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Citation: Chin. Phys. B . 2020, 29(5): 057301 . **doi:** 10.1088/1674-1056/ab7d96
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Influence of spherical inclusions on effective thermoelectric properties of thermoelectric composite materials*

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(Received 1 December 2019; revised manuscript received 15 February 2020; accepted manuscript online 9 March 2020)

A homogenization theory is developed to predict the influence of spherical inclusions on the effective thermoelectric properties of thermoelectric composite materials based on the general principles of thermodynamics and Mori–Tanaka method. The closed-form solutions of effective Seebeck coefficient, electric conductivity, heat conductivity, and figure of merit for such thermoelectric materials are obtained by solving the nonlinear coupled transport equations of electricity and heat. It is found that the effective figure of merit of thermoelectric material containing spherical inclusions can be higher than that of each constituent in the absence of size effect and interface effect. Some interesting examples of actual thermoelectric composites with spherical inclusions, such as insulated cavities, inclusions subjected to conductive electric and heat exchange and thermoelectric inclusions, are considered, and the numerical results lead to the conclusion that considerable enhancement of the effective figure of merit is achievable by introducing inclusions. In this paper, we provide a theoretical foundation for analytically and computationally treating the thermoelectric composites with more complicated inclusion structures, and thus pointing out a new route to their design and optimization.

Keywords: thermoelectric composites, effective properties, spherical inclusions

PACS: 73.50.Lw, 84.60.Rb, 84.60.Bk, 46.25.Cc **DOI:** 10.1088/1674-1056/ab7d96

1. Introduction

Thermoelectric materials directly convert heat into electricity and *vice versa* due to the Seebeck effect and Peltier effect, and thus they are widely used for power generation in harvesting wasted heat, refrigeration, solar energy harvesting, and carbon reduction.[1–9] The thermoelectric devices have unique advantages: they are more compact, robust, and noiseless than the conventional mechanical providers of power generation and refrigeration because the thermoelectric devices are solid state heat engines and have no moving parts.[10–13] However, thermoelectric materials have a critical weakness of low conversion efficiency which is governed by a dimensionless figure of merit $ZT = \alpha^2\sigma T/k$, where α, σ, and k are the Seebeck coefficient, electric conductivity, and heat conductivity, respectively, T being the absolute temperature.[3,14] Thermoelectric materials with high figure of merit at a temperature T should combine with a large Seebeck coefficient α to provide a large electric potential difference, a large electric conductivity σ to reduce the Joule heat loss, and a small heat conductivity k to maintain a large temperature gradient.[15] As a result, most efforts dedicating to improving the conversion efficient have involved alloying, doping, quantum, and other size effects in thin thermoelectric films and mesoscopic systems.[16–27]

Lots of papers have focused on improving the properties of thermoelectric materials by using size effect and interface effect, but very few researchers have examined the classical composite mixtures as a route to enhancing the thermoelectric characteristics. In such thermoelectric composites, the granularity and heterogeneity are on a large enough scale that the physical processes can be well described in each individual phase by the classical thermodynamic theory.[28] The problem of thermoelectric properties of a composite medium was studied by Bergman and Levy,[29] and they found that the effective figure of merit of a thermoelectric composite material was bounded by its constituents from the micromechanical analysis. However, this conclusion was drawn based on the assumption that both $\alpha\sigma T$ and $\alpha^2\sigma T$ are material constants independent of temperature T, which linearizes the constitutive equations. In fact, both the effective figure of merit and the effective conversion efficiency of layered heterogeneous medium can be higher than all of its constituents, indicated by solving the rigorous nonlinearly coupled transport equations of electricity and heat.[30,31] The closed-form solutions of effective properties of a thermoelectric composite containing an elliptic inhomogeneity or an arbitrarily shaped hole were provided in Refs. [32,33]. A theoretical model to analyze the energy conversion efficiency of a cracked thermoelectric material with finite height and width based on the nonlinearly coupled transport equations of electricity and heat was developed by Zhang *et al.*[34] The three-dimensional ellipsoidal in-

*Project supported by the Ningbo Natural Science Foundation, China (Grant Nos. 2019A610151 and 2018A610081), the Natural Science Foundation of Zhejiang Province, China (Grant Nos. LY17A020001 and LY20A020002), the National Natural Science Foundation of China (Grant No. 11402063), and the K C Wong Magna Fund in Ningbo University, China.

