Thermophysical Properties of U_3Si_2 to 1773 K

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Abstract

Use of U_3Si_2 in nuclear reactors requires accurate thermophysical property data to capture heat transfer within the core. Compilation of the limited previous research efforts focused on the most critical property, thermal conductivity, reveals extensive disagreement. Assessment of this data is challenged by the fact that the critical structural and chemical details of the material used to provide historic data is either absent or confirms the presence of significant impurity phases. This study was initiated to fabricate high purity U_3Si_2 to quantify the coefficient of thermal expansion, heat capacity, thermal diffusivity, and thermal conductivity from room temperature to 1773 K. Datasets provided in this manuscript will facilitate more detailed fuel performance modeling to assess both current and proposed reactor designs that incorporate U_3Si_2 .

Keywords: Uranium silicide, U_3Si_2 , laser flash analysis, thermal conductivity

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1. Introduction

Uranium-rich compounds within the uranium-silicon binary system have seen broad usage as nuclear fuels in low power reactors [1]. Although U_3Si was initially preferred due to its superior uranium density, U_3Si_2 has supplanted it in recent decades primarily due to its enhanced resistance to in-pile swelling induced by rapid amorphization [2–5]. Both U_3Si and U_3Si_2 have seen most extensive use as dispersion fuels, where an aluminum metal matrix incorporates uranium silicide particles. This approach is most prominent for U_3Si_2 , but mitigating the formation of interaction layers between the fuel particles and metallic matrix remains an ongoing challenge [6].

More recently, interest has grown in the exploration of U_3Si_2 for service in more demanding reactor applications such as those of commercial light water reactors (LWR)[7]. This is predominantly driven by its uranium density, anticipated improvement in thermal conductivity relative to that of reference uranium dioxide (UO_2), and limited existing irradiation database. The uranium density of U_3Si_2 is 11.3 g-U/cm³; this is appreciably higher than UO_2 (9.7 g-U/cm³). Such an increase in uranium density could provide reactor performance gains at constant enrichment, or be used to offset a neutronic penalty incurred by replacement of low neutron cross section zirconium cladding with a more oxidation resistant option.

Improving upon the thermophysical properties of UO_2 is also a primary focus of any alternative LWR fuel. The comparatively poor thermal conductivity of UO_2 drives steep thermal gradients and high centerline temperatures during reactor startup and operation. Improvements in the thermal conductivity of the fuel will limit thermally-induced stresses; this may result in reduced pellet cracking. Lower temperatures throughout the fuel pellet will also reduce diffusion, and thereby retard species transport and in-pile restructuring. All of these evolutions contribute to operational uncertainty.

The limited availability of thermophysical properties for U_3Si_2 suggests that it possesses a thermal conductivity vastly superior to that of UO_2 . However, significant scatter exists in the literature, and limited data is available at temperature relevant to LWR operation. This uncertainty has not impeded use of U_3Si_2 in low power reactor applications, as the cooling requirements and need for an accurate understanding of the temperature profiles within the fuel are greatly relaxed compared to those of commercial power reactors. This study was undertaken to determine the thermal conductivity, heat capacity, and thermal expansion of U_3Si_2 from room temperature to near the

38 melt point to evaluate its potential as an LWR fuel. Accurate results of fuel
39 performance modeling executed with this objective requires thermophysical
40 cal property data across a wide range of temperatures in order to capture
41 steady state operation and design basis accident scenarios. If reactor performance
42 is found acceptable, further evaluation of irradiation behavior under
43 LWR conditions, cladding compatibility, off-normal behavior, economic considerations,
44 and the numerous other development pathways needed to scope
45 commercial viability is justified.

46 2. Experimental

47 Preparation of single phase U_3Si_2 in geometries suitable for thermophys-
48 ical property measurement using differential scanning calorimetry (DSC),
49 dilatometry, and laser flash analysis (LFA) was first necessary for execution
50 of this work. Arc melting was chosen as the most efficient means to syn-
51 thesize gram quantities of U_3Si_2 for subsequent fabrication into the required
52 geometries. The high volatility of silicon requires an excess beyond stoichio-
53 metric quantities to compensate for anticipated losses in the molten state.
54 Significant process refinement was undertaken to systematically explore the
55 minimum excess silicon necessary to ensure that the resulting button was as
56 nearly stoichiometric U_3Si_2 as possible without the presence of either Si-rich
57 or U-rich secondary phases. Additional efforts were taken to prevent $\text{O}_2(\text{g})$
58 contamination during the fabrication steps to minimize formation of oxide
59 phases. Detailed characterization of the samples' microstructure and chemi-
60 cal impurities is also essential to facilitate discussion of the phase content of
61 the material measured.

