# Supplementary Materials

## Thermoelectric transport properties of $BaFe_2Fe_{16}O_{27}$ hexaferrites

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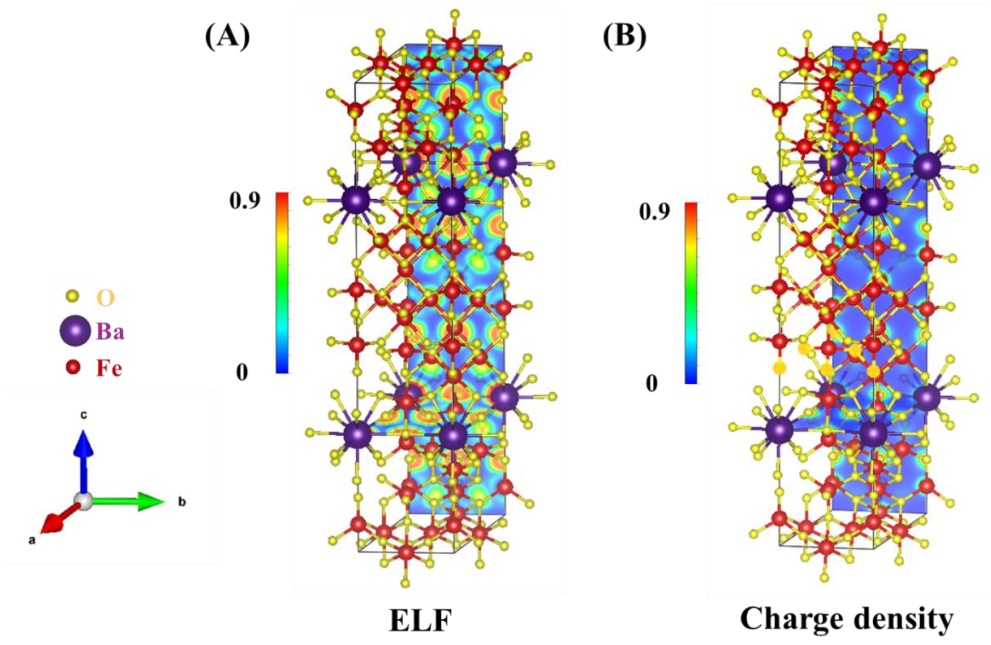


**Figure S1.** Calculated electron localization function (ELF) and charge transfer of $Fe_2W$.

We fitted the $C_p$ data with one Debye model and two Einstein models based on [1,2]

$$\frac{C_p}{T} = \varphi + \beta T^2 + A\Theta_{E1}{}^2 (T^2)^{-3/2} \cdot \frac{e^{\Theta_{E1}/T}}{(e^{\Theta_{E1}/T}-1)^2} + B\Theta_{E2}{}^2 \cdot \frac{e^{\Theta_{E2}/T}}{(e^{\Theta_{E2}/T}-1)^2} \quad (1)$$

where $\varphi$, $\beta$ are the electronic and Debye lattice contribution, respectively. $\Theta_{Ei}$ (i = 1, 2) is the

Einstein temperature of the two local vibrational modes, and A and B are the constants. In addition, Debye temperature can be estimated according to [2]

$$\Theta_D = (12\pi^4 pR/5\beta)^{1/3} \quad (2)$$

where $R$ is the gas constant and $p$ is the number of atoms in each molecule.

**Table S1.** Parameters obtained by fitting the low-temperature specific heat capacity of $BaFe_2Fe_{16}O_{27}$ using Debye-Einstein model.

| Parameter (Unit) | Value |
|---|---|
| $\varphi$ ($10^{-3}$ J/mol $K^{-2}$) | 44.5 |
| $\beta$ ($10^{-4}$ J/mol $K^{-2}$) | 6.5 |
| $\Theta_{E1}$ (K) | 47.7 |
| $\Theta_{E2}$ (K) | 127 |
| A | 10.5 |
| B | 70.0 |
| $\theta_D$ (K) | 516 |

