Supplement Information

A list of constants used in the formula

$$a_1 = \frac{J^2 fL}{2\sigma_1 \kappa_1} + \frac{J^2(1-f)L}{2\sigma_1\sigma_2}\frac{\sigma_1(1-f)+\sigma_2 f}{\Lambda} - \frac{J(1-f)L(\varepsilon_2-\varepsilon_1)T_h - \kappa_2\Delta T}{L\Lambda},$$

$$b_1 = T_h,$$

$$a_2 = \frac{J^2(1+f)L}{2\sigma_2\kappa_2} - \frac{J^2 fL}{2\sigma_1\sigma_2}\frac{\sigma_1(1-f)+\sigma_2 f}{\Lambda} + \frac{JfL(\varepsilon_2-\varepsilon_1)T_c + \kappa_1\Delta T}{L\Lambda},$$

$$b_2 = -\frac{J^2 fL^2}{2\sigma_2\kappa_2} + \frac{J^2 fL^2}{2\sigma_1\sigma_2}\frac{\sigma_1(1-f)+\sigma_2 f}{\Lambda} + \frac{T_c[\kappa_1(1-f)+\kappa_2 f - Jf^2 L(\varepsilon_2-\varepsilon_1)] - \kappa_1\Delta T}{\Lambda},$$

$$c_1 = \phi_h,$$

$$c_2 = \phi_c - \frac{\varepsilon_2 J^2 L^2}{2\sigma_2\kappa_2} + (\frac{J}{\sigma_2} + \varepsilon_2 a_2)L,$$

$$\Lambda = \kappa_1(1-f) + \kappa_2 f + J(\varepsilon_2-\varepsilon_1)(1-f)fL,$$

$$\Omega = \frac{1}{2}(\varepsilon_2-\varepsilon_1)f(1-f)[\sigma_1(1-f)+\sigma_2 f],$$

$$\Phi = [\sigma_1(1-f)+\sigma_2 f][\kappa_1(1-f)+\kappa_2 f],$$

$$\Gamma = (\varepsilon_2-\varepsilon_1)f(1-f)\sigma_1\sigma_2,$$

$$\Psi = [\kappa_1\varepsilon_2(1-f)+\kappa_2\varepsilon_1 f]\sigma_1\sigma_2,$$

$$\Theta = [\kappa_1(1-f)+\kappa_2 f]\sigma_1\sigma_2.$$

Material constants used for numerical evaluations

TABLE I: Thermoelectric properties of Bi_2Te_3 and the second phase that is optimally matched with Bi_2Te_3 under different temperatures.

Material	ε ($\times 10^{-6}$V/K)	σ ($\times 10^3$S/m)	κ (W/m/K))	$Z(\times 10^{-3} K^{-1})$
Bi_2Te_3	200.000	110.000	1.60000	2.75000
800K Optimal	225.322	550.933	9.98962	2.80000
1000K Optimal	232.553	517.242	9.99032	2.80000
1200K Optimal	238.713	490.981	9.99217	2.80000

Proof of enhanced efficiency

To prove mathematically that the layered thermoelectric composite can achieve higher conversion efficiency than its constituents, we consider the bilayered composite shown in Fig. 4a, where we have taken $Z_1 = Z_2 = Z$ for simplicity. Since the various thermoelectric properties of constituent phase are tailored to match the optimal current density, it is evident that

$$H_{U,i} \leq H_{U,b} = \frac{T_h - T_m}{T_h} \frac{\sqrt{1 + Z\frac{T_h + T_m}{2}} - 1}{\sqrt{1 + Z\frac{T_h + T_m}{2}} + \frac{T_m}{T_h}}, \tag{1}$$

$$H_{L,i} \leq H_{L,b} = \frac{T_m - T_c}{T_m} \frac{\sqrt{1 + Z\frac{T_m + T_c}{2}} - 1}{\sqrt{1 + Z\frac{T_m + T_c}{2}} + \frac{T_c}{T_m}}, \tag{2}$$

where the subscriptions U and L denote the upper and lower segments separated by T_m, while subscript i is to denote phase 1 and 2; the bilayered composite is indicated by the subscript b. The overall efficiency for is thus,

$$H_i = 1 - (1 - H_{U,i})(1 - H_{L,i}) < H_b = 1 - (1 - H_{U,b})(1 - H_{L,b}), \tag{3}$$

proving that the optimal conversion efficiency of the bilayered composite can be higher than that of the constituent phases.