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clusion problem in thermoelectric materials and the effective material properties of the matrix–inclusion system were studied by Wang *et al.*[35] From Refs. [30,32–35], it is also found that the effective figure of merit can exceed that of each constituent. A continuum linear theory for thermoelectric materials was developed by Liu under the conditions of small variations of temperature, electric potential, and their gradients, and this linear theory is further used to predict effective properties of thermoelectric composites.[36] An asymptotic homogenization model for three-dimensional thermoelectric composites was proposed and applied to the finite element simulations by Yang *et al.*[37] Most of the above papers have paid attention to the two-dimensional problem, these studies motivate us to developing a rigorous homogenization theory based on the three-dimensional thermoelectric constitutive model which can predict the macroscopic behavior of thermoelectric composites and furnish the design strategies for desired effective properties.

In this paper, we develop a homogenization theory to analyze the effective properties of thermoelectric composites based on the Mori–Tanaka method.[38] The rest of the present paper is organized as follows. The general framework of homogenization for thermoelectric composites is provided in Section 2. In Section 3, closed-form solutions of electric potential and temperature fields for the thermoelectric material containing a spherical inclusion are obtained by solving the nonlinear governing equations. We proceed to determine the effective properties of thermoelectric composites containing spherical inclusions based on the developed homogenization theory, and numerical results are given to illustrate that the effective figure of merit of such a material can be higher than that of each constituent in Section 4. Finally, some conclusions are drawn from the present study in Section 5.

2. General framework for effective properties of thermoelectric composites

2.1. Basic formulations for thermoelectric material

The properties of thermoelectric materials can be described by considering an isotropic homogeneous body $V \in R^n$, where $n = 2$ or 3 is the dimension of space. The thermodynamic state of the body is described by the absolute temperature T and electrochemical potential μ. The usual suffix notation is used in this paper, a repeated suffix is summed over x, y, and z, and suffixes preceded by a comma denote differentiation. The absolute temperature T, electrochemical potential μ, electric current density e_i, and heat flux density q_i are related by[2]

$$e_i = -\sigma\mu_{,i} - \sigma\alpha T_{,i}, \tag{1a}$$

$$q_i = -kT_{,i} + \alpha T e_i. \tag{1b}$$

The energy in the considered system is transported through both electrochemical potential and heat, and the energy flux density is defined as $u_i = \mu e_i + q_i$. The conservation of both electric charges and energy imply

$$e_{i,i} = 0, \tag{2a}$$

$$u_{i,i} = 0 \tag{2b}$$

for the stationary case where neither free electric charge nor heat source exists. From Eqs. (1) and (2), it can be seen that the electric potential and temperature are fully coupled, and this always makes a boundary value problem nonlinear and untractable. This difficulty may be addressed by introducing an additional state function $F = \mu + \alpha T$ which represents the total electrochemical potential including Seebeck effect.[39] Then, the new constitutive equation and governing equation can be expressed as

$$e_i = -\sigma F_{,i}, \tag{3a}$$

$$u_i = -\sigma F F_{,i} - kT_{,i}, \tag{3b}$$

$$\nabla^2 F = 0, \tag{4a}$$

$$k\nabla^2 T + \sigma F_{,i}^2 = 0. \tag{4b}$$

It should be noted that the total electrochemical potential F in Eq. (4) is uncouple from the whole problem and satisfies the Laplace's equation, so that the function F may be solved first from the theory of electrostatics, then the temperature function T can be determined with the aid of the known F and the boundary conditions. In addition, it is convenient to represent the temperature function T as the sum of two functions T_1 and T_2, where $\nabla^2 T_1 = 0$ and $k\nabla^2 T_2 + \sigma F_{,i}^2 = 0$. The solution of T_2 is $T_2 = -\left(\sigma F^2\right)/k$, then the energy flux density u_i can be obtained as

$$u_i = -kT_{1,i}. \tag{5}$$

Now this problem is reduced to determine the temperature function T_1 satisfying the Laplace's equation which has been studied very well.

