Thermal conductivity reduction in \((\text{Zr}_{0.25}\text{Ta}_{0.25}\text{Nb}_{0.25}\text{Ti}_{0.25})\text{C}\) high entropy carbide from extrinsic lattice defects

Cody A. Dennett\(^a\(^{,}\)\(^b\), Zilong Hua\(^a\), Eric Lang\(^c\), Fei Wang\(^d\) and Bai Cui\(^{d,e}\)

\(^a\)Materials Science and Engineering Department, Idaho National Laboratory, Idaho Falls, ID, USA; \(^b\)Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA, USA; \(^c\)Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, NM, USA; \(^d\)Department of Mechanical and Materials Engineering, University of Nebraska-Lincoln, Lincoln, NE, USA; \(^e\)Nebraska Center for Materials and Nanoscience, University of Nebraska-Lincoln, Lincoln, NE, USA

ABSTRACT
High entropy carbides ceramics with randomly-distributed multiple principal cations have shown high temperature stability, low thermal conductivity, and possible radiation tolerance. While chemical disorder has been shown to suppress thermal conductivity in these materials, little investigation has been made on the effects of additional, extrinsically-generated structural defects on thermal transport. Here, \((\text{Zr}_{0.25}\text{Ta}_{0.25}\text{Nb}_{0.25}\text{Ti}_{0.25})\text{C}\) is exposed to Zr ions to generate a micron-scale, structural-defect-bearing layer. The reduction in lattice thermal transport is measured using laser thermoreflectance. Conductivity changes from different implantation temperatures suggest dislocation loops contribute little to phonon scattering while nanoscale defects serve as effective scatterers, offering a pathway for thermal engineering.

1. Introduction
High entropy carbide ceramics (HECs) have emerged in the last several years as a promising class of materials for applications involving high temperatures or other environmental extremes. Borrowing from the maturing field of complexity-engineering and entropic-stabilization in metal alloys [1], new high entropy ceramics with complex selections of cations have emerged across oxides, borides, nitrides, sulfides, silicides, and carbides [2,3]. HECs, in particular, have been synthesized as bulk specimens and as thin films and have been shown to have a variety of physical characteristics attributed to the presence of a disordered cation sublattice including high hardness [4–6], low thermal conductivity [7,8], oxidation resistance [9–11], and radiation tolerance [12].

To date, experimental studies of the thermal transport characteristics of these HEC materials have been relatively few and primarily focused on five-cation chemistries including \((\text{Hf}_{0.2}\text{Zr}_{0.2}\text{Ta}_{0.2}\text{Nb}_{0.2}\text{Ti}_{0.2})\text{C}\) [7,8,13] and \((\text{Hf}_{0.2}\text{Zr}_{0.2}\text{Ta}_{0.2}\text{Mo}_{0.2}\text{W}_{0.2})\text{C}\) [8]. These studies have exclusively focused on the thermal transport characteristics of as-synthesized materials in addition to their resulting structure. While some progress has been made in computationally treating transport in these systems, large discrepancies exist in the estimation of the lattice contribution to thermal conductivity between simulation and experiment [14]. While noted as a promising feature of these systems, the ability to thermally engineer HECs for targeted applications has not yet been exploited in a systematic manner.
In particular, the interplay between chemical disorder, stoichiometry, and structural defects in relation to thermal transport has yet to be explored in HECs or, to a large extent, in any high entropy ceramic. Frameworks for understanding the effects of structural defects, and tailoring performance by extrinsically introducing such defects, are well-established for single-principal-element materials and compounds [15,16]. Effects on electron and phonon thermal conductivity are routinely expressed in terms of the scattering strength of structural defects of particular types including 0D point defects, 1D dislocations, 2D dislocation loops or boundaries, and 3D inclusions [17–20]. However, how thermal carriers scatter from each of these classes of structural features in the limit of maximized chemical disorder is not yet well understood. For HECs in particular, the relative partitioning of these thermal carriers (electrons and phonons) depends sensitively on stoichiometry [8] and introduces additional challenges for developing a complete understanding of transport phenomena. However, controlling carrier partitioning, therefore, could also eventually serve as a tool to engineer thermal properties.

