Department of Materials Science
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Utilizing novel enhancement principles to develop high performance thermoelectric materials & devices

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Development of thermoelectric (TE) materials is important, as in addition to energy saving via waste heat power generation [1], they can also serve as dynamic power sources for innumerable IoT sensors and devices [2]. To achieve enhanced thermoelectric performance, it is necessary to find ways to overcome the traditional tradeoffs between the key properties, namely, between the Seebeck coefficient S and electrical conductivity σ, and between the electrical and thermal conducivity κ. Find ways to enhance S, and also selectively lower κ [3]. I would like to systematically present several principles we have been developing, most which can be widely applied to different materials [4 and references therein].

For the first tradeoff overcoming, we have found that magnetism can be utilized to to enhance the Seebeck coefficent and overall power factor. Strong coupling of the electrical carriers with magnetic moments, can lead to effective magnon drag, e.g. like for $CuFeS_2$ chalcopyrite and the recently indicated origin of the huge power factor in metastable Fe_2VAl-based thin films, and furthermore paramagnon drag, where magnetic ion doping into nonmagnetic materials could enhance S. Spin fluctuation and spin entropy has also been demonstrated to enhance the Seebeck coefficient.

For the second aspect, in addition to various nanostructurings, intrinsic low κ mechanisms have been demonstrated. Materials informatics approach led to identification of a material catalogue with low κ. Particular doping into SnTe was shown to lead to softening of the lattice and a dramatic reduction of thermal conductivity largely exceeding the contribution from phonon scattering. Finally, the heterogeneous bonding in mixed anion compounds was shown to result in exceptional low thermal conductivity.

Defect engineering has also been shown to be a powerful method. Cr doping in GeTe serendipitously lowered the formation energy of Ge defects leading to homogenously distributed Ge precipitations and vacancies, coupled with typical band convergence doping to obtain $ZT\sim2$. A high entropy approach of $AgInTe_2$ alloying into GeTe, stabilized the cubic phase, thereby enabling enhanced doping of Bi, leading to the first stable n-type conduction in GeTe. The hidden role of rhombohedral distortion degree on the Ge-vacancy formation energy was revealed and utilized leading to high power factor and ZT_{av}. Incidentally, a combined theoretical and experimental screening of some unusual dopants of GeTe revealed Zr to be an effective dopant.

Recently an interesting dual effect of small amounts of Cu doping in Mg_3Sb_2 was revealed. Interstitial Cu doping was indicated to lower the phonon group velocity, while Cu doping into the grain boundaries promoted grain growth and unusual optimum chemical composition leading to very high mobilities similar to single crystals, while being a polycrystalline material with low thermal conductivity. An initial realistic 8 pair module composed of Cu doped Mg_3Sb_2-type and MgAgSb exhibited an efficiency of 7.3% @hot temperature side of 320ºC, with an estimated efficiency from the actual performance of materials actually reaching close to 11% [5]. Tuning toward room temperature yielded an initial realistic 8 pair module with an efficiency of 2.8% with temperature difference of 95 K from RT and cooling of 56.5 K [6]. Once again, the performance of materials are much higher, so further improvements are expected as the module technology becomes developed.

I will also present an overview on different thermoelectric power generation devices which can be utilized for energy harvesting for possible applications [4 and references therein]. We have also recently fabricated a miniaturized in-plane π-type thermoelectric device utilizing the microfabrication techniques of photolithography and dry etching which are industrial compatible [7].

[1] L. E. Bell, Science 321, 1457 (2008), JOM, 68, 2673-2679 (2016); [2] T. Mori, S. Priya, MRS Bulletin 43, 176 (2018), I. Petsagkourakis et al, Sci. Tech. Adv. Mater. 19 (2018) 836, N. Nandihalli et al., Nano Energy, 78 105186 (2020); [3] T. Mori, Small 13, 1702013 (2017); [4] T. Hendricks, T. Caillat, and T. Mori, Energies, 15, 7307 (2022); [5] Z. Liu et al., Joule, 5, 1196-1208 (2021); [6] Z. Liu et al., Nature Commun. 13, 1120 (2022); [7] I. Ohkubo et al., Mater. Today Energy, 28, 101075 (2022).

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