

Supporting Information

Inertial Effective Mass as an Effective Descriptor for Thermoelectrics via Data-driven Evaluation

Ady Suwardi[§], Daniil Bash^{§,†}, Hong Kuan Ng^{§,‡}, Jose Recatala Gomez^{§,⊥}, D V Maheswar
Repaka[§], Pawan Kumar[§] and Kedar Hippalgaonkar^{§#}

[§]Institute of Materials Research and Engineering, #08-03, 2 Fusionopolis Way, Agency for
Science, Technology and Research, Singapore 138634

[†]Department of Chemistry, National University of Singapore, Singapore 117543, Singapore

[‡]NUS Graduate School for Integrative Sciences and Engineering, National University of
Singapore, Center for Life Sciences, #05-01, 28 Medical Drive, Singapore 117456

[⊥]Department of Chemistry, University of Southampton, University Road, Highfield,
Southampton SO17 1BJ, United Kingdom

[#]Corresponding author: kedarh@imre.a-star.edu.sg

Seebeck vs inertial effective mass

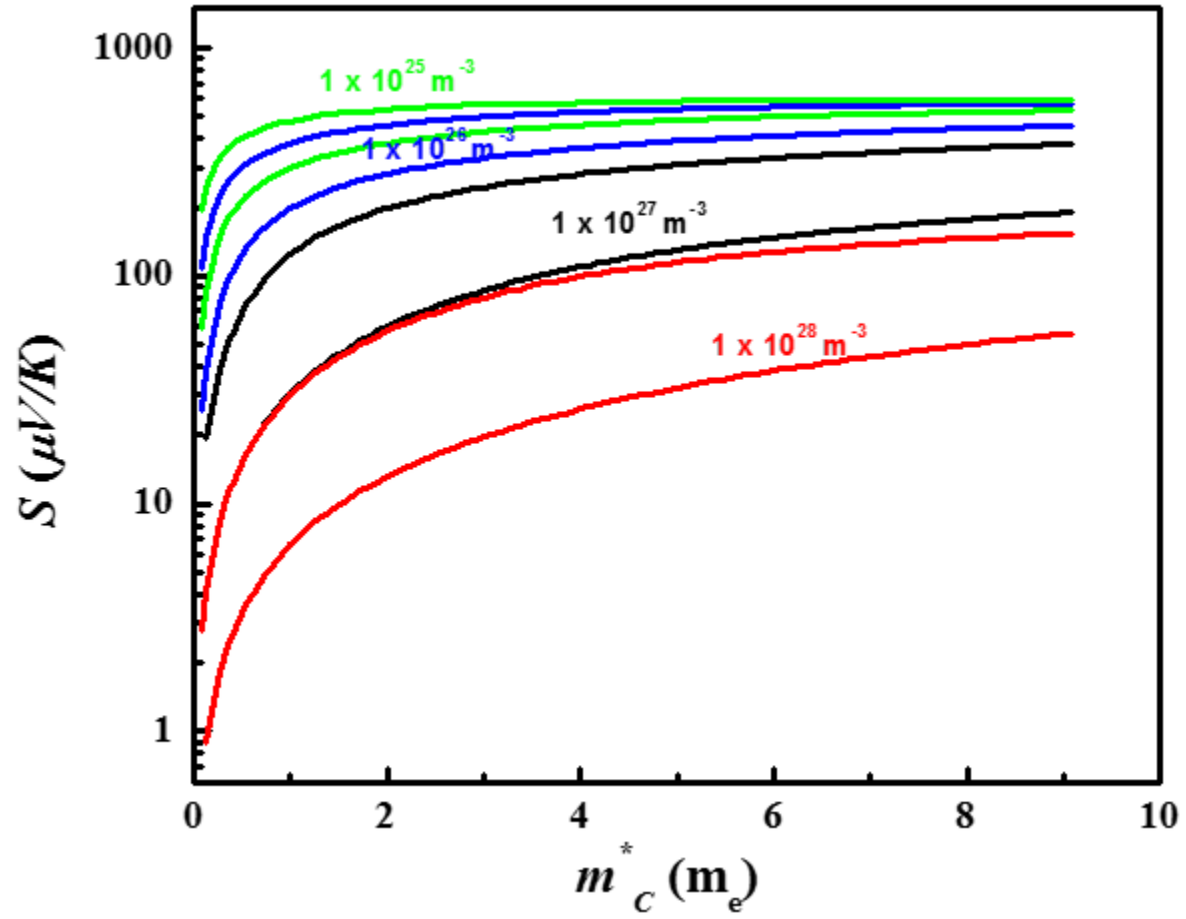


Figure S1. Seebeck vs inertial effective mass of p-type compounds at 300 K for carrier concentration ranging from $1 \times 10^{25} \text{ m}^{-3}$ to $1 \times 10^{28} \text{ m}^{-3}$. Each carrier concentration is color coded differently. Solid lines represents lower and upper bound of $N_V^* K^*$ of 1 and 10, respectively.

