capacity

Specific heat capacity is an intensive quantity that illustrates the amount of heat needed to change the body of fuels temperature. A high specific heat capacity helps stabilize fuel elements thereby reducing the probability of fuel failure. Furthermore, specific heat capacity is also important in the estimation of stored energy in the fuel during potential accident scenarios. Table 1 summarizes available specific heat capacity data for the U-10Mo alloy.

Table 1. Specific heat capacity data for U-10Mo.

	Cp [J g^{-1}°C^{-1}]			
T (°C)	Farkas[1]	Fackelmann[2]	Burkes[3]	Average
100	0.142	0.141	0.143	0.142 ± 0.001
200	0.149	0.148	0.144	0.147 ± 0.003
300	0.157	0.156	0.148	0.154 ± 0.005
400	0.164	0.164	0.155	0.161 ± 0.006
500	0.172	0.171	0.165	0.169 ± 0.004
600	0.179	0.178	0.167	0.175 ± 0.007
700	0.187	0.186	0.170	0.181 ± 0.009
800	0.194	0.193	0.179	0.189 ± 0.008
900	0.201	0.200		0.201 ± 0.001
1000	0.209	0.208		0.208 ± 0.001

Specific heat capacity of the U-10Mo alloy increases near linearly with respect to temperature. There is minimal difference in the specific heat capacity of the alloy up to 600 °C for the three references provided here. Variations in the data can be attributed to differences in the experimental method and procedure used to perform the measurement, alloy and sample preparation techniques, and chemical variations. References can be directly consulted to investigate the impacts of such variations.

The least squares method was applied to the data, approximating a second order polynomial that best fit the data, represented by Equation 1.

$$Cp_{U-10Mo} = \left(0.137 \pm 3.31\times10^{-3}\right) + \left(5.12\times10^{-5} \pm 1.41\times10^{-5}\right)\cdot T + \left(1.99\times10^{-8} \pm 1.29\times10^{-8}\right)\cdot T^2 \tag{1}$$

In the above equation, $Cp_{U\text{-}10Mo}$ is the specific heat capacity in J g^{-1}°C^{-1} and T is the temperature from 100 to 1000 °C. The average specific heat capacity values and the best-fit polynomial are shown graphically in Figure 1. Also included in the figure is an approximated value for a depleted uranium (DU) – 10Mo alloy employing the Neumann-Kopp rule. Values for the Neumann-Kopp rule will not be entirely accurate since α-U is used in the calculation from room temperature to 669 °C, β-U from 669–776 °C, and finally γ-U above 776 °C. As observed in the figure, the Neumann-Kopp rule is generally not an adequate representation of the specific heat capacity for the γ-phase U-10Mo alloy.