62 2.1. Sample preparation

63 High purity feedstock of depleted uranium plate was used as the starting
64 material. Inductively coupled mass spectrometry was conducted to verify
65 impurity content of the U feedstock. Impurities in the uranium metal were
66 below the detectable limit for all elements with the exception of 31 weight
67 parts-per-million carbon. The plate was then wire electro-discharge machined
68 into discs weighing approximately 10 grams each. The silicon used was Alfa
69 Aesar (Alfa Aesar, USA) stock with 99.999% purity in the form of small
70 irregular shapes up to 5 mm in size. An excess of 0.1 weight percent (w/o)
71 Si was used to minimize either U-rich or Si-rich phases under the arc melting
72 conditions and processing used in this study.

73 Buttons of U_3Si_2 were prepared by arc-melting the disc shaped uranium
74 feedstock to produce an approximate mass of 10 g each. Fabrication was
75 performed in an arc melter (5TA Reed Tri-Arc, Centorr Vacuum Industries,
76 USA) constant current welder connected to a water cooled 2% thoriated
77 tungsten electrode and a water cooled copper hearth with small hemispherical
78 cavities to adequately contain the uranium feedstock. A gettered argon gas
79 stream fed the arc melter chamber with an oxygen mole fraction (χ_{O_2}) less
80 than 10^{-10} parts per million (ppm). A molten Ti getter was also maintained
81 inside the arc melter itself to capture any remaining oxygen in the chamber

during melting of the charge. U_3Si_2 is reported to form congruently at 2043 K according to the most recent published phase diagram [8]. When cooled from the molten state, the Si-rich compound (as was synthesized in this study based upon pre- and post-arc melt weights) is reported to pass through a eutectic at 1813 K to form U_3Si_2 and USi .

Chemical analysis was conducted on three separate samples as prepared from a comminuted button. Inductively coupled plasma-optical emission spectroscopy (ICP-OES) was used to determine transition and transuranic elements, while the WO_3 combustion catalyst method was utilized to determine H, C, and N content. Elemental O analysis is not possible using this experimental technique, and was therefore not evaluated in this study. ICP-OES required 50 mg of powder to be digested in a solution of nitric, hydrofluoric, and hydrochloric acid. Chemical analysis was performed by ALS Environmental of Tuscon, Arizona (USA). Three measurements were conducted on powder from the representative pellet to explore potential chemical variations within the feedstock. The results of the chemical analysis for detectable elements are shown in Table 1. It is noted that the buttons were processed with 1 w/o excess Si, while the resulting button chemistry is slightly Si-lean. It is expected that this degree of Si loss can be attributed to the volatilization of Si during arc-melting.

2.2. Sintering U_3Si_2 powder

Specimens for thermophysical property measurements were prepared by a powder metallurgy route. Arc-melted U_3Si_2 buttons were comminuted in a high energy SPEX mill (8000M, SPEX Sample Prep, USA) using a stainless steel jar and milling media. The resulting powder was sieved at -200 mesh within a nitrogen glove box maintained below 20 ppm O_2 concentration to minimize oxidation of the powder. Sieved powders were then pressed at 300 MPa in a steel die set, which utilized zinc stearate as a die lubricant. Geometric green densities ranged between 55% and 65% of the theoretical density. Binders were not used during fabrication of the pellets in order to minimize potential oxygen or carbon contamination. Pellets were sintered using a W metal furnace on W foil under flowing Ar gas using a constant heating and cooling rates of $5 \text{ K}\cdot\text{min}^{-1}$ to a maximum temperature of 1773 K for 12 h. Densities were determined using a geometric method. The balance used to determine masses in this study was calibrated to 0.01 mg. Resulting pellets were between 96-98% of the theoretical density of U_3Si_2 taken as $12.2 \text{ g}\cdot\text{cm}^{-3}$ [9], which was confirmed using digital image analysis [10] to be $98\%\pm 2\%$. The

119 Si vapor pressure for U_3Si_2 is $\sim 6 \times 10^{-8}$ atm [11] at this sintering temperature.
120 It is therefore expected that the Si content of the sintered pellets Si remained
121 unchanged.