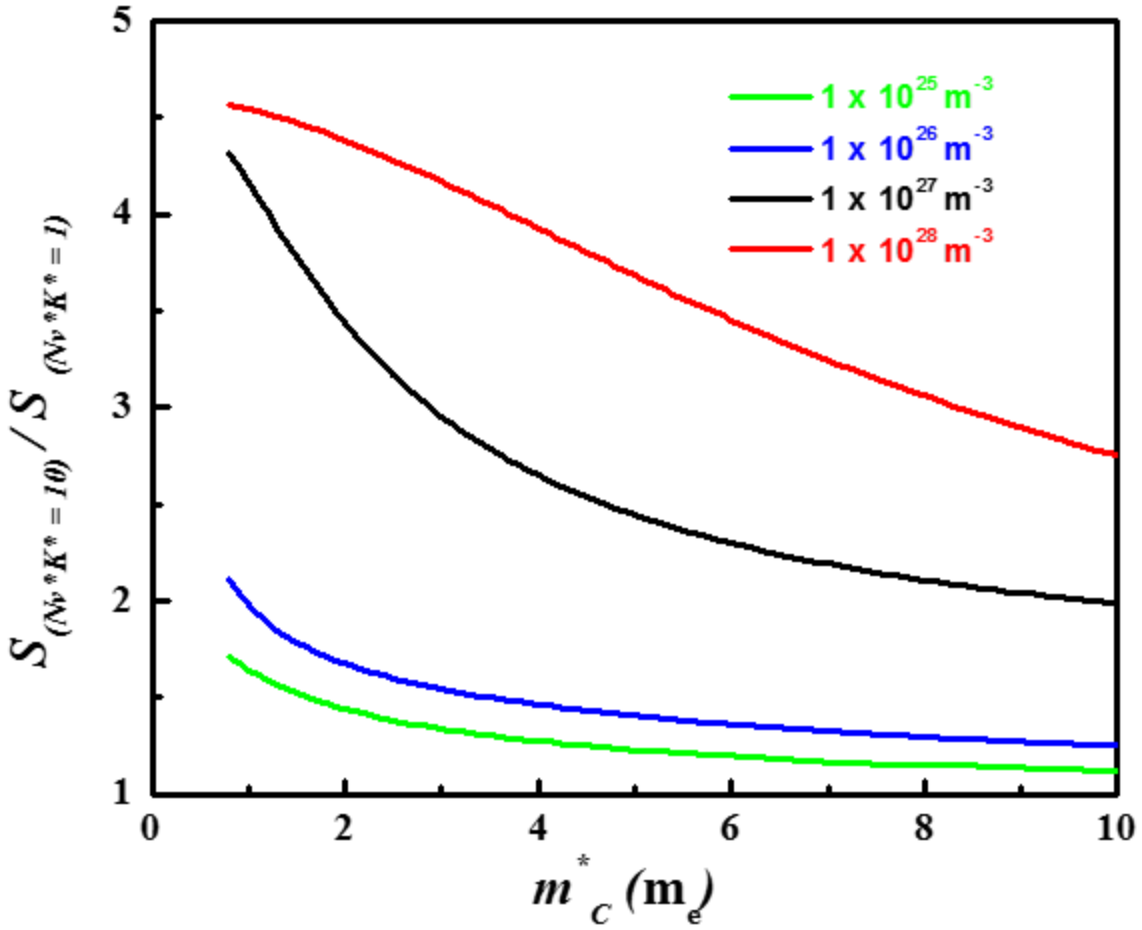


Figure S2. Ratio of Seebeck coefficients at $N_V^* K^*$ of 10 to $N_V^* K^*$ of 1, showing higher enhancements at lower inertial effective mass across a wide range of carrier concentration. High carrier concentration has higher enhancement at any given value of inertial effective mass over the low carrier concentration counterparts. of p-type compounds at 300 K for carrier concentration ranging from $1 \times 