Here, the coupled effects of chemical disorder and extrinsically-imposed lattice defects are studied in \((Zr_{0.25}Ta_{0.25}Nb_{0.25}Ti_{0.25})C\). This four-cation HEC was originally synthesized Hf-free by Wang and coworkers to study its potential, if any, for use in nuclear systems, as Hf has a large neutron absorption cross section [12]. Specimens of \((Zr_{0.25}Ta_{0.25}Nb_{0.25}Ti_{0.25})C\) were exposed to Zr ion beam irradiation to generate a micron-scale defect-bearing layer at the surface of bulk specimens at a series of different temperatures. Following exposure, a spatial domain thermal reflectance (SDTR) method is used to determine the total thermal conductivity of the defect-bearing region using a multi-layer thermal transport model. A combination of SDTR and electrical resistivity measurements on a pristine bulk HEC are used to determine the total heat capacity and estimate the electron contribution to thermal conductivity. The resulting thermal performance is interpreted in the context of previously-measured concentrations of defects from electron microscopy, showing that dislocation loops serve as much less effective thermal scatters in these chemically disordered structures than would be expected from classical phonon scattering theory.

### 2. Materials and methods

The \((Zr_{0.25}Ta_{0.25}Nb_{0.25}Ti_{0.25})C\) specimens investigated in this work have been described previously [12], so only a brief summary of their synthesis and ion beam processing will be described here. Bulk specimens were fabricated from commercial powders of the constituent binary carbides through ball milling the as-received powders and consolidation with spark plasma sintering (SPS). Scanning electron microscopy indicated an as-synthesized mean grain size of 19 \(\mu\)m. X-ray diffraction patterns of as-synthesized specimens are indexed as a single-phase rock-salt structure, similar to other multication carbides [12]. The theoretical density is calculated from the XRD lattice constant as 8.46 g/cm\(^3\) and the measured density of the bulk materials is 8.25 g/cm\(^3\), resulting in a relative density of 97.5%. Mechanically polished specimens were exposed to 3 MeV Zr\(^{2+}\) ions to a total fluence of \(8.0 \times 10^{15}\) ions/cm\(^2\) at three temperatures, 25, 300, and 500 °C. Ions at this energy generate structural defects over an \(\sim 1.5 \mu\)m depth at the sample surface, with peak defect production at \(\sim 800\) nm [21,22]. Extensive transmission electron microscopy (TEM) characterization by Wang et al. showed the generation of a population of both perfect and faulted Frank loops with consistently small diameters (\(\sim 2\) nm) at all three temperatures, with density peaking near the peak defect generation region, and no other visible defects [12]. A spatially-averaged summary of the measured dislocation loop size and density is listed in Table 1, noting that no loops were observed in the as-synthesized material. No radiation-induced chemical segregation was observed near the grain boundaries for the as-synthesized or irradiated materials.

| Irradiation Temperature | Ion Fluence | Ave. Loop Diameter \(d\) | Ave. Loop Density \(n\) |
|-------------------------|-------------|--------------------------|------------------------|
| 25°C                    | \(8 \times 10^{15}\) ions/cm\(^2\) | 1.6 nm                   | \(9.5 \times 10^{17}\) loops/cm\(^3\) |
| 300°C                   | \(8 \times 10^{15}\) ions/cm\(^2\) | 2.0 nm                   | \(7.4 \times 10^{17}\) loops/cm\(^3\) |
| 500°C                   | \(8 \times 10^{15}\) ions/cm\(^2\) | 2.1 nm                   | \(6.0 \times 10^{17}\) loops/cm\(^3\) |