122 2.3. Microstructure analysis

123 Powder X-ray diffraction (XRD) was performed on a Bruker D2 diffrac-
124 tometer using $\text{CuK}\alpha$ radiation. Measurements were conducted in a Bragg-
125 Brentano geometry from $20 - 90^\circ 2\theta$ with a step size of 0.01° . Powder spec-
126 imens were prepared by comminuting buttons using a mortar and pestle.
127 This was done inside a N_2 atmosphere controlled glovebox to minimize oxy-
128 gen contamination where O_2 contamination was maintained below 30 ppm.
129 The powder was mounted on a Si crystal zero-background plate using a thin
130 layer of vacuum grease, and then sealed inside a polymer dome while in-
131 side the glovebox. The polymer dome had an air scatter shield installed to
132 minimize the contribution of the background to the signal.

133 Figure 1 confirms that U_3Si_2 is the dominant phase present in the sample.
134 For comparison, the characteristic peaks of other U-Si phase in the vicinity
135 of U_3Si_2 are shown. No other U-Si phases are identified in the XRD pattern.
136 The authors acknowledge that many of the U-Si XRD peaks are convoluted
137 making identification and quantitative analysis of secondary U-Si phases dif-
138 ficult. Characteristic peaks for UO_2 were included as this phase is typical of
139 oxidized U-Si compounds [12, 13]. These specimens exhibited no evidence of
140 UO_2 presence.

141 Scanning electron microscopy (Inspect, FEI, Hillsboro, Oregon) and po-
142 larized light microscopy (Axio Imager.M2m, Carl Zeiss AB, Sweden) was also
143 used to further investigate the microstructures of the synthesized materials.
144 Samples of U_3Si_2 were cross sectioned, ground, and polished with $0.05 \mu\text{m}$
145 colloidal silica used as the final step. Although the majority of the polished
146 sample was observed as a single phase, one secondary phase was identified
147 using electron backscatter detection (see Figures 2 (a) and (b)). The precip-
148 itates appeared darker in contrast in the backscatter image indicating higher
149 silicon content and are observed as $5\text{-}15 \mu\text{m}$ in size. These precipitates con-
150 stituted less than one volume percent as established by the microstructural
151 analysis technique. Energy dispersive spectroscopy measurements confirmed
152 that this secondary silicide phase consisted only of U and Si, and was Si-
153 rich compared to the surrounding matrix. It is suspected that the secondary
154 phase is USi based on the accepted phase diagram [8]. Grain sizes for the
155 U_3Si_2 phase are approximately $35 \mu\text{m}$ in size.

156 *2.4. Differential scanning calorimetry*

157 Specimens for heat capacity measurements were prepared according to
158 §2.2 using a 5.2 mm punch and die set. Resulting cylindrical pellets weighed
159 0.215 g and had diameters of 4.46 mm $\pm$ 0.01 mm and thickness of 1.21 mm
160 $\pm$ 0.01 mm. Measurements in the DSC used Pt pans with Al₂O₃ liners, which
161 prevented reaction of the U₃Si₂ sample with the pans. Specimen mass varied
162 by <0.01 mg between measurements.

163 The ratio method was utilized to determine the molar heat capacity,* C_p ,
164 of the specimens in a differential scanning calorimeter (Pegasus 404C, Net-
165 zsch Instruments, Germany). Measurements were conducted at a heating
166 rate of 20 K·min⁻¹ to a final temperature of 1243 K. Three separate mea-
167 surements were conducted to indicate the error of the heat capacity mea-
168 surement, which was determined to be 3% in this study. The atmosphere in
169 the DSC was comprised of ultrahigh purity Ar(g) that flowed through a Cu
170 getter source (2A-100, Centorr Vacuum Industries, USA), which decreased
171 the O₂(g) content in the gas stream to 10⁻¹⁵ ppm. Oxygen content on the
172 gas stream was monitored on the exhaust by an in-stream partial pressure of
173 O₂(g), p_{O_2} , sensor (Rapidox 3100 OEM, Cambridge Sensotec, UK) to verify
174 minimal O₂(g) contamination in the furnace chamber. Thermocouple cal-
175 ibrations were conducted using melt point standards over the entire range
176 of temperatures investigated. Based on the temperature calibrations, the
177 instrument had an error in temperature of $\pm$ 1 K.