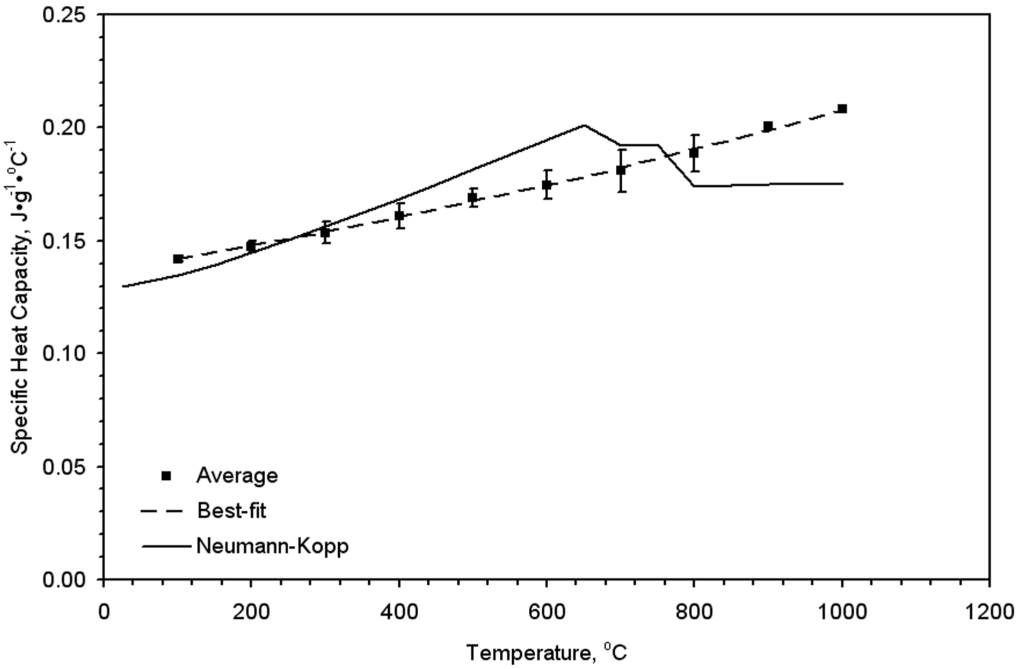

Figure 1. Specific heat capacity of the U-10Mo alloy and specific heat capacity of a DU-10Mo alloy predicuted using the Neumann-Kopp approximation.

2.2 Coefficient of Linear Thermal Expansion

Coefficient of thermal expansion (CTE) data is critical to modeling fuel for nuclear reactors. Thermal expansion increases the volume and decreases the density of the fuel system, which in turn increases the neutron leakage and allows for negative feedback of the system. Table 2 summarizes available coefficient of linear thermal expansion data for the U-10Mo alloy.

Table 2. Coefficient of linear thermal expansion data for U-10Mo.

	CTE [$\times 10^{-6}$ °C^{-1}]		
T (°C)	Burkes[3]	Saller[4]	Average
RT-100	11.8 ± 0.2	11.5	11.7 ± 0.2
RT-200	12.6 ± 0.2	11.8	12.4 ± 0.4
RT-300	14.1 ± 0.9	12.4	13.7 ± 1.1
RT-400	16.1 ± 1.9	12.7	15.2 ± 2.3
RT-500	16.4 ± 1.7	13.0	15.6 ± 2.2
RT-600	16.6 ± 1.3	13.5	15.8 ± 1.9
RT-700	16.7 ± 0.9		16.7 ± 0.9
RT-800	17.2 ± 1.1		17.2 ± 1.1

The data presented in Table 2 represent the instantaneous coefficient of linear thermal expansion. This value is obtained by evaluating the slope of displacement with respect to temperature over a given temperature range. Values can be affected by changes in the slope of the displacement-temperature curve. These types of changes can be the result of porosity within the sample, homogenization or decomposition of the alloy during testing, alleviation of residual stresses, etc. The least squares method was used to calculate a best-fit line provided in Equation 2, where T is the temperature from RT to 800 °C and CTE_{U-10Mo} is the coefficient of linear thermal expansion $\times 10^{-6}$ °C^{-1}.

$$CTE_{U-10Mo} = (11.2 \pm 0.6) + (8.07 \times 10^{-3} \pm 1.14 \times 10^{-3}) \cdot T \quad (2)$$

The average values for the instantaneous coefficient of thermal expansion of U-10Mo show minor divergence from the best fit values, as shown in Figure 3.