Temperature-dependent lattice thermal conductivity could be modeled by using the Debye approximation

$$\kappa_L = \frac{k_B}{2\pi^2 v}\left(\frac{k_B T}{\hbar}\right)^3 \int_0^{\theta_D/T} \frac{x^4 e^x}{\tau_C^{-1}(e^x-1)^2} d \quad (3)$$

where the dimensionless quantity can be expressed as

$$x = \hbar\omega/k_B T \quad (4)$$

where $k_B$ is the boltzmann constant, ω is the phonon frequency, $\hbar$ is the reduced Planck constant, $\theta_D$ is the Debye temperature, $v$ is the sound velocity, and $\tau_C$ is the phonon-scattering relaxation time. The phonon-scattering relaxation time $\tau_C$ can be expressed by

$$\tau_C^{-1} = \frac{v}{L} + A\omega^4 + B\omega^2 T \exp\left(-\frac{\theta_D}{3T}\right) \quad (5)$$

where $L$ is the grain size and the coefficients A, B are the fitting parameters which related to the point defect scattering and Umklapp scattering, respectively. The terms on the right side of Equation 10 represent the influence of grain boundary, point defect, and phonon-phonon umklapp, respectively. $\theta_D$ and $v$ for $Fe_2W$-based ceramics are calculated by DFT calculation and values are 517 K and 3667 m/s (listed in Table 1), respectively.

$$A = \frac{\Omega_0}{4\pi v^3}\Gamma_{exp} \quad (6)$$

$$\Gamma_{mass}(U_u V_v W_w) = \frac{u\left(\frac{M_u}{M_m}\right)^2\Gamma(U)+v\left(\frac{M_v}{M_m}\right)^2\Gamma(V)+w\left(\frac{M_w}{M_m}\right)^2\Gamma(W)}{u+v+w} \quad (7)$$

$$\Gamma = \Phi(1-\Phi)\left(\frac{\Delta M}{M_{av}}\right)^2 \quad (8)$$

$$\Delta M = M_i - M_h \quad (9)$$

$$M_{av} = \Phi M_i + (1-\Phi)M_h \quad (10)$$

$$\Gamma_{strain} = \Gamma_{exp} - \Gamma_{mass} \quad (11)$$

where $\Omega_0$ is the volume of the primitive cell and $\Gamma_{exp}$ is an experimental disorder scattering parameter. $M_i$ and $M_h$ are the masses of the impurity and host atoms, respectively.

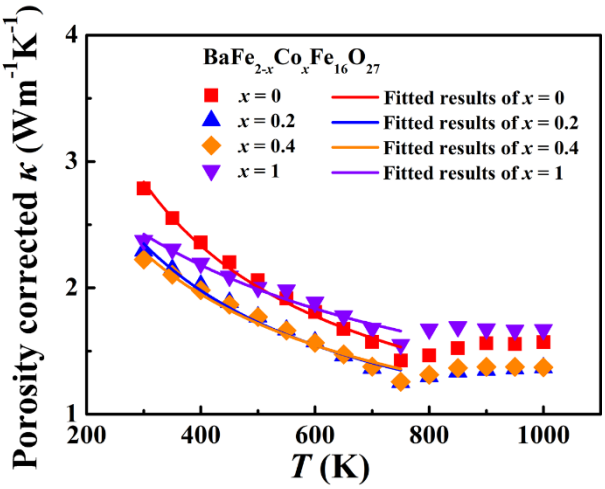


**Figure S2.** Porosity corrected thermal conductivities as functions of content of temperature for all $BaFe_{2-x}Co_xFe_{16}O_{27}$ ($x$ = 0, 0.2, 0.4, 1.0) ceramics ranged from 300 K to 750 K.

## REFERENCES


1. Dimitrov IK, Manley ME, Shapiro SM, Yang J, Zhang W, et al. Einstein modes in the phonon density of states of the single-filled skutterudite $Yb_{0.2}Co_4Sb_{12}$. *Phy Rev B* 2010;82:174301. DOI: 10.1103/PhysRevB.82.174301.

2. Jiang B, Qiu P, Eikeland E, Chen H, Song Q, et al. $Cu_8GeSe_6$-based thermoelectric materials with an argyrodite structure. *J Mater Chem C* 2017;5:943-52. DOI: 10.1039/C6TC05068A.