2.2. General framework of homogenization for thermoelectric composites

The general framework of homogenization for thermoelectric composites is developed based on the Mori–Tanaka method in this Subsection.[38] Consider a three-dimensional thermoelectric material containing uniformly distributed spherical inclusions. The geometrical and physical quantities with superscripts "m" and "s" refer to those for the thermoelectric matrix material and internal inclusions, respectively, and the full quantities without a superscript refer to those for the entire thermoelectric composite material. The total volume of the solid can be expressed as $V = V^{\mathrm{m}} + V^{\mathrm{s}}$, and the volume fraction of the inclusions to the thermoelectric

composite material is defined as $f = V^{\rm s}/V$. The overall quantities defined are based on a volume-averaged concept, therefore the average electrochemical potential $\bar{\mu}$, electrochemical potential gradient $\bar{\mu}_{,i}$, electric current density $\bar{e}_i$, temperature $\bar{T}_1$, temperature gradient $\bar{T}_{1,i}$, and energy flux density $\bar{u}_i$ are expressed as

$$\begin{aligned}&[\bar{\mu},\ \bar{\mu}_{,i},\ \bar{e}_i,\ \bar{T}_1,\ \bar{T}_{1,i},\ \bar{u}_i]\\ &= \frac{1}{V}\int_V [\mu,\ \mu_{,i},\ e_i,\ T_1,\ T_{1,i},\ u_i]\,{\rm d}V.\end{aligned} \tag{6}$$

Similarly, the volume-averaged quantities for the inclusions and the thermoelectric matrix material can be written as follows:

$$\begin{aligned}&\left[\bar{\mu}^{\rm s},\ \bar{\mu}^{\rm s}_{,i},\ \bar{e}^{\rm s}_i,\ \bar{T}^{\rm s}_1,\ \bar{T}^{\rm s}_{1,i},\ \bar{u}^{\rm s}_i\right]\\ &= \frac{1}{V^{\rm s}}\int_{V^{\rm s}} [\mu,\ \mu_{,i},\ e_i,\ T_1,\ T_{1,i},\ u_i]\,{\rm d}V,\end{aligned} \tag{7}$$

$$\begin{aligned}&\left[\bar{\mu}^{\rm m},\ \bar{\mu}^{\rm m}_{,i},\ \bar{e}^{\rm m}_i,\ \bar{T}^{\rm m}_1,\ \bar{T}^{\rm m}_{1,i},\ \bar{u}^{\rm m}_i\right]\\ &= \frac{1}{V^{\rm m}}\int_{V^{\rm m}} [\mu,\mu_{,i},e_i,T_1,T_{1,i},u_i]\,{\rm d}V.\end{aligned} \tag{8}$$

The average quantities of the inclusions and thermoelectric matrix are related to those of the entire solid by volume fraction rule, that is

$$\bar{e}_i = (1-f)\,\bar{e}^{\rm m}_i + f\bar{e}^{\rm s}_i, \tag{9a}$$

$$\bar{F}_{,i} = (1-f)\,\bar{F}^{\rm m}_{,i} + f\bar{F}^{\rm s}_{,i}, \tag{9b}$$

$$\bar{u}_i = (1-f)\,\bar{u}^{\rm m}_i + f\bar{u}^{\rm s}_i, \tag{10a}$$

$$\bar{T}_{1,i} = (1-f)\,\bar{T}^{\rm m}_{1,i} + f\bar{T}^{\rm s}_{1,i}. \tag{10b}$$