For both pristine and ion irradiated \((Zr_{0.25}Ta_{0.25}Nb_{0.25}Ti_{0.25})C\), SDTR is used to extract thermal properties including thermal conductivity, \(\kappa\), thermal diffusivity, \(D\), and, in the case of the as-synthesized material, the specific heat capacity, \(C_P\). In SDTR, periodic local temperature variations are induced using an intensity-modulated 660 nm CW heating laser and detected using a 532 nm CW probing laser through the thermoreflectance effect [23,24]. The heating and probing lasers are both focused to spot sizes on the order \(\sim 1\) \(\mu\)m using a high-power optical objective and the probing laser scanned across a distance of \(\sim 20\) \(\mu\)m centered on the heating spot. By coating specimens with thin transducer layers of gold, a strong thermoreflectance response at 532 nm
is ensured. The relative phase lag between the heating and probe laser as a function of separation distance at several modulation frequencies in the range 5–100 kHz is recorded using lock-in detection [23]. A multi-layer thermal model taking into account the thermal properties of an arbitrary number of layers using the thermal quadrupole method is used to return best-fit values for the properties of selected layers [25,26].

The thermal layering model used for pristine and as-irradiated (Zr0.25Ta0.25Nb0.25Ti0.25)C is shown in Figure 1. For pristine specimens, Figure 1(a), a relatively thick 81 nm Au film was deposited in order to achieve the highest joint sensitivity to thermal conductivity and thermal diffusivity based on analysis shown in the Supplementary Material. With known density, independent thermal diffusivity based on analysis shown in the Supplementary MATER. RES. LETT. is assumed constant for both defect-bearing and pristine layers, as there is little evidence to suggest that heat capacity is meaningfully affected by structural defects. In each optimization, the thermal parameters of the pristine substrate layer are fixed to those measured independently on the pristine specimen and \( D \) and \( \kappa \) optimized self-consistently for the defect-bearing layer. A table of the fixed and varying optimization parameters used in the two- and three-layer models are detailed in the Supplementary Materials.

Figure 1. Thermal layering model used for pristine (a) and ion irradiated (b) HECs, overlaid with the defect generation profile from Zr ion implantation. The thick gold transducer layer used on the pristine specimen allows for all three thermal parameters, \( \kappa \), \( D \), and \( C_p \), to be extracted from the two-layer thermal model. The defect generation profile and ion irradiated layer are to scale, while the gold transducer thicknesses are not.

To ensure this parameterization results in the highest sensitivity to the thermal properties of the defect-bearing layer, a detailed sensitivity analysis is carried out, see Figure 2. Relative sensitivity is calculated for \( D \) and \( \kappa \) of the irradiated layer, the thickness of the irradiated layer, and the thickness of the gold transducer film as

\[
S(\xi) = \frac{\varphi(\xi - \Delta\xi) - \varphi(\xi)}{(\Delta\xi/\xi)},
\]

where \( \varphi(\xi) \) is the spatially-varying thermal phase delay and \( \xi \) is the parameter in question. Figure 2 is calculated for a baseline set of parameters \( D = 3.70 \text{ mm}^2/\text{s} \) and \( \kappa = 9.14 \text{ W/m} \cdot \text{K} \) for the pristine materials, with a 10% reduction in \( D \) and \( \kappa \) in the defect-bearing layer, and assuming a 14 nm thick deposited gold film (with corresponding thermal conductivity [28,29]). An \( \sim 10 \text{ nm} \) gold film was chosen as the set point as the sensitivity to a ‘sandwiched’ thermal layer is higher when the transducer layer is thin; 14 nm was eventually deposited on these irradiated samples. As can be seen in Figure 2, the highest sensitivity at both the intermediate and highest modulation frequencies used, 20 kHz and 100 kHz, is to the thermal properties of the defect-bearing layer. Only very weak sensitivities to the precise gold coating thickness and segmented irradiated layer thickness are observed. Together, these sensitivities ensure that the values reported from the three-layer optimization are representative of the defect-modified thermal characteristics.

Total thermal conductivity as measured in these carbide ceramics contains contributions from both electron and lattice (or phonon) thermal transport [8]. To extract the relative contribution of electrons to thermal conductivity, the electrical resistivity of the un-coated side of the pristine bulk (Zr0.25Ta0.25Nb0.25Ti0.25)C specimen was measured using a 4-point probe. Traditionally, the electrical resistivity can be used to estimate the electronic thermal conductivity as \( \kappa_e = \frac{LT}{\rho_\epsilon} \), where most commonly \( L = L_0 = 2.44 \times 10^{-8} \text{ W} \Omega \text{K}^{-2} \) is the

