A Recommendation Engine for Suggesting Unexpected Thermoelectric Chemistries: Characterization of $RE_{12}Co_5Bi$ (RE=Gd,Er)

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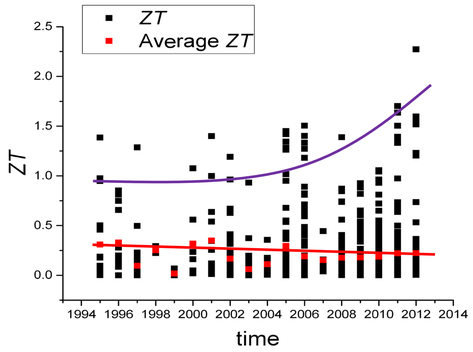
Most thermoelectric research is incremental

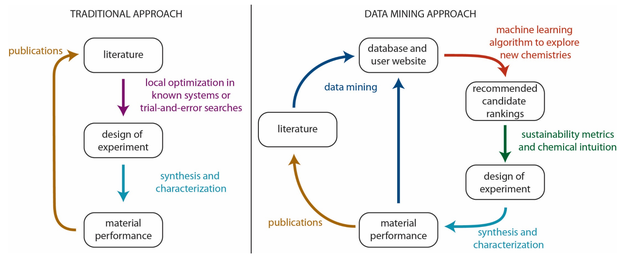
Maximum *zT* has been steadily increasing with time but these tend to be outliers. The average *zT* has been slowly declining. Exploring new compounds is high risk and low probability of success.



http://www.mrl.ucsb.edu:8080/datamine/thermoelectric.jsp
Gaultois et al. "Data -driven review of thermoelectric materials: Performance and resource considerations," *Chemistry of Materials*, **25**, [15] 2911-2920 (2013).

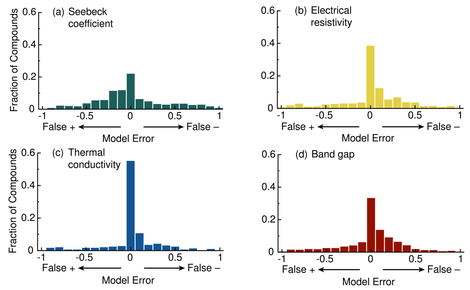
New tools of discovery are needed to explore chemical white space

Traditional approaches for materials research in thermoelectrics and other technologies tend to rely on local optimization of known systems or chemical trial and error searches guided by limited chemical intuition. Data mining could provide the bases for a new machine learning based tool that generates materials recommendations with high probability of good performance.



A recommendation engine based on a large dataset has been created and validated

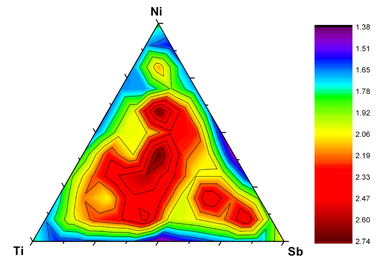
Leave-one-out cross-validation shows that the model has very few false positives or false negatives. This recommendation engine predicts probability of performance within a given range, not an actual calculated value.



Oliynyk, Anton O., et al. "Gd12Co5. 3Bi and Gd12Co5Bi, Crystalline Doppelgänger with Low Thermal Conductivities." Inorganic chemistry (2016). APA

Performance probability phase diagrams can be constructed quickly and easily

Prediction only requires composition, not crystal structure. This means that thousands of compounds can be screened in minutes. The four properties relevant to thermoelectrics (Seebeck Coefficient, electrical resistivity, thermal conductivity, and band gap) can be calculated for different phase space and plotted on a ternary diagram.



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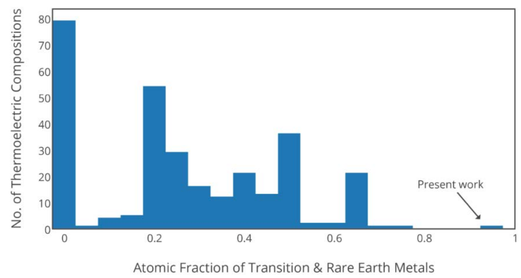
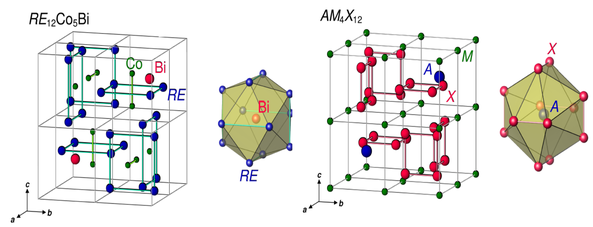
$RE_{12}Co_5Bi$ offers experimental validation in a new class of materials

The recommendation engine is a tool to explore chemical white space so we chose to validate this concept on a compound that is extremely different from standard thermoelectric materials. Model suggested high confidence in $\kappa < 10\ W/mK$ and $\rho < 10^{-2}\ \Omega cm$ but low confidence in $S > 100\ \mu V/K$ and a band gap.



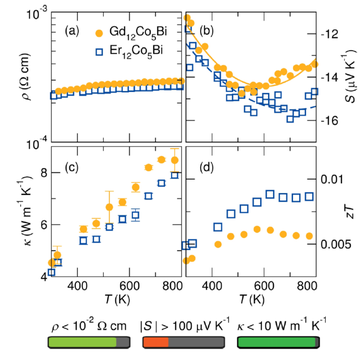
$Re_{12}Co_5Bi$ is nearly antitype to skutterudite structure

Structural similarity to skutterudite might explain the observed low thermal conductivity, but cannot explain the low predicted thermal conductivity because the algorithm uses composition alone (not crystal structure).



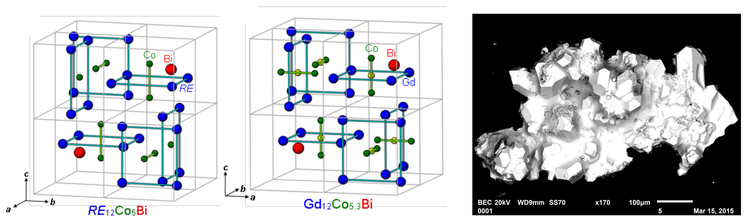
Measured properties agree well with predictions

Interestingly, thermal conductivity rises with increasing temperature. Synchrotron diffraction revealed subtle secondary phases hidden within the primary phase.



Unusual thermal transport led to subsequent detailed investigation of structure-property relationships

Single crystal phase space exploration led to the discovery of a new crystal structure which was very structurally and chemically similar to our expected composition.



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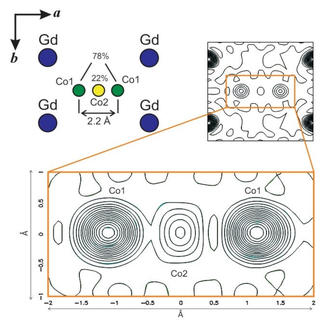
$RE_{12}Co_{5.3}Bi$ has additional Co disorder compared to $RE_{12}Co_5Bi$

Electron density map shows two distinct sites, not one elongated site. This leads to the different electrical resistivity with temperature but the thermal conductivity was dramatically different.



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http://thermoelectrics.citrination.com

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