If the thermoelectric transport process in the inclusions and thermoelectric material matrix follows Eqs. (3a) and (5), the average electric current density is related to the average electrochemical potential gradient by

$$\bar{e}^{\rm s}_i = -\sigma^{\rm s}\bar{F}^{\rm s}_{,i} \ \text{ in } \ V^{\rm s}, \tag{11a}$$

$$\text{or } \ \bar{F}^{\rm s}_{,i} = -\rho^{\rm s}\bar{e}^{\rm s}_i \ \text{ in } \ V^{\rm s}, \tag{11b}$$

with ρ being the electric resistivity (*i.e.*, the reciprocal of the electric conductivity σ), and the average energy flux density is related to the average temperature gradient by

$$\bar{u}^{\rm s}_i = -k^{\rm s}\bar{T}^{\rm s}_{1,i} \ \text{ in } \ V^{\rm s}, \tag{12a}$$

$$\text{or } \ \bar{T}^{\rm s}_{1,i} = -R^{\rm s}\bar{u}^{\rm s}_i \ \text{ in } \ V^{\rm s}, \tag{12b}$$

where R is the thermal resistivity, and $R = 1/k$. For the thermoelectric material matrix, we also have

$$\bar{e}^{\rm m}_i = -\sigma^{\rm m}\bar{F}^{\rm m}_{,i} \ \text{ in } \ V^{\rm m}, \tag{13a}$$

$$\text{or } \ \bar{F}^{\rm m}_{,i} = -\rho^{\rm m}\bar{e}^{\rm m}_i \ \text{ in } \ V^{\rm m}, \tag{13b}$$

$$\bar{u}^{\rm m}_i = -k^{\rm m}\bar{T}^{\rm m}_{1,i} \ \text{ in } \ V^{\rm m}, \tag{14a}$$

$$\text{or } \ \bar{T}^{\rm m}_{1,i} = -R^{\rm m}\bar{u}^{\rm m}_i \ \text{ in } \ V^{\rm m}. \tag{14b}$$

For a thermoelectric material with internal inclusions subject to homogeneous boundary conditions, in general, the electric current density and energy flux density inside the inclusions can be expressed as

$$\bar{e}^{\rm s}_i = \bar{H}\bar{e}^{\rm m}_i, \tag{15a}$$

$$\bar{u}^{\rm s}_i = \bar{G}\bar{u}^{\rm m}_i, \tag{15b}$$

where $\bar{H}$ and $\bar{G}$ are the local concentration factors to be determined. Substituting Eq. (15) into Eqs. (9a) and (10a), respectively, we have

$$\bar{e}^{\rm m}_i = (1-f+f\bar{H})^{-1}\,\bar{e}_i, \tag{16a}$$

$$\bar{u}^{\rm m}_i = \left(1-f+f\bar{G}\right)^{-1}\bar{u}_i. \tag{16b}$$

Using Eqs. (9b), (10b), (13b), (14b), and (16), the average electric current density and energy flux density are obtained as follows:

$$\bar{F}_{,i} = -\left[(1-f)\,\rho^{\rm m} + f\bar{H}\rho^{\rm s}\right](1-f+f\bar{H})^{-1}\bar{e}_i, \tag{17a}$$

$$\bar{T}_{1,i} = -\left[(1-f)\,R^{\rm m} + f\bar{G}R^{\rm s}\right]\left(1-f+f\bar{G}\right)^{-1}\bar{u}_i. \tag{17b}$$

The effective electric resistivity $\bar{\rho}$ and thermal resistivity $\bar{R}$ of the thermoelectric composite material are respectively defined as

$$\bar{F}_{,i} = -\bar{\rho}\bar{e}_i, \tag{18a}$$

$$\bar{T}_{1,i} = -\bar{R}\bar{u}_i, \tag{18b}$$

and can be given by

$$\bar{\rho} = \left[(1-f)\,\rho^{\rm m} + f\bar{H}\rho^{\rm s}\right](1-f+f\bar{H})^{-1}, \tag{19a}$$

$$\bar{R} = \left[(1-f)\,R^{\rm m} + f\bar{G}R^{\rm s}\right]\left(1-f+f\bar{G}\right)^{-1}. \tag{19b}$$