\[
\rho_\epsilon = \frac{\Delta 3 \text{ MeV } 2^+}{\text{ damage profile}}
\]

\[
\kappa = 9.14 \text{ W/m} \cdot \text{K}
\]

\[
D = 3.70 \text{ mm}^2/\text{s}
\]

\[
\kappa_e = \frac{LT}{\rho_\epsilon}
\]

\[
\rho_\epsilon = 2.44 \times 10^{-8} \text{ W} \Omega \text{K}^{-2}
\]

\[
\kappa_e = \frac{LT}{\rho_\epsilon}
\]
Figure 2. Sensitivity of the three-layer thermal model to several parameters at both 20 kHz and 100 kHz thermal wave modulation frequencies in terms of phase delay difference as a function of SDTR scan distance, the distance between heating and probe laser spots. Initial parameters are taken as 14 nm of Au coating, fixed values of semi-infinite substrate thermal properties, and a 10% reduction in conductivity in the middle, defect-bearing layer. To ensure a constant heat capacity, the values of \( D \) and \( \kappa \) in the defected layer are changed in the same proportion in this analysis. Of these parameters, the highest sensitivity is to the thermal properties of the layer of interest.

Lorentz number and \( \rho_e \) is the electrical resistivity. However, in single-element metal carbides, \( L_0 \) has been shown to overestimate \( \kappa_e \), such that an effective Lorentz number as calculated by Makinson should be used [8,30,31]. A description of the 4-point probe measurements as well as the conversion of \( \rho_e \) to \( \kappa_e \) is provided in the Supplementary Materials. From \( \kappa_e \) and the measured total thermal conductivity, \( \kappa_{\text{tot}} \), the phonon thermal conductivity can be estimated as \( \kappa_p = \kappa_{\text{tot}} - \kappa_e \). The relative electron contribution to \( \kappa_{\text{tot}} \) has been shown to be a sensitive function of carbon stoichiometry, and therefore bonding character, in binary carbides and these HECS [8,32]. The carbon stoichiometry of as-synthesized (Zr\(_{0.25}\)Ta\(_{0.25}\)Nb\(_{0.25}\)Ti\(_{0.25}\))C is estimated using X-ray photoemission spectroscopy (XPS) to place measured values of \( \kappa_e \) in context. As extraction of the electrical resistivity of solely the defect-bearing layers in the irradiated materials was not possible from bulk specimens, \( \kappa_e \) is assumed to be constant for the defect-bearing material for this initial investigation. This practical limitation will lead to an overestimation of the lattice thermal conductivity reduction, as both electrons and phonons are known to scatter from point defects in refractory carbide and nitride ceramics [31].

3. Results and discussion

SDTR measurements of pristine (Zr\(_{0.25}\)Ta\(_{0.25}\)Nb\(_{0.25}\)Ti\(_{0.25}\))C with a thick gold transducer return room-temperature thermal property values of \( \kappa_{\text{tot}} = 9.14 \pm 0.20 \text{ W/m} \cdot \text{K} \) and \( D = 3.70 \pm 0.06 \text{ mm}^2/\text{s} \), which imply a specific heat capacity of \( C_p = 293 \pm 41 \text{ J/kg} \cdot \text{K} \), with all uncertainties listed as the standard error of multiple spatially-varying measurements. Although no direct comparisons exist for this 4-cation HEC in the literature, these values are consistent with the thermal properties measured in bulk (Hf\(_{0.2}\)Zr\(_{0.2}\)Ta\(_{0.2}\)Nb\(_{0.2}\)Ti\(_{0.2}\))C synthesized using the same procedure [7]. Table 2 compares the as-measured values of the 4- and 5-cation HEC thermal properties. That the 4-cation HEC measured here retains a higher thermal conductivity is consistent with the lower levels of mass and force constant scattering expected with one fewer principal elements. In addition, the distinctly higher value for \( C_p \) is consistent with excluded cation, Hf, having the lowest \( C_p \) of any of the constituent binary carbides [7]. Confirmation measurements of \( C_p \) made using DSC returned a value \( 310 \pm 5 \text{ J/kg} \cdot \text{K} \) at \( \sim 335 \text{ K} \), consistent with the theoretically monotonically-rising heat capacity in refractory carbides in this temperature range [33].