178 *2.5. Dilatometry*

179 The thermal expansion specimen was prepared according to §2.2 using
180 a 5.2 mm punch and die set. Mass of the thermal expansion specimen was
181 0.899 g and had diameters of 4.42 mm $\pm$ 0.01 mm and thicknesses of 4.90 mm
182 $\pm$ 0.02 mm.

183 Measurement of the physical coefficient of thermal expansion, α_p , was de-
184 termined using a dilatometer (DIL 402CD, Netzsch Instruments, Germany).
185 A heating rate of 2.5 K·min⁻¹ was used to a maximum temperature of 1732
186 K. A baseline correction was applied to the data to correct for the thermal
187 expansion of the Al₂O₃ fixturing. The Ar atmosphere was monitored with a
188 p_{O_2} sensor on the outlet. Initial p_{O_2} values were 0.1-1 ppm, which dropped

*Specific heat capacity values were determined in the instrument, which was converted to molar values using a molecular weight of 770.26 g mol⁻¹.

189 to 10^{-12} ppm during the measurement and returned to the initial value on
 190 cooling. Specimen mass increased by <0.5 mg between measurements, which
 191 can be attributed to surface oxidation of the specimen. Four separate α_p
 192 measurements using identical profiles and baseline corrections were used to
 193 determine the standard error of 8%.

194 2.6. Thermal diffusivity measurements

195 A cylindrical specimen was fabricated according to §2.2 using a 13 mm
 196 punch and die set for LFA measurements. The specimen weight was 2.55 g
 197 with a thickness of $2.24 \text{ mm} \pm 0.02 \text{ mm}$. Pellet diameters were $11.0 \text{ mm} \pm 0.1$
 198 mm in size. Diffusivity values were determined by fitting the temperature
 199 rise signal with a modified Cape-Lehman model [14]. Calibration of the
 200 thermocouple used to measure the sample temperature was achieved using
 201 the Curie temperature of Fe; the minimum in the diffusivity curve produced
 202 was located at 1045.2 K, slightly above the accepted value of 1043 K. As such,
 203 an uncertainty of ± 3 K is ascribed to the temperatures of each measurement.

204 The LFA (LFA 427, Netzsch Instruments, Germany) method was utilized
 205 to determine the thermal diffusivity, D , of the specimens. In this technique,
 206 a laser is pulsed on one side of the specimen, while the heat rise is determined
 207 on the opposite side by measuring the infrared blackbody radiation. All sur-
 208 faces of the sample were coated with a graphite coating (Graphit 33, Kontakt
 209 Chemie, Germany) to improve absorption of the laser energy on the bottom
 210 face and enhance emissivity of the upper face. Diffusivity data was collected
 211 from room temperature to 1773 K in 50 K increments on cooling, collecting
 212 three data points per temperature and averaged for the reported value. Ad-
 213 ditional data was collected on heating and were within the experimental error
 214 of the technique (3%). The reported data in this study is limited to values
 215 taken on cooling. Al_2O_3 fixturing was used throughout the sample chamber,
 216 and gettered argon was again flowed throughout the measurement. Oxygen
 217 content of the outlet gas stream was monitored with a p_{O_2} sensor during each
 218 measurement and was $< 10^{-15}$ ppm for all measurements. Changes in sample
 219 mass were 1-3 mg, which is attributed to a limited surface reaction with the
 220 graphite coating. Error introduced from thermal expansion of the specimen
 221 was calculated from $D = 0.1388 \cdot d^2 \cdot t_{1/2}^{-1}$ [15] using the α_p value measured in
 222 this study, where d is the thickness and $t_{1/2}$ is the half-time. Error result-
 223 ing from thermal expansion is calculated at less than 1.5% over the entire
 224 temperature range and was not included in this study.

