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The Electron Thermal Conductivity of Pu and Zr Substituted Gamma-Uranium

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Report Name:

The Electron Thermal Conductivity of Pu and Zr Substituted Gamma-Uranium

Report Category

Nuclear Energy

Scientific Achievement

Density Functional Theory calculations are applied to gamma-U, U-Pu, U-Zr, and U-Pu-Zr compositions, followed by application of the linearized Boltzmann transport equation under the relaxation time approximation [Madsen 2018] and a semi-empirical electron relaxation time calculation method [Zhou 2020] to calculate electron thermal conductivity for each composition (10 in total). The electrical and electron thermal conductivity are calculated using one of two methods based on literature [Zhou 2020, Zhou 2021]: the direct method, which assumes electron relaxation time is independent of alloying, and the mixed method, calculates conductivities by mixing alloying and host atom properties. Electron band structures are also calculated for gamma-U, U-Pu, and U-Zr compositions. The Sawtooth system of High Performance Computing at Idaho National Laboratory was used primarily, specifically the dual Xeon Platinum processors on the Sawtooth system. The Lemhi system was also used to a lesser degree, specifically the dual Xeon Gold processors.

Significance

Three important findings result from this work. First, the electron thermal conductivity decays more slowly with increase in atomic percent Pu than with increase in atomic percent Zr, both in the respective binary compounds with U, and in U-Pu-Zr. This is especially important for U-Pu-Zr, as it implies more Pu can be recycled per fuel element of U-Pu-Zr. Second, of the two methods applied, the mixed method performs better if Zr is present, and the direct method if Zr is not. This is attributed to the large mass difference, and to a lesser extent, electron band flattening, Zr induces. Therefore, we suggest that if alloying and host atoms differ significantly in mass and or valance electron number (specifically, if the alloying atom has less valance electrons), the mixed method should be applied, while if they do not, the direct method is appropriate. This finding is applicable to all metallic alloys, and can be applied for investigation of the radiation resistance of high-entropy alloys, which often consist of at least four species. Third, when Pu content is doubled, from 12.5 to 25.0 at. % (an increase of 100 %) the electron thermal conductivity is lowered by only 7.7 %. This again implies that greater Pu content in U-Pu-Zr is possible, allowing for faster recycling of spent fuel. Fourth, because the mass difference of Am and Cm (other recyclable long-lived actinides) relative to U is similar to that of Pu and these elements have more valance electrons than U, Am and Cm should behave similar to Pu. Therefore, not only can Pu be recycled faster increasing its content in U-Pu-Zr, but this is likely true for Am and Cm as well.

Key Publications

The Electron Thermal Conductivity of Pu and Zr Substituted Gamma-U (status: In Progress)

Sponsor/ Program

AMMT

Images

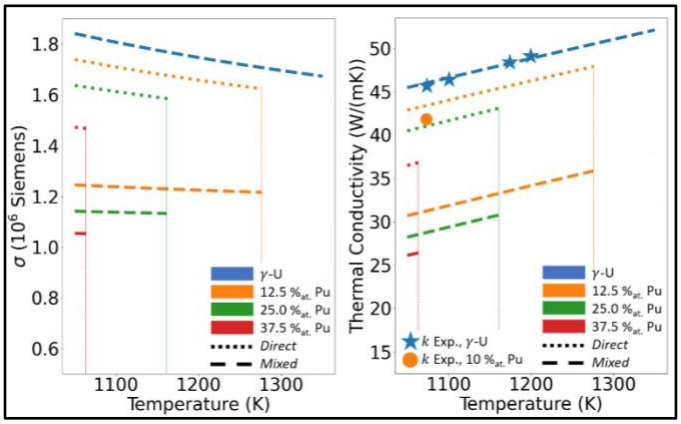

*Figure 1. Thermal and Electrical Conductivity of U-Pu Systems: **Left**: Calculated σ^{direct} and σ^{mixed} for U-Pu systems up to their solidus temperatures (denoted by vertical dashed lines)* [85]*, with σ^{direct} and σ^{mixed} for γ-U for comparison. **Right**: Calculated k_e^{direct} and k_e^{mixed} are compared against experimental k* [16] *for U-Pu systems up to their solidus temperatures (denoted by vertical dashed lines)* [85]*. Calculated k_e^{direct}, k_e^{mixed}, and experimental* [36] *k for γ-U is included for comparison.*

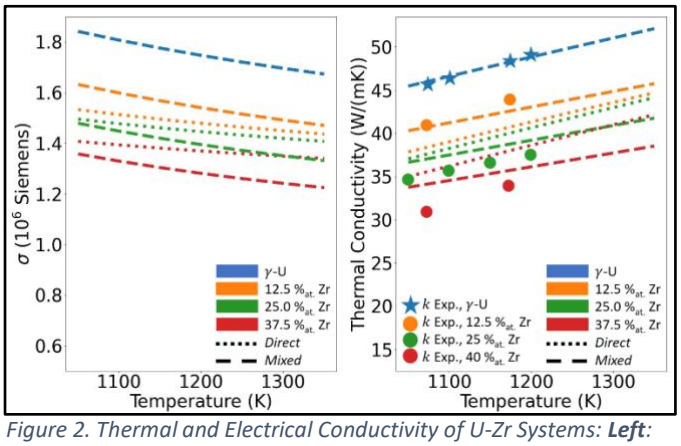

*Figure 2. Thermal and Electrical Conductivity of U-Zr Systems: **Left**: Calculated σ^{direct} and σ^{mixed} for U-Zr systems, with σ^{direct} and σ^{mixed} for γ-U for comparison. **Right**: Calculated k_e^{direct} and k_e^{mixed} are compared against experimental k* [37], [42] *for U-Zr systems. Calculated k_e^{direct}, k_e^{mixed}, and experimental k* [36] *for γ-U is included for comparison.*

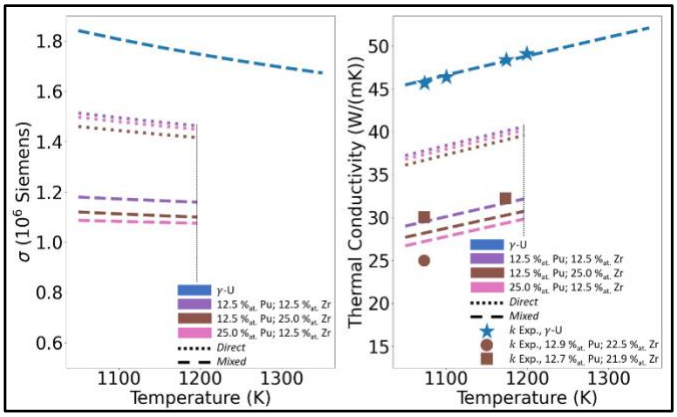

Figure 3. Thermal and Electrical Conductivity of U-Pu-Zr Systems: ***Left****: Calculated* σ^{direct} *and* σ^{mixed} *for U-Pu-Zr systems up to the highest experimentally available solidus temperature for U-Pu-Zr (denoted by a vertical dashed line)* [17]*, with* σ^{direct} *and* σ^{mixed} *for* γ*-U for comparison.* ***Right****: Calculated* k_e^{direct} *and* k_e^{mixed} *are compared against experimental* k [15], [22] *for U-Pu-Zr systems up to the highest experimentally available solidus temperature for U-Pu-Zr (denoted by vertical dashed lines)* [17]*. Calculated* k_e^{direct}*,* k_e^{direct}*, and experimental* [36] k *for* γ*-U is included for comparison.*