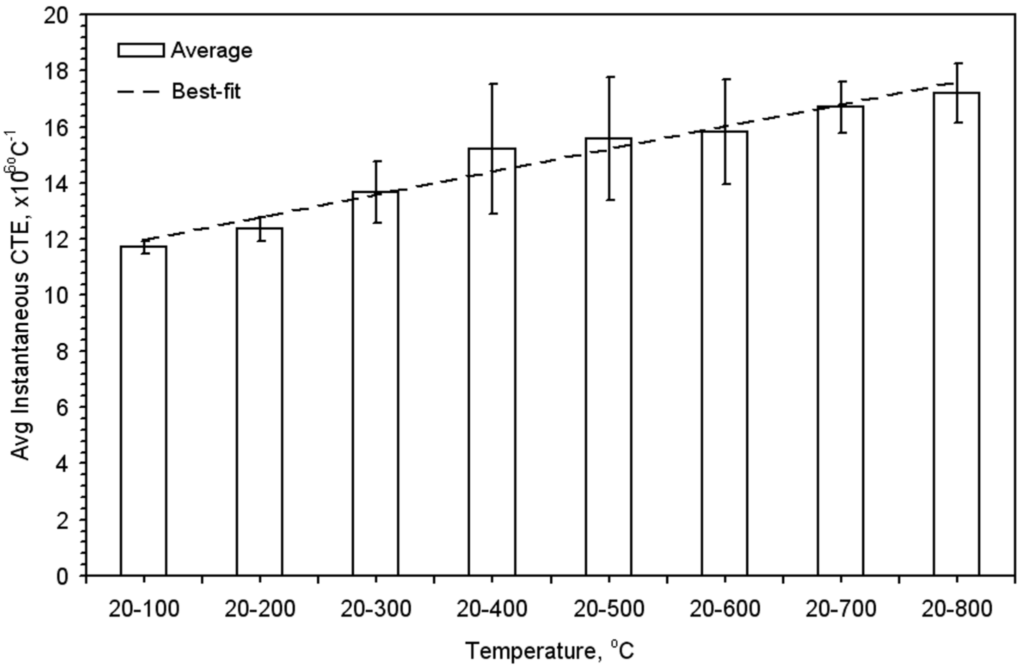

Figure 2. Instantaneous coefficient of linear thermal expansion for the U-10Mo alloy as a function of temperature range.

2.3 Density

The value for density of the U-10Mo alloy is calculated by the rule of mixtures using atomic mass and density of the alloy constituents. This method finds the theoretical density of U-10Mo alloy using the rule of mixtures shown in Equation 3.

$$\rho_{U-Mo} = X_{Mo}\rho_{Mo} + (1 - X_{Mo})\rho_U \quad (3)$$

In Equation 3, X_{Mo} and X_U are the mole fractions of Mo and U in the U-Mo alloy, and $\rho_{U\text{-}Mo}$, ρ_{Mo}, and ρ_U are the densities of the U-Mo alloy, elemental Mo, and elemental U, respectively with units of g•cm^{-3}. The density of a natural U-10Mo alloy is calculated to be 17.2 g•cm^{-3}. The representative U-10Mo fuel foils will have a significantly higher U-235 content than natural uranium. The recommended data for the density of U-10Mo has a uranium composition enriched to 19.75 wt% uranium-235 and assumes the uranium-234 content is unchanged from natural uranium. The remaining 80.25 wt% consists of uranium-238. Using values from Reference 5 and Equation 3, the molar mass of the LEU-10Mo fuel foil would be 237.456 g•mol^{-1}. Assuming that the total atom density does not change, the density of LEU will be less than natural uranium due to the lower atomic weight, resulting in an LEU density of 19.054 g•cm^{-3}. Using Equation 3 again, the LEU-10Mo fuel foil at 20 °C will have a theoretical density of 17.145 g•cm^{-3}. A change of approximately 0.05 g•cm^{-3} occurs when the density of natural uranium is changed to roughly 20 percent enriched in uranium-235.

As the temperature of the alloy is increased, the density will correspondingly decrease due to thermal expansion. Table 3 summarizes density data for the U-10Mo alloy as a function of temperature. It is important to note the known differences of the data in Table 7. Data from Reference 3 was obtained from a DU-10wt% Mo alloy (therefore a theoretical density of 17.2 g•cm^{-3} rather than 17.15 g•cm^{-3}) and samples were documented to contain roughly 4% porosity. The values were not corrected for the presence of this porosity (although values are roughly 4% lower than those in the other References). References 6 and Reference 7 do not mention the enrichment of the uranium used in their alloys. Furthermore, there is no mention made whether the samples contained porosity, and whether the data was corrected for that porosity. Since the sensitivity of density for changes in uranium enrichment is a possible source of error, average values are not presented for density as a function of temperature.

Table 3. Mass density data for U-10Mo alloys as a function of temperature.

	$\rho_{U\text{-}10Mo}$ [g•cm^{-3}]		
T (°C)	Burkes[3]	Klein[6]	Bridge[7]
20		17.13	17.12
100	16.38	17.06	17.06
200	16.31	16.97	16.97
300	16.23	16.88	16.88
400	16.14	16.80	16.79
500	16.06	16.71	16.71
550	16.02	16.66	
600	15.98		16.62
700	15.90		16.53
Slope	-8.315×10^{-4}	-8.846×10^{-4}	-8.719×10^{-4}

Alternatively, the slope of the density change as a function of temperature can be determined from the data available on U-10Mo alloys, since this effect should be independent of uranium enrichment and only slightly dependent upon small amounts of porosity in the test samples. The average slope of the data presented in Table 3 is -8.63 × 10-4 ± 2.77 × 10^{-5} g•cm^{-3} (within ±3%). An expression for the density of a fully dense LEU-10Mo fuel foil can therefore be created using the average slope from data in Table 3 and the room temperature theoretical density discussed above. This expression is provided in Equation 4 and shown graphically in Figure 3.

$$\rho_{U-10Mo} = 17.15 - \left(8.63\times10^{-4} \pm 2.77\times10^{-5}\right)\cdot\left(T+20\right) \tag{4}$$

In Equation 4, $\rho_{U\text{-}10Mo}$ is density of LEU-10Mo in g•cm^{-3} and T is temperature from 20 to 700 °C.