10^{25} \text{ m}^{-3}$ to $1 \times 10^{28} \text{ m}^{-3}$. Each carrier concentration is color coded differently, in the same order according to S1.

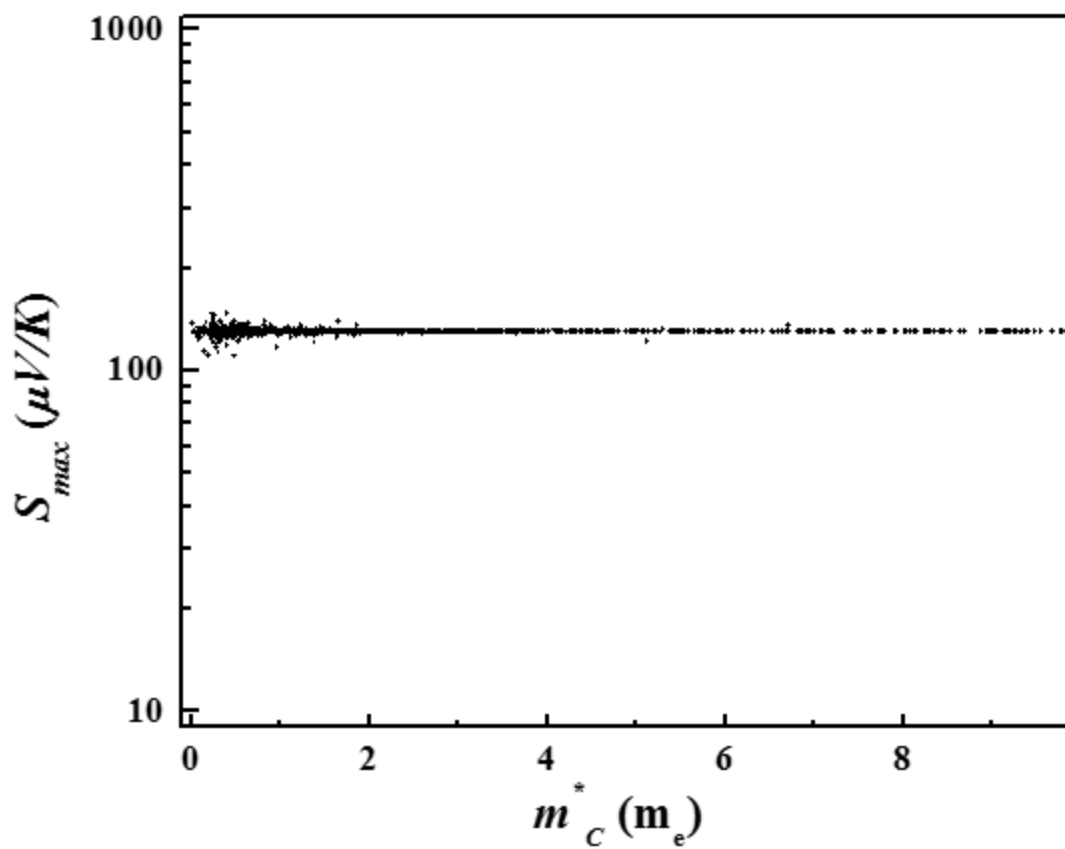


Figure S3. Seebeck coefficients at optimum carrier concentration for all compounds, plotted against their inertial effective mass. The optimal Seebeck values of 130 $\mu\text{V/K}$ is consistent with the case for constant relaxation time assumption (*i.e.* $r = 0$).¹