3. Results and Discussion

3.1. Density and coefficient of thermal expansion

Specimen length change (L/L_0) and α_p are shown in Figure 3 as a function of temperature to 1673 K. Expansion of U_3Si_2 is observed to depend linearly with temperature. Fitting dL/L_0 with a linear regression analysis produces a coefficient of determination, $R^2 = 0.998$. Calculating the instantaneous derivative of dL/L_0 at a given temperature produces α_p , shown by ($\square$) in Figure 3. The physical coefficient of thermal expansion for this data set has a mean value of $16.1 \times 10^{-6} \text{ K}^{-1} \pm 1.3 \times 10^{-6} \text{ K}^{-1}$. The mean ($\bar{\alpha}_p$) and standard deviation (σ) values are represented in Figure 3 by the (—) and (---) lines, respectively. Good agreement is observed with the data set collected here except for the values near 1000 K. For the U_3Si_2 phase, there is no physical explanation for deviations in this temperature range, and it is expected that this observation is an experimental anomaly.

Loch *et al.* determined the technical coefficient of thermal expansion referenced to 273 K as a function of temperature to 1223 K, which varied from $15.2 \times 10^{-6} \text{ K}^{-1}$ to $14.6 \times 10^{-6} \text{ K}^{-1}$ and is within 9% of the value determined in this study [16]. Specific details regarding the sample fabrication route and microstructural information were not included in Loch's investigation making comparison difficult with this and other investigations in the literature. Shimizu measured $\alpha_p = 17.3 \times 10^{-6} \text{ K}^{-1}$ for arc-cast U_3Si_2 to 1473 K, which is in good agreement with the values measured in this study [17]. Taylor and McMurtry measured the thermal expansion on sintered U_3Si_2 specimens and determined a value of $17.3 \times 10^{-6} \text{ K}^{-1}$, which is within 6.8% of the value determined in this study [18]. Qualitatively, the microstructures provided in Shimizu's study are typical of arc-cast specimens, while Taylor and McMurtry's micrographs are similar in appearance to those provided in Figure 2. Possible sources for the differences between the thermal expansion provided in this dataset and the references detailed above is not possible given the limited characterization of microstructural and phase information in the literature.

Calculation of the density as a function of temperature is achieved in this study using Equation 1:

$$\rho = \frac{\rho_0}{[1 + \alpha_p (T - T_0)]^3}, \quad (1)$$

where ρ_0 is the density determined at the reference temperature, T_0 . The calculated mean value of α_p provided previously was used to determine the

258 density as a function of temperature. An error of <0.5% is calculated for the
 259 density value based on a propagation of error analysis using the prescribed
 260 error in α_p .

261 3.2. Heat capacity of U_3Si_2

262 The heat capacity of U_3Si_2 as determined by the ratio method in the
 263 DSC is shown in Figure 4 as a function of temperature to 1240 K following
 264 conversion of the experimental data (collected as specific heat capacity, c_p)
 265 to molar heat capacity (C_p). Use of heat capacity data for engineering ap-
 266 plications is typically more direct when expressed as specific heat capacity.
 267 The collected c_p values are summarized and tabulated in Appendix A.

A linear regression analysis of the data is shown as the red line, which is represented by:

$$C_p = 0.02582 \times T + 140.5 \text{ (J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}) \quad (2)$$

268 with an $R^2 = 0.972$, which is within 3% of the experimental data over the
 269 entire temperature range. Equation (2) is used to calculate the thermal con-
 270 ductivity values presented in the following section. No literature values for
 271 U_3Si_2 heat capacity could be located. It is common to encounter approxi-
 272 mations for the heat capacity of U_3Si_2 in the literature derived from a study
 273 executed by Cape and Taylor [19]. However, the samples evaluated in their
 274 study are reported to have contained 6.1 w/o Si, significantly U-rich com-
 275 pared with the composition of U_3Si_2 . Heat capacity data from Cape and
 276 Taylor’s study (shown in Figure 4) has a considerably larger slope than the
 277 value presented in this study and is likely a result of the differing composi-
 278 tions. A more recent thermochemical database [20] exhibits good agreement
 279 over the entire temperature range with the current dataset.