As a result, the electric conductivity $\bar{\sigma}$ and heat conductivity $\bar{k}$ are obtained as follows:

$$\bar{\sigma} = \sigma^{\rm m} - \frac{f\bar{H}\,(\sigma^{\rm m}-\sigma^{\rm s})\,\sigma^{\rm m}}{(1-f)\,\sigma^{\rm s} + f\bar{H}\sigma^{\rm m}}, \tag{20a}$$

$$\bar{k} = k^{\rm m} - \frac{f\bar{G}\,(k^{\rm m}-k^{\rm s})\,k^{\rm m}}{(1-f)\,k^{\rm s} + f\bar{G}k^{\rm m}}. \tag{20b}$$

It should be noted that $\bar{\sigma}$ and $\bar{k}$ for thermoelectric composites with internal inclusions have the same expressions as those obtained based on the Mori–Tanaka method. If the open-circuit boundary condition $e_i = 0$ is used, then the effective Seebeck coefficient $\bar{\alpha}$ can be derived as $\alpha = -\mu_{,i}/T_{1,i}$. Thus

$$\bar{\alpha} = -\frac{(1-f)\,\bar{\mu}^{\rm m}_{,i} + f\bar{\mu}^{\rm s}_{,i}}{(1-f)\,\bar{T}^{\rm m}_{1,i} + f\bar{T}^{\rm s}_{1,i}}. \tag{21}$$

Using the relationship $F = \mu + \alpha T$ and noting that $F_{,i} = 0$, $\bar{\alpha}$ is finally obtained as

$$\bar{\alpha} = \alpha^{\rm m} - \frac{f\,(\alpha^{\rm m}-\alpha^{\rm s})\,\bar{T}^{\rm s}_{1,i}/\bar{T}^{\rm m}_{1,i}}{1-f+f\bar{T}^{\rm s}_{1,i}/\bar{T}^{\rm m}_{1,i}}, \tag{22a}$$

or

$$\bar{\alpha} = \alpha^{\mathrm{m}} - \frac{f(\alpha^{\mathrm{m}} - \alpha^{\mathrm{s}})k^{\mathrm{m}}\bar{G}/k^{\mathrm{s}}}{1 - f + fk^{\mathrm{m}}\bar{G}/k^{\mathrm{s}}}. \tag{22b}$$

In the absence of internal inclusions (that is, $f = 0$), equation (22b) is reduced to $\bar{\alpha} = \alpha^{\mathrm{m}}$. If $f = 1$, equation (22b) has the form of $\bar{\alpha} = \alpha^{\mathrm{s}}$. The effective figure of merit for the thermoelectric composite material can be defined as $\bar{Z} = \bar{\alpha}^2\bar{\sigma}/\bar{k}$ from the effective properties. It is noted that the derivation process of the effective electric conductivity, heat conductivity and Seebeck coefficient are independent of the geometrical configuration of the internal inclusions, therefore equations (20) and (22) are suitable for all the problem of thermoelectric composites containing internal inclusions when the local concentration factors $\bar{H}$ and $\bar{G}$ are obtained. However, the $\bar{H}$ and $\bar{G}$ are determined based on the knowledge of the geometrical configuration of the internal inclusions.

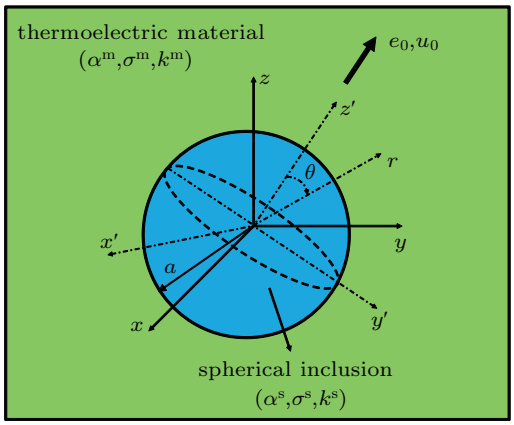


Fig. 1. Schematic diagram of thermoelectric material containing a spherical inclusion subjected to the electric current and energy flux in z' direction.