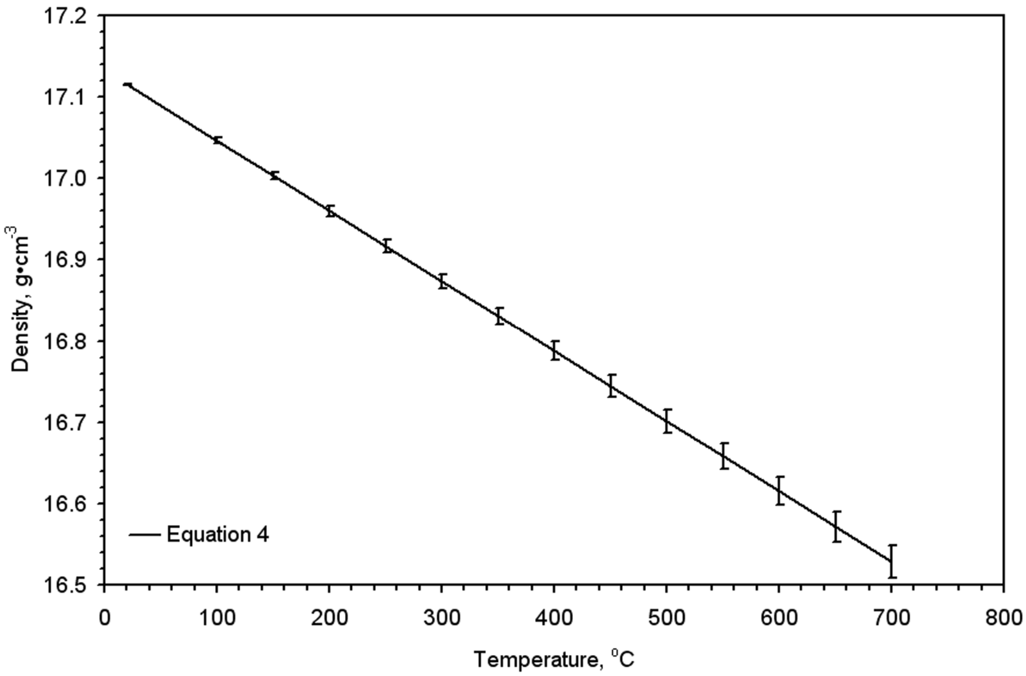

Figure 3. Density of LEU-10Mo as a function of temperature.

2.4 Thermal Conductivity

Thermal conductivity is one of the most important properties of a nuclear fuel. This property ultimately plays a significant role in determining the maximum operating power of a fuel element and the available safety margins. Economics of utilizing the fuel is therefore directly impacted by thermal conductivity. Benefits of increased thermal conductivity include decreased thermal strain, increased operating lifetime, and improved fuel integrity. Table 4 summarizes available thermal conductivity data for the U-10Mo alloy as a function of temperature.

Table 4. Thermal conductivity data for U-10Mo as a function of temperature.

	λ [W m^{-1}°C^{-1}]				
T (°C)	Burkes[3]	Klein[6]	McGeary[8]	Touloukian[9]	Average
20		12.1	9.7	12.1	11.3 ± 1.4
100		14.2	11.7	13.8	13.2 ± 1.3
200	20.0	17.2	14.0	17.3	17.1 ± 2.4
300	23.9	20.1	17.2	20.1	20.3 ± 2.7
400	27.1	23.0	21.6	23.3	23.7 ± 2.3
500	31.2	26.4	25.7	27.2	27.6 ± 2.5
600	35.5	30.1		30.1	31.9 ± 3.1
700	36.9	33.9			35.4 ± 2.1
800	37.4	37.7			37.5 ± 0.2