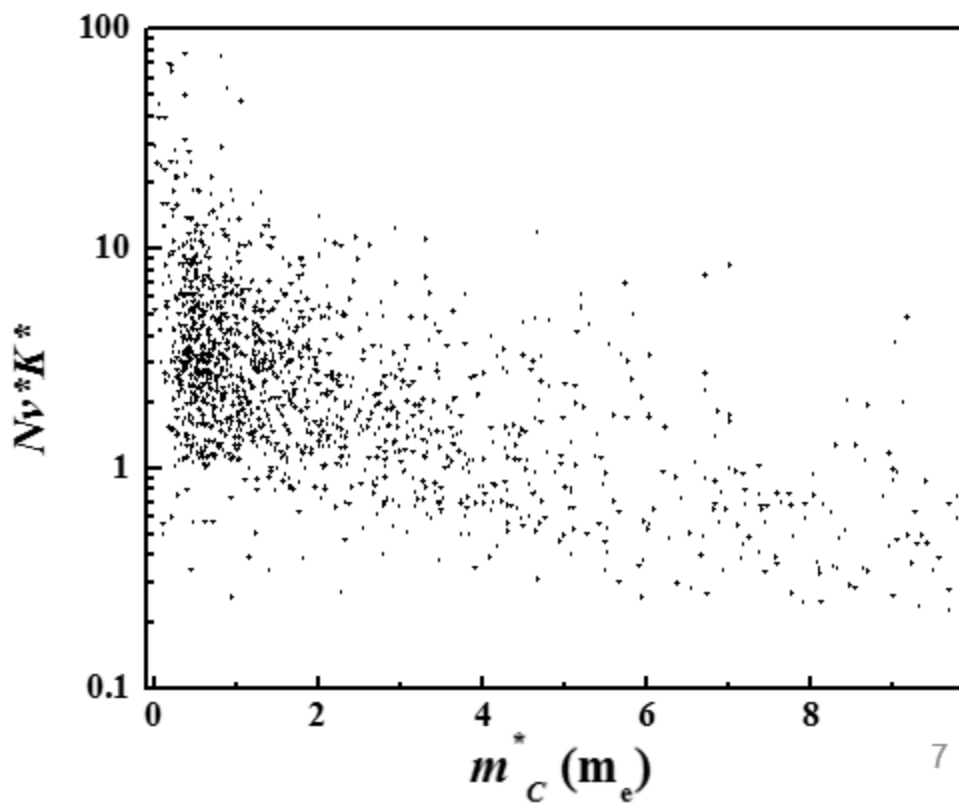


Figure S4. $N_V^*K^*$ plotted against inertial effective mass. Qualitatively, at a particular carrier concentration, low effective mass compounds will possess higher reduced fermi potential (*i.e.* the Fermi level is deeper into the conduction/valence bands). Hence, utilizing more orbitals (higher orbital degeneracy), resulting in higher $N_V^*K^*$.

Calculation for effective Seebeck and electrical conductivity

The effective Seebeck coefficient and average conductivity for polycrystalline compounds²:

$$S = \frac{S_{xx}\sigma_{xx} + S_{yy}\sigma_{yy} + S_{zz}\sigma_{zz}}{3\sigma_{avg}} \quad (1)$$

$$\sigma_{avg} = \frac{\sigma_{xx} + \sigma_{yy} + \sigma_{zz}}{3} \quad (2)$$

Calculation for optimal power factor

In order to minimize computational cost, the optimal power factor is calculated by assuming constant Seebeck effective mass (m_s^*) (*i.e.* taken at $n = 1 \times 10^{20} \text{ cm}^{-3}$) since the optimal doping concentration for many thermoelectric compounds lie near to this doping concentration. Using the effective Seebeck coefficients obtained from Equation (1) for each compound, the reduced fermi potential (η) can be calculated using the Boltzmann transport equation expression for Seebeck coefficient in Equation (3)¹ and by solving the Fermi-Dirac integrals code in MATLAB.