4-point probe measurements indicated that pristine (Zr\(_{0.25}\)Ta\(_{0.25}\)Nb\(_{0.25}\)Ti\(_{0.25}\))C specimens have an electrical resistivity \( \rho_e = 149 \pm 2 \text{ \Omega} \cdot \text{cm} \). This is higher than that of binary carbides such as TiC and TaC (68 and 25 \( \mu \Omega \cdot \text{cm} \), respectively [34]), similar to that for (Hf\(_{0.2}\)Zr\(_{0.2}\)Ta\(_{0.2}\)Nb\(_{0.2}\)Ti\(_{0.2}\))C reported as \( \sim 115 \mu \Omega \cdot \text{cm} \) by Wen and coworkers for sintered pellets [13], but lower than the range reported for thick films of (Hf\(_{0.2}\)Zr\(_{0.2}\)Ta\(_{0.2}\)Mo\(_{0.2}\)W\(_{0.2}\))C\(_{1-x}\) by Rost and coworkers as a function of carbon stoichiometry as \( \sim 200–1200 \mu \Omega \cdot \text{cm} \) [8]. That the measured resistivity is low compared to the range reported for precisely carbon-controlled HECS implies the final, as-synthesized bulk samples are possibly carbon deficient and retain some metallic bonding that would not be present in a perfectly stoichiometric compound. From \( \rho_e, \kappa_e \) is estimated as 3.7 \text{ W/m} \cdot \text{K} , indicating that 40% of the thermal conductivity is due to electrons in the as-synthesized material and 60% is due to the lattice. From XPS measurements, the metal to carbon ratio is M:C \( \approx 1 \), as detailed in the Supplementary Materials, indicating that an approximately stoichiometric carbide has been maintained. While Rost and coworkers reported generally higher resistivity for stoichiometric

\[ D | \text{mm}^2/\text{s} | \kappa_{\text{tot}} | \text{W/m} \cdot \text{K} | C_p | \text{J/kg} \cdot \text{K} \]

| 4-cation\(^a\) | 3.70 | 9.14 | 293 |
| 5-cation\(^b\) | 3.60 | 6.45 | 191 |

\(^a\)(Zr\(_{0.25}\)Ta\(_{0.25}\)Nb\(_{0.25}\)Ti\(_{0.25}\))C \[^b\](Hf\(_{0.2}\)Zr\(_{0.2}\)Ta\(_{0.2}\)Nb\(_{0.2}\)Ti\(_{0.2}\))C [7]
compounds, the deposition of excess, non-metal-bonded carbon in sputter deposited materials complicates a direct comparison [8]. Further correlated microscopy and XPS should be carried out in the future on bulk, sintered ceramics to more fully characterize possibly heterogeneous carbon stoichiometry following synthesis. To ensure that thermal property measurements account for any such variability, ten or more randomly-selected spatially varying measurements are made on each specimen prior to reporting statistical averages.

Using the three-layer thermal transport model as described above, Figure 3(a) shows the total and phonon thermal conductivities as a function of irradiation temperature for the fixed ion fluence applied to each specimen. The electronic contribution is plotted as the constant value estimated for the as-synthesized material. The general trend of a larger decrease in thermal conductivity at lower irradiation temperatures is consistent with expectations from most material systems where extrinsic structural defect production is driven by radiation bombardment. Namely, higher irradiation temperatures increase the mobility and therefore the recombination of any Frenkel defects or small clusters generated by displacement damage, leaving fewer thermal scattering sites [28,35,36]. Using the simplified assumption that $\kappa_e$ is unaffected by structural defects and the entire reduction is due to phonon scattering, the fractional reduction in lattice thermal conductivity is plotted in Figure 3(b).