280 Heat capacity in metallic systems is typically comprised of lattice, di-
 281 lational, and electronic components. The Debye-like lattice contribution is
 282 typically saturated to the Dulong–Petit value ($124.71 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$) above the
 283 Debye temperature, which is unknown for U_3Si_2 . Based on other U–Si binary
 284 compound Debye temperature values [13, 21], it is estimated that U_3Si_2 has
 285 an approximate Debye temperature of 200 K, which is below the temper-
 286 ature range investigated in this study. Contributions to the heat capacity
 287 from the dilational term are approximated using the well-known formula
 288 $C_d = \alpha_p^2 V k_T^{-1} T$, where V and k_T are the molar volume and isothermal com-
 289 pressibility, respectively. Values for k_T were not found during the literature

review. Approximate values were calculated using Young's modulus of 128 GPa and a Poisson ratio, ν , of 0.185 [18] by applying $k_T = 3(1 - 2\nu) \cdot E^{-1}$. Using the aforementioned approximations, the dilational heat capacity term contributes <0.7% over the entire temperature range. Electronic contributions to heat capacity for metallic materials depend linearly with temperature and can be represented by $C_e = \gamma T$. Analysis of Equation (2) suggests that $\gamma = 52.0 \text{ mJ}\cdot\text{mol}^{-1}\cdot\text{K}^{-2}$, given that the Debye component is saturated. No γ values exist in the literature for U_3Si_2 to the authors' knowledge.

3.3. Thermal diffusivity and conductivity of U_3Si_2

Thermal diffusivity data collected in the LFA on U_3Si_2 are shown in Figure 5 as a function of temperature to 1773 K. Effects of probable second phase USi as identified by microscopy in these specimens are considered to be negligible given the low fraction present in the specimens as discussed in §2.3. Thermal transport for U_3Si_2 increases non-linearly with temperature over the entire range investigated in this study. No thermal diffusivity data was found in the literature for comparison.

The product of density, specific heat capacity, and thermal diffusivity produces the thermal conductivity, which is shown in Figure 6. Thermal conductivity values have also been corrected for porosity, P , using:

$$\lambda_{TD} = \frac{\lambda_{exp}}{1 - 3/2(1 - P)}, \quad (3)$$

where λ_{TD} and λ_{exp} are the thermal conductivity values for a theoretically dense specimen and experimental values, respectively. The error bars in Figure 6 represent a propagation of error analysis for the three techniques used in this study, which calculates $\Delta\lambda/\lambda = 5\%$. A linear regression analysis of the thermal conductivity data is shown in Figure 6, which is represented by:

$$\lambda = 0.0151 \times T + 6.004 \text{ (W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}\text{)}, \quad (4)$$

where λ is the thermal conductivity and $R^2 = 0.997$.

Shimizu fabricated arc-cast U_3Si_2 specimens that were tested at three separate institutions using either an axial ($\circ$ and ∇) or radial ($\triangle$) heat flow method (see Figure 6) [17]. Low magnification micrographs of pre-irradiated U_3Si_2 microstructures did not exhibit significant cracking, although it is expected that some degree of microcracking exists in Shimizu's specimens

321 similar to the buttons produced in this study. This would likely explain
 322 the lower thermal conductivity values determined by Shimizu relative to this
 323 study. An induction cast specimen measured by the radial flow method (Δ)
 324 in Shimizu’s study is considerably lower than this data set, although cracking
 325 was observed in those specimens. Data presented by Taylor and McMurtry
 326 on sintered specimens using the comparator method is considered erroneous
 327 given that the difference in thermal conductivity between the reference ma-
 328 terial (Ni) and U_3Si_2 is inadequate for a reliable measurement [17, 22].

329 Thermal transport in metallic conductors consists predominantly of phonon
 330 scattering and electronic contributions. It is assumed in this work that the
 331 phonon terms are negligible in this temperature range relative to the elec-
 332 tronic component. This approximation is qualitatively justified because the
 333 phonon thermal conductivity typically depends inversely with temperature,
 334 while the electronic term depends linearly as a function temperature. Linear
 335 electronic behavior is expressed by the well known Wiedemann-Franz law:

$$L_0 = \frac{\lambda}{\sigma \cdot T} = 2.45 \times 10^{-8} \text{ (V}^2 \cdot \text{K}^{-2}) \quad (5)$$

336 where L_0 and σ are the Lorenz number and electrical conductivity. Values
 337 for $L(T)$ were calculated from the electrical resistivity data on arc-cast U_3Si_2
 338 [17] and from the thermal conductivity collected in this study, and shown in
 339 the inset of Figure 6. Calculated values for $L(T)$ are consistently larger than
 340 the theoretical Lorentz value shown in Equation (5). Positive deviations
 341 from L_0 are possible in metals when higher order terms of Equation (5)
 342 are included [23]. However, it is suspected that based on the low thermal
 343 conductivity data presented by Shimizu would also lead to artificially high
 344 electrical resistivity data from measuring arc-cast specimens. Therefore, the
 345 Lorentz values calculated in this study are suspected to be overestimated.