$$S = \pm \frac{k_B}{e} \left[\eta - \frac{\frac{5}{2} F_{(3/2)}(\eta)}{\frac{3}{2} F_{(1/2)}(\eta)} \right] \quad (3)$$

From Equation (3), for every value of Seebeck coefficient, a one-to-one correspondence can be found on η . Subsequently, the n (carrier concentration) corresponding to each η can be calculated from the following equation:³

$$n = \frac{2}{\sqrt{\pi}} \int_0^\infty N_{C,3D} \sqrt{\varepsilon} f_{FD}(\varepsilon) d\varepsilon \quad (4)$$

which essentially reduces to half-order Fermi integral that can be solved numerically:

$$n = N_{C,3D} F_{\frac{1}{2}}(\eta) \quad (5)$$

Here, $N_{C,3D}$ represents the 3 dimensional effective density of states and can be expressed in terms of m_s^* as follows:

$$N_{C,3D} = 12 (2\pi m_s^* k_B T / h^2)^{\frac{3}{2}} \quad (6)$$

Hence, by taking the value of m_s^* for each compound and calculating the corresponding η from Seebeck coefficient, n can be obtained. Finally, the optimum carrier concentration and maximum power factor $S^2\sigma$ can be determined by varying the value of n to get the corresponding S and σ that gives the highest power factor, as shown in Table T2.

Calculation for $N_V^*K^*/n$ slope

The slope for $N_V^*K^*/n$ was calculated from the range of $n = 1 \times 10^{19} \text{ cm}^{-3}$ to $1 \times 10^{20} \text{ cm}^{-3}$. Using the effective Seebeck coefficient, carrier concentration, as well as temperature, the Seebeck effective mass m_s^* was calculated using Boltzmann transport equations using MATLAB. The Fermi-Dirac integrals was solved using MATLAB code as described in ⁴. $N_V^*K^*$ is subsequently estimated by the following equation:

$$m_s^* = (N_V^*K^*)^{2/3} m_c^* \quad (7)$$

Where: $m_c^* = 3 \left(\frac{1}{m_x^*} + \frac{1}{m_y^*} + \frac{1}{m_z^*} \right)^{-1}$

Subsequently, the slope of $N_V^*K^*/n$ was calculated using linear approximation of the following:

$$\frac{\partial N_V^* K^*}{\partial n} = \frac{N_V^* K^* (1 \times 10^{20} \text{ cm}^{-3}) - N_V^* K^* (1 \times 10^{19} \text{ cm}^{-3})}{n} \quad (8)$$

Averaging methods for m_b^* and m_c^*

Different averaging methods were used for m_b^* and m_c^* .⁵ The single valley effective mass, m_b^* can be calculated from geometric average:

$$m_b^* = (m_x^* m_y^* m_z^*)^{1/3} \quad (9)$$

The inertial effective mass, m_c^* , which determines the carrier mobility can be calculated from the harmonic average along each direction:

$$m_c^* = 3 \left(\frac{1}{m_x^*} + \frac{1}{m_y^*} + \frac{1}{m_z^*} \right)^{-1} \quad (10)$$

The effective anisotropy parameter K^* for non-ellipsoidally shaped Fermi surface can be defined as:

$$K^* = \left(\frac{m_b^*}{m_c^*} \right)^{\frac{3}{2}} \quad (11)$$

References

1. P. Pichanusakorn and P. Bandaru, *Materials Science and Engineering: R: Reports*, 2010, **67**, 19-63.
2. D. S. Parker, A. F. May and D. J. Singh, *Physical Review Applied*, 2015, **3**, 064003.
3. H. Ng, D. Chi and K. Hippalgaonkar, *Journal of Applied Physics*, 2017, **121**, 204303.
4. N. Mohankumar and A. Natarajan, *physica status solidi (b)*, 1995, **188**, 635-644.
5. M. Cagnoni, D. Führen and M. Wuttig, *Advanced Materials*, 2018, **30**, 1801787.