Thermoelectric Properties of Gadolinium Copper Tellurides

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Abstract:

Since thermoelectrics are an important way to produce electricity from waste heat, the thermoelectric properties of gadolinium copper telluride, $GdCuTe_2$, together with the doped compounds gadolinium samarium copper telluride, $Gd_{1-x}Sm_xCuTe_2$, and gadolinium dysprosium copper telluride, $Gd_{1-x}Dy_xCuTe_2$, with x = 0.02 and x = 0.05 were investigated. Upon doping, a decrease in thermal conductivity and an increase in electrical conductivity were observed leading to an improved figure of merit for the heavily doped samples. An interesting peak around 200°C in the electrical conductivity was also found, stimulating further research into the cause of this nonlinear trend, which will hopefully be done in the future.

Introduction:

With waste heat as a common problem, thermoelectric materials have gained interest for the generation of electricity. While they have high potential, the current low efficiency of the materials is still a limiting factor in commercializing these systems. The generation of electricity from a temperature gradient applied to a material is known as the Seebeck effect, with the Seebeck coefficient, α, defined as the electrical potential difference, ΔV, divided by the temperature difference, ΔT. The efficiency of a thermoelectric material can be probed through the calculation of the thermoelectric figure of merit (*ZT*), defined as

$$ZT = \frac{\sigma \alpha^2 T}{\lambda}$$ *Equation 1.*

with σ as the electrical conductivity, T as the temperature and λ as the thermal conductivity [1]. This project focused on the thermoelectric characterization of gadolinium copper tellurides.

Gadolinium copper telluride, $GdCuTe_2$, was chosen for its small band gap semiconductor nature and for its potential magnetic properties. In addition, other rare earth copper tellurides of the same crystal structure have been found to have good ZT values [2]. The Gd site was doped with either samarium or dysprosium to form $Gd_{1-x}Sm_xCuTe_2$ or $Gd_{1-x}Dy_xCuTe_2$ respectively, with x = 0.02 and x = 0.05. The structural disorder induced by the dopants, together with their magnetic moments and the lower valence state of Sm compared to Gd were expected to have a beneficial effect on the thermoelectric properties of the compound.

Methods:

The different material systems were synthesized by combining stoichiometric ratios of the materials in a glove box and sealing them in quartz tubes under vacuum. The tubes were placed in a furnace to directly react the materials. They were heated to 1050°C in ten hours and kept there for six hours, then cooled to 750°C in three hours and kept there for 24 hours. The samples were then reground in a glove box and the heating steps were repeated. Spark Plasma Sintering was performed on each sample to densify it. X-ray diffraction (XRD) was performed to determine the structure and look for secondary phases. The heat capacity was measured using differential scanning calorimetry. The thermal diffusivity, the Seebeck coefficient and the electrical conductivity were measured from 50°C to 450°C using a Laser Flash Apparatus (LFA) for the former and ZEM-2 for the two latter.

Results and Conclusions:

The crystal structure of $GdCuTe_2$ was verified to be monoclinic with a C2/m space group using XRD [3], although some peaks could be ascribed to secondary phases. The heat capacity was measured and a phase transition was observed around 500°C, therefore all other measurements were stopped at 450°C to avoid this transition. The electrical conductivity measured using ZEM was found to be dependent on doping, with an interesting peak around 200°C as seen in

Figure 1. This peak could be due to a structural transition or a secondary phase. It is also worth mentioning that the 2% and the 5% doped compounds behave with similar patterns, independently of the specific dopant. The Seebeck coefficient, Figure 2, also showed correlation to the electrical conductivity measurement with a transition at 200°C. While the Seebeck coefficient was impacted by doping, no systematic change could be observed. The thermal conductivity was found to be rather low characteristic of the layered nature of the compound. It was observed to further decreasing upon doping, as shown in Figure 3. Given the anisotropic crystal structure of the material it is important to note that the ZT calculated with the thermal properties measured perpendicular to the pressing direction and the electrical properties parallel to it may be overestimated. As seen in Figure 4, the figure of merit was increased to 0.22 at 200°C with heavy doping and the peak around 200°C is still present in both of the 5% compounds.