The behavior of the lattice thermal conductivity in the presence of these structural defects is most easily explored using the simplified Klemens-Callaway-Debye model where

$$\kappa_p = \frac{1}{3} \int_0^{\omega_{\text{max}}} d\omega C(\omega)v_g(\omega)^2\tau(\omega),$$

(2)

and $C(\omega)$, $v_g(\omega)$, and $\tau(\omega)$ are the phonon-frequency-dependent heat capacity, group velocity, and relaxation time, respectively [37]. The overall relaxation time is normally considered as a combination of different scattering mechanisms, phonon-phonon scattering, impurity scattering, etc., as summed using Matthiessen’s rule. Classical models have been used with success to describe scattering from both perfect (pl) and faulted (fl) dislocation loops in terms of the density and average size of the dislocation loops present as

$$\tau_{\text{pl}}^{-1} \propto nd_l\omega^2,$$

(3)

and

$$\tau_{\text{fl}}^{-1} \propto nd_f^2\omega^2,$$

(4)

where $n$ is the dislocation loop density and $d_l$ is the loop diameter, with each relationship containing prefactors, not shown here, to maintain the correct dimensionality [16]. As Wang and coworkers showed that some number of both perfect and faulted loops were present in these specimens [12], it is useful to compare the products $nd_l$ and $nd_f^2$ to compare the relative effects of extrinsic dislocation loops on $\kappa_p$. This comparison is shown in Table 3 and demonstrates that scattering from either perfect or faulted dislocation loops should be nearly identical in each of the three specimens with extrinsic structural defects measured here.

This theoretically identical scattering strength from loops, coupled with the similarly in thermal conductivity...
between the pristine sample and that exposed at 500 °C, implies that dislocation loops are effectively not scattering phonons in this chemically complex ceramic. This conclusion is reached by noting that at 500 °C, few residual point defects are expected following irradiation given the increased defect mobility at high temperatures. These data suggest, therefore, that the interactions between phonons and dislocation loops is inherently distinct in materials with a high degree of chemical complexity compared with traditional ceramics. Analytical dislocation loop scattering models, after Klemens, propose that the long-range strain field associated with dislocations is responsible for the majority of phonon scattering [18,38]. Here, in a maximally-chemically disordered carbide ceramic, dislocations appear to contribute little to phonon scattering, implying their strain fields are suppressed. This long-range suppression could occur as a result of the inherent strain associated with the disordered cation sublattice, although further experimental and computational work is required to investigate these effects directly.

Even though the reduction in $\kappa_p$ is likely overestimated here due to assumption of constant $\kappa_e$ across exposure conditions, electrons scatter little from extended loops. Therefore, the conclusion that residual nanoscale structural defects are the primary contributors to conductivity reduction remains unchanged. Altogether, that lattice thermal conductivity is reduced by ~20% at 25 °C, in a microstructure expected to contain a high density of both nanoscale and extended defects, supports the conjecture that heat carrier scattering from extrinsic nanoscale defects (point defects and small clusters below TEM resolution) can be used as a tool with which thermal conductivity may be engineered in these systems. Although ion beam irradiation was used to generate extrinsic structural defects in this work, other post-synthesis processing treatments could similarly be used for such defect injection and thermal engineering in the future.

### 4. Conclusion

Here, the impact of lattice defects extrinsically-generated using ion irradiation on thermal transport in a chemically complex carbide ceramic with four principal cations has been systematically studied using a laser thermoreflectance technique. Multi-layer thermal modeling allows the defect-affected thermal conductivity of a micron-thick damaged layer generated by ion implantation to be measured. Additional electrical resistivity measurements on as-synthesized material shows that this HEC has a thermal carrier partitioning, between electrons and phonons, in line with that previously observed in similar materials. The reduction in thermal conductivity is observed to be greatest in materials irradiated at low temperatures where a significant population of retained nanoscale defects is expected. At high temperatures, however, little reduction in overall or phonon thermal conductivity is observed. Coupled with prior investigations showing that small dislocation loops have formed under these exposure conditions at all temperatures, these observations indicate that dislocation loops are contributing little to phonon scattering, likely due to a suppression of their long-range strain fields. Nanoscale defects still serve as effective scatterers of thermal carriers in this system, opening a pathway for controlled thermal transport engineering in the presence of maximized chemical disorder in high entropy ceramics.