346 4. Conclusions

347 The coefficient of thermal expansion, heat capacity, thermal diffusivity,
348 and thermal conductivity of U_3Si_2 were determined from room temperature
349 to 1773 K. A constant coefficient of thermal expansion was determined over
350 the entire temperature range of $16.1 \times 10^{-6} \text{ K}^{-1}$. Heat capacity data on the
351 stoichiometric material increased linearly with temperature and is consistent
352 with thermodynamic databases in the literature. Metallic thermal conduc-
353 tion is observed in U_3Si_2 which increases linearly with temperature.

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Table 1: Chemical analysis conducted on comminuted U_3Si_2 buttons. Elements included in the analysis, but below the detection limit are included as follows: Al, Sb, Ba, Be, Cd, Ca, Cu, Pb, Li, Mg, Mn, Mo, Ni, P, K, Se, Na, Sr, Sn, Ti, Zn, Zr, B, Gd, Nb, Ta, and W.

Elements	Si	C	H	N	Fe	As	Cr
Contents (w/o)	7.28	0.08	0.27	0.12	0.04	0.03	0.006

Figure 1: X-ray diffraction analysis of a comminuted U_3Si_2 button that was sealed within an atmosphere controlled polymer holder. Reference files are from: 01-091-2241 (U_3Si_2) [9], 04-008-2456 (UO_2) [24], 04-003-0522 (USi) [25].

Figure 2: Microstructure of sintered U_3Si_2 specimen captured in the light microscope using a polarizing light filter (a) to emphasize the grain size of the U_3Si_2 phase, while the precipitate phase is highlighted in (b) using a backscatter detector in the SEM.

Figure 3: Thermal expansion of U_3Si_2 as a function of temperature (—) along with α_p ($\square$) measured in this study. Literature values for α_p are provided from (—) — [18], (— · —) — [16], and (— · · —) — [17].

Figure 4: Molar heat capacity of U_3Si_2 collected as a function of temperature using the ratio method from 356 K to 1220 K. The calculated Dulong–Petit value for the system is included as a (– –). Literature data is included from (– · –) – [19] and (– · ·) – [20].

Figure 5: Thermal Diffusivity data collected on cooling for U_3Si_2 as a function of temperature from 273 K to 1773 K.

Figure 6: Calculated thermal conductivity data for U_3Si_2 as a function of temperature to 1773 K collected in this study ($\square$). Reference data from Shimizu [17] is represented by ($\circ$), (∇), and ($\triangle$). Literature values for UO_2 ($\star$) are included from [26]. An inset is provided to compare the expected Lorenz values ($-$) with calculated values ($-$).

432 **Appendix A. Tabulated Thermal Property Values Measured in this**
433 **Study**

Table A.2: Thermophysical property values of U_3Si_2 determined in this study. Values that are *italicized* have been extrapolated from experimental values, as described in the text. Density values have been calculated assuming a constant value of α_p .

T K	$D \times 10^6$ $\text{m}^2 \cdot \text{s}^{-1}$	c_p $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$	$\rho \times 10^3$ $\text{kg} \cdot \text{m}^{-3}$	λ_{TD} $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$
1775	11.6	<i>242.6</i>	11.2	32.1
1674	11.2	<i>239.2</i>	11.2	30.9
1574	10.8	<i>235.8</i>	11.3	29.4
1474	10.5	<i>232.5</i>	11.4	28.4
1374	10.1	<i>229.1</i>	11.4	26.9
1274	9.62	<i>225.8</i>	11.5	25.4
1174	9.14	213.5	11.5	23.9
1074	8.68	217.2	11.6	22.5
974	8.2	217.7	11.6	21.0
874	7.72	211.4	11.7	19.6
775	7.18	208.8	11.7	18.0
675	6.61	202.9	11.8	16.4
575	5.99	200.3	11.8	14.7
476	5.32	199.5	11.9	12.9
376	4.63	194.7	12.0	11.1
320	4.17	187.0	12.0	9.91