Future Work:

More investigation will need to be done to determine what the nature of the transition around 200°C. This may lead to a better understanding of how to manipulate the thermoelectric properties of a material. It would also be important to determine the actual composition of the material and determine what effects secondary phases may be playing.

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References:

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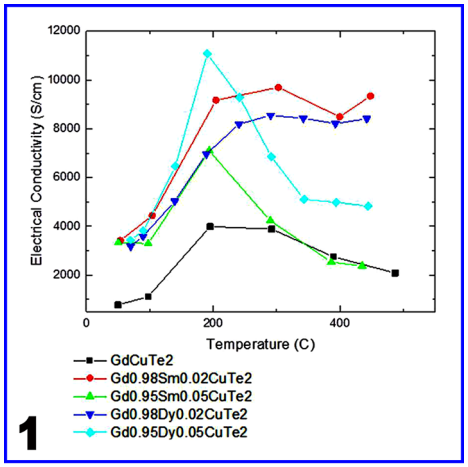

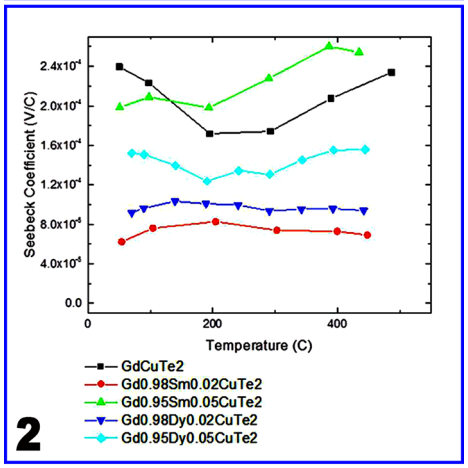

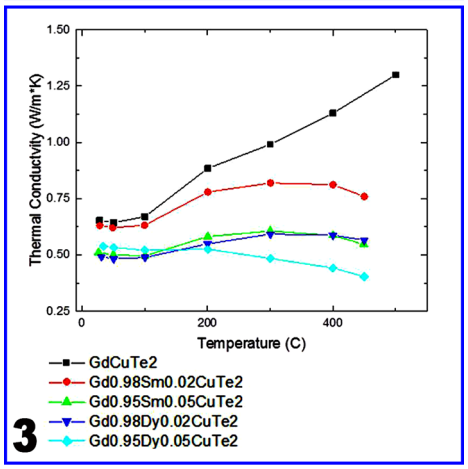

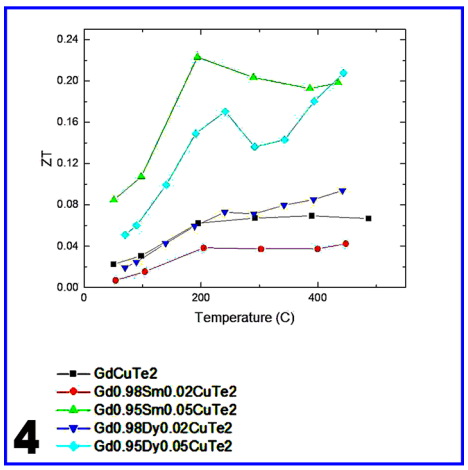

Figure 1: The electrical conductivity of the $GdCuTe_2$. It was found that the electrical conductivity was dependent on the amount of doping done on the gadolinium site.

Figure 2: The Seebeck coefficient measurements of $GdCuTe_2$.

Figure 3: The thermal conductivity of the $GdCuTe_2$ found with LFA. It was found that the thermal conductivity was decreased with doping the gadolinium site.

Figure 4: The calculated figure of merit for the gadolinium copper tellurides. It is observed that the 5% doped samples have an increase in the figure of merit.