### Acknowledgments

The authors would like to thank S.G. Rosenberg and M. Meyerman at SNL for performing XPS measurements. The views expressed in this article do not necessarily represent the views of the U.S. DOE of the United States Government.

### Disclosure statement

No potential conflict of interest was reported by the author(s).

### Funding

This work was supported through the INL Laboratory Directed Research & Development Program under U.S. Department of Energy Idaho Operations Office Contract DE-AC07-05ID14517. C.A.D. and Z.H. acknowledge support from the Center for Thermal Energy Transport under Irradiation (TETI), an Energy Frontier Research Center funded by the US Department of Energy, Office of Science, Office of Basic Energy Sciences. This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the DOE Office of Science. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. DOE’s National Nuclear Security Administration under contract DE-NA-0003525.

### ORCID

Cody A. Dennett [http://orcid.org/0000-0003-2989-9550](http://orcid.org/0000-0003-2989-9550)
Eric Lang [http://orcid.org/0000-0001-5136-1875](http://orcid.org/0000-0001-5136-1875)
Bai Cui [http://orcid.org/0000-0002-0585-6698](http://orcid.org/0000-0002-0585-6698)
References

[1] Miracle DB, Senkov ON. A critical review of high entropy alloys and related concepts. Acta Mater. 2017;122:448–511.
[2] Oses C, Toher C, Curtarolo S. High-entropy ceramics. Nat Rev Mater. 2020;5:295–309.
[3] Feng L, Fahrenholz WG, Brenner DW. High-entropy ultra-high-temperature borides and carbides: a new class of materials for extreme environments. Annu Rev Mater Res. 2021;51(1):165–185.
[4] Sarker P, Harrington T, Toher C, et al. High-entropy high-hardness metal carbides discovered by entropy descriptors. Nature Commun. 2018;9:4980.
[5] Ye B, Wen T, Nguyen MC, et al. First-principles study, fabrication and characterization of (Zr0.25Nb0.25Ta0.25V0.25)C high-entropy ceramics. Acta Mater. 2019;170:15–23.
[6] Harrington TJ, Gild J, Sarker P, et al. Phase stability and mechanical properties of novel high entropy transition metal carbides. Acta Mater. 2019;166:271–280.
[7] Yan X, Constantin L, Lu Y, et al. (Hf0.2Zr0.2Ta0.2Nb0.2Ti0.2)C high-entropy ceramics with low thermal conductivity. J Am Ceram Soc. 2018;101(10):4486–4491.
[8] Rost CM, Borman T, Hossain MD, et al. Electron and phonon thermal conductivity in high entropy carbides with variable carbon content. Acta Mater. 2020;196:231–239.
[9] Zhou J, Zhang J, Zhang F, et al. High-entropy carbide: a novel class of multicomponent ceramics. Ceram Int. 2018;44(17):22014–22018.
[10] Ye B, Wen T, Liu D, et al. Oxidation behavior of (Hf0.2Zr0.2Ta0.2Nb0.2Ti0.2)C high-entropy ceramics at 1073–1473 K in air. Corros Sci. 2019;153:327–332.
[11] Ye B, Wen T, Chu Y. High-temperature oxidation behavior of (Hf0.2Zr0.2Ta0.2Nb0.2Ti0.2)C high-entropy ceramics in air. J Am Ceram Soc. 2020;103(1):500–507.
[12] Wang F, Yan X, Wang T, et al. Irradiation damage in (Zr0.25Ta0.25Nb0.25Ti0.25)C high-entropy carbide ceramics. Acta Mater. 2020;195:739–749.
[13] Wen T, Ye B, Nguyen MC, et al. Thermophysical and mechanical properties of novel high-entropy metal nitride-carbides. J Am Ceram Soc. 2020;103(11):6475–6489.
[14] Dai FZ, Wen B, Sun Y, et al. Theoretical prediction on thermal and mechanical properties of high-entropy metal nitride-carbides (Zr0.3Hf0.3Ti0.3Ta0.3)C by deep learning potential. J Mater Sci Tech. 2020;43:168–174.
[15] Gurunathan R, Hansu R, Dylla M, et al. Analytical models of phonon–point-defect scattering. Phys Rev Appl. 2020;13:034011.