Thermal conductivity of the U-10Mo alloy increases near linearly with respect to temperature. The data of Reference 3 was determined utilizing the laser flash thermal diffusivity (LFTD) method and was corrected for the approximate porosity of the DU-10Mo alloy, as determined by Archimedes method on the sample. When this data is compared to the thermal conductivity data of the other references, the values of Reference 3 tend to be roughly 20% higher. In most cases, the thermal conductivity of the alloys from previous literature were determined from electrical conductivity measurements and converted using the Wiedemann-Franz law. Thermal conductivity measurements obtained from electrical conductivity should be lower than those measured in a direct or semi-direct manner, since electrical conductivity only considers the electronic contribution to thermal conductivity and phonon-phonon scattering is not taken into account. It must also be noted that in the case of older literature, the experimental method and resultant phase analysis of the alloys investigated is not always available or clear. References can be directly consulted to further investigate any impacts of such variations.

The least squares method was applied to the data, approximating a best-fit line represented by Equation 5, where $\lambda_{U\text{-}10Mo}$ is the thermal conductivity in W m^{-1}°C^{-1} and T is the temperature from 20 to 800 °C. The average thermal conductivity values and the best-fit line are shown graphically in Figure 4.

$$\lambda_{U-10Mo} = (10.2 \pm 0.688) + \left(3.51 \times 10^{-2} \pm 1.61 \times 10^{-3}\right) \cdot T \quad (5)$$

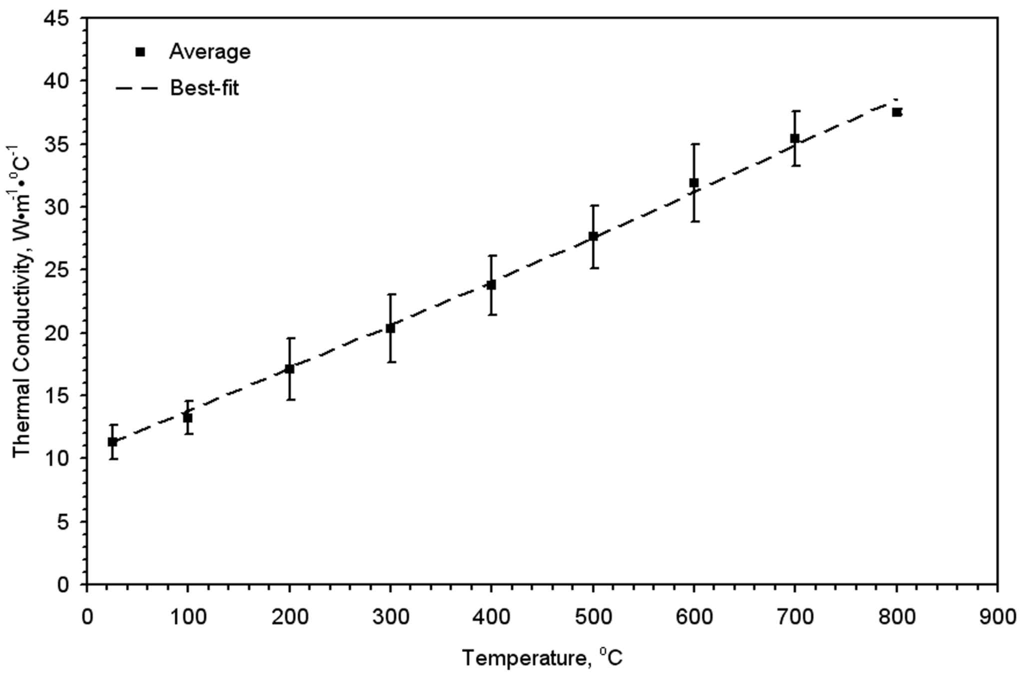

Figure 4. Thermal conductivity of U-10Mo as a function of temperature.

3. CONCLUSIONS

The data and analyses in this report are intended to serve as a catalyst towards standardization of models and simulations using the U-10 wt% Mo fuel alloy under development. Additional documents are being prepared to fully characterize monolithic U-10Mo, as well as cladding and structural materials. It is critical to begin new research and continue current efforts to validate or modernize the existing research available, since the available references on U-10Mo are approaching a half century since publication. The report includes historical data along with data that has been collected by the GTRI fuel development program to date. Thermophysical properties including specific heat capacity, coefficient of thermal expansion, density, and thermal conductivity as a function of temperature are summarized in this report. Temperature dependent equations are suggested for use based on best-fit polynomials placed through the existing data.

4. REFERENCES

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