

## High Throughput Generation of High- $zT$ Thermoelectrics with Precise Stoichiometric Controls

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Formula	zT
Pb0.933Cu0.067Te	1.55
Ge0.965Zn0.035Te	2.11
Bi1.942Te0.058Te3	1.96
Ge0.943Cu0.057Te	1.2
Sn0.931K0.069Se	1.72
Bi1.973Br0.027Te3	1.31
Mg2.978Cl0.022Sb2	1.92
Bi1.958Ba0.042Te3	1.93
Ge0.99Cu0.01Te	1.79
Bi1.948Br0.052Te3	1.32
Pb0.92Mg0.06Na0.02Te0.85Se0.15	1.89
Te0.041Co4Sb1	1.28
Ge0.986Zn0.014Te	1.27
Bi1.975La0.025Te3	1.59
Ge0.981Li0.019Te	1.29
Pb0.979K0.021Te	1.21
Br0.081Co4Sb1	1.25
Bi1.976Te0.024Te3	1.83
Bi1.982Ba0.018Te3	1.58
Mg2.98Ba0.02Sb2	1.38
Ge0.93Li0.07Te	1.48
Bi1.98La0.02Te3	1.99
Pb0.953Li0.047Te	1.45
Bi1.931Cl0.069Te3	1.45
Ge0.948K0.052Te	1.32
Bi1.965Cl0.035Te3	1.02
Mg2.917Ba0.083Sb2	1.97
Ge0.966Li0.034Te	1.27
Br0.079Co4Sb1	1.02
Pb0.953K0.047Te	1.85
Mg2.921I0.079Sb2	1.75
Pb0.984Ag0.016Te	1.39
Pb0.971Zn0.029Te	1.84
Pb0.916Li0.084Te	1.91
Mg2.986La0.014Sb2	1.29
Bi1.955Cl0.045Te3	1.4
Br0.037Co4Sb1	1.97
Ge0.943Ag0.057Te	1.43
Mg2.926Cl0.074Sb2	1.01
Ge0.916Zn0.084Te	1.04
Pb0.965Li0.035Te	1.74

Pb0.914Cu0.086Te	1.52
Ge0.975Ag0.025Te	1.54
Pb0.955Li0.045Te	1.01
Mg2.928La0.072Sb2	1.01
Ge0.969Zn0.031Te	1.5
Bi1.995I0.005Te3	1.72
Sn0.916Zn0.084Se	1.73
Bi1.994Yb0.006Te3	1.96
Ge0.941Zn0.059Te	1.83
Ba0.013Co4Sb1	1.06
Ge0.937Cu0.063Te	1.96
Sn0.989Li0.011Se	2.21
Pb0.924Ag0.076Te	1.3
Bi1.927Yb0.073Te3	1.28
Ge0.966K0.034Te	1.75
Pb0.958Ag0.042Te	1.15
Yb0.078Co4Sb1	1.28
Ge0.977Cu0.023Te	1.59
La0.021Co4Sb1	1.56
Bi1.943Ba0.057Te3	1.18
Mg3.05Zn0.15Sb1.48Bi0.52	1.62
Mg2.964Ba0.036Sb2	1.73
Pb0.914Ag0.086Te	1.51
Mg2.983I0.017Sb2	1.15
Ge0.957Ag0.043Te	1.36
Bi1.936Ba0.064Te3	1.78
Ba0.069Co4Sb	1.18
Pb0.934Na0.066Te	1.25
Bi1.977Cl0.023Te3	1.4
Pb0.962Na0.038Te	1.22
Pb0.983Zn0.017Te	1.05
Mg2.989Yb0.011Sb2	2.15
Bi1.917Br0.083Te3	1.98
Br0.029Co4Sb1	1.51
Mg2.941Yb0.059Sb2	1.35
Te0.052Co4Sb1	2.3
La0.076Co4Sb1	1.61
Bi1.955La0.045Te3	2.1
Mg2.986Cl0.014Sb2	2.2
Bi1.965Ba0.035Te3	1.46
Sn0.978Ag0.022Se	1.3
Ge0.921Cu0.079Te	1.71
Ge0.971Cu0.029Te	1.23

Sn0.954Li0.046Se	1.69
Pb0.943Na0.057Te	2.17
Bi1.928Yb0.072Te3	1.44
Ge0.92Ag0.08Te	1.07
Pb0.945K0.055Te	1.99
Pb0.937K0.063Te	1.16
Ge0.928Na0.072Te	1.24
Sn0.979Na0.021Se	1.15
Bi1.945I0.055Te3	1.29
Mg2.932I0.068Sb2	2.17
Mg2.96Br0.04Sb2	1.17
Te0.01Co4Sb1	1.98
Li0.03K0.26Cu1.71Se	2.21
Ge0.963K0.037Te	2.1
Pb0.975Cu0.025Te	1.6
Pb0.933Cu0.067Te	1.55
Ge0.965Zn0.035Te	2.11
Bi1.942Te0.058Te3	1.96
Ge0.943Cu0.057Te	1.2

Table S1. The other 99 generated potential high-performance thermoelectric materials

Stoichiometric Formula	Calculation Settings	DFT-calculated Formation Energy (eV/atom)	Roost-predicted Formation Energy (eV/atom)	Maximum Predicted zT (Temperature)
$\text{Pb}_{0.92}\text{Mg}_{0.06}\text{Na}_{0.02}\text{Te}_{0.85}\text{Se}_{0.15}$	VASP, PAW pseudopotential, HSE06 functional, 500 eV cutoff, 12×12×12 k-point mesh	-0.39	-0.35	1.89(700K)
$\text{Mg}_{3.05}\text{Zn}_{0.15}\text{Sb}_{1.48}\text{Bi}_{0.52}$	VASP, PAW pseudopotential, HSE06 functional, 500 eV cutoff, 12×12×12 k-point mesh	-0.47	-0.40	1.62(600K)
$\text{Li}_{0.03}\text{K}_{0.26}\text{Cu}_{1.71}\text{Se}$	VASP, PAW pseudopotential, HSE06 functional, 500 eV cutoff, 12×12×12 k-point mesh	-0.33	-0.42	2.21(1000K)

Table S2. DFT validation results of additional thermoelectric candidates