[16] Khafizov M, Pakarinen J, He L, et al. Impact of irradiation induced dislocation loops on thermal conductivity in ceramics. J Am Ceram Soc. 2019;102(12):7533–7542.
[17] Alfred LCR. Theory of the resistivity change in a metal due to multiple point imperfections. Phys Rev. 1966;152:693–698.
[18] Klemens PG. Thermal conductivity and lattice vibrational modes. London, UK: Academic Press; 1958. (Solid State Physics; 7). p. 1–98.
[19] Turk LA, Klemens PG. Phonon scattering by impurity platelet precipitates in diamond. Phys Rev B. 1974;9:4422–4428.
[20] Morelli DT, Perry TA, Farmer JW. Phonon scattering in lightly neutron-irradiated diamond. Phys Rev B. 1993;47:131–139.
[21] Ziegler JF, Ziegler MD, Biersack JP. SRIM: the stopping and range of ions in matter (2010). Nucl Instrum Meth Phys Res B. 2010;268(11–12):1818–1823.
[22] Weber WJ, Zhang Y. Predicting damage production in monoatomic and multi-elemental targets using stopping and range of ions in matter code: challenges and recommendations. Curr Opin Solid State Mater Sci. 2019;23(4):100757.
[23] Hurley DH, Schley RS, Khafizov M, et al. Local measurement of thermal conductivity and diffusivity. Rev Sci Instrum. 2015;86(12):123901.
[24] Khafizov M, Chauhan V, Wang Y, et al. Investigation of thermal transport in composites and ion beam irradiated materials for nuclear energy applications. J Mater Res. 2017;32(1):204–216.
[25] Maillet D, André S, Batsale JC, et al. Thermal quadrupoles: solving the heat equation through integral transforms. Chichester, UK: John Wiley & Sons, LTD; 2000.
[26] Hua Z, Fleming A, Ban H. The study of using a multi-layered model to extract thermal property profiles of ion-irradiated materials. Int J Heat Mass Transf. 2019;131:206–216.
[27] Riyad MF, Chauhan V, Khafizov M. Implementation of a multilayer model for measurement of thermal conductivity in ion beam irradiated samples using a modulated thermoreflectance approach. J Nucl Mater. 2018;509:134–144.
[28] Dennett CA, Hua Z, Khanolkar A, et al. The influence of lattice defects, recombination, and clustering on thermal transport in single crystal thorium dioxide. APL Mater. 2020;8:111103.
[29] Chen G, Hui P. Thermal conductivities of evaporated gold films on silicon and glass. Appl Phys Lett. 1999;74(20):2942–2944.
[30] Makinson Reb. The thermal conductivity of metals. Math Proc Cambridge Philos Soc. 1938;34(3):474–497.
[31] Zheng Q, Mei AB, Tuteja M, et al. Phonon and electron contributions to the thermal conductivity of VNx epitaxial layers. Phys Rev Mater. 2017;1:065002.
[32] Hossain MD, Borman T, Kumar A, et al. Carbon stoichiometry and mechanical properties of high entropy carbides. Acta Mater. 2021;215:117051.
[33] Wolf W, Podloucky R, Anttretter T, et al. First-principles study of elastic and thermal properties of refractory carbides and nitrides. Phil Mag B. 1999;79(6):839–858.
[34] Williams WS. Electrical properties of hard materials. Int J Refract Hard Met. 1999;17(1):21–26.
[35] Khafizov M, Yablinsky C, Allen TR, et al. Measurement of thermal conductivity in proton irradiated silicon. Nucl Instrum Meth Phys Res B. 2014;325:11–14.
[36] Ferry SE, Dennett CA, Woller KB, et al. Inferring radiation-induced microstructural evolution in single-crystal niobium through changes inthermal transport. J Nucl Mater. 2019;523:378–382.
[37] Toherer ES, Zevalkink A, Snyder GJ. Phonon engineering through crystal chemistry. J Mater Chem. 2011;21:15843–15852.
[38] Klemens PG. The scattering of low-frequency lattice waves by static imperfections. Proc Phys Soc A. 1955;68(12):1113–1128.