The local concentration factors $\bar{H}$ and $\bar{G}$ can be calculated for the single thermoelectric inclusion with the involved electrical and thermal properties being the overall ones by using the Mori–Tanaka approach. The effective electric conductivity, heat conductivity and Seebeck coefficient are calculated according to Eqs. (20) and (22) through the average taken over all the possible orientations of the inclusion in the thermoelectric matrix material. For this, the prime coordinate system (x', y', z') shown in Fig. 1 is introduced, and the unit normal in the prime coordinate system are related to those in the physical coordinate system by the Euler angles Θ and Φ:

$$\boldsymbol{n}_i = \beta_{ij}\boldsymbol{n}'_j \tag{23}$$

with

$$\beta_{ij} = \begin{bmatrix} -\sin\Theta & -\sin\Phi\cos\Theta & \cos\Phi\cos\Theta \\ \cos\Theta & -\sin\Phi\sin\Theta & \cos\Phi\sin\Theta \\ 0 & \cos\Phi & \sin\Phi \end{bmatrix}, \tag{24}$$

where $0 \leqslant \Theta \leqslant 2\pi$ and $0 \leqslant \Phi \leqslant \pi/2$. The local concentration factors $\bar{H}'$ and $\bar{G}'$ can be determined by the solutions of the single inclusion, then $\bar{H}'$ and $\bar{G}'$ are transformed back to the physical coordinates according to Eq. (23) to obtain $\bar{H}$ and $\bar{G}$, the results are finally averaged over all the possible orientations of the inclusions to obtain the overall thermoelectric properties[40,41]

$$\bar{H}_{ij} = \frac{N}{2\pi}\int_0^{2\pi}\int_0^{\pi/2}\beta_{im}\beta_{jn}\bar{H}'_{mn}\cos\Phi\mathrm{d}\Phi\mathrm{d}\Theta, \tag{25a}$$

$$\bar{G}_{ij} = \frac{N}{2\pi}\int_0^{2\pi}\int_0^{\pi/2}\beta_{im}\beta_{jn}\bar{G}'_{mn}\cos\Phi\mathrm{d}\Phi\mathrm{d}\Theta, \tag{25b}$$

with β_{jn} being the transformation tensor defined in Eq. (24) and N the total number of internal inclusions per unit volume.

3. Solutions for thermoelectric material with a spherical inclusion

The practical use of the homogenization theory for the thermoelectric composites is illustrated by considering the case of internal spherical inclusions (see Fig. 1) in the thermoelectric matrix. In sequence, the model involves the determination of (i) the distribution of electric current density and energy flux density inside a single spherical inclusion to obtain the $\bar{H}'$ and $\bar{G}'$; (ii) the $\bar{H}$ and $\bar{G}$ according Eq. (25); (iii) the effective electric conductivity, heat conductivity, and Seebeck coefficient according to Eqs. (20) and (22); and (iv) the effective thermoelectric figure of merit.

The incoming electric current density e_0 and energy flux density u_0 in a three-dimensional thermoelectric material are assumed to be disturbed by spherical thermoelectric inclusions with an average radius a, and the applied e_0 and u_0 are aligned to be in the z' direction (Fig. 1). The following spherical coordinate (r, θ, ϕ) is introduced:

$$(x', y', z') = (r\sin\theta\cos\phi, r\sin\theta\sin\phi, r\cos\theta). \tag{26}$$

For a spherical inclusion with ϕ-symmetry, the solutions of temperature fields T_2 can be obtained by $T_2 = -(\sigma F^2)/k$, and the solution of F and T_1 have the form of

$$F^{\mathrm{m}} = \left(A_{1\mathrm{m}}r + B_{1\mathrm{m}}/r^2\right)\cos\theta, \quad \text{for } r \geqslant a, \tag{27a}$$

$$F^{\mathrm{s}} = A_{1\mathrm{s}}r\cos\theta, \quad \text{for } r < a, \tag{27b}$$

$$T_1^{\mathrm{m}} = D_{0\mathrm{m}} + \left(D_{1\mathrm{m}}r + E_{1\mathrm{m}}/r^2\right)\cos\theta + \left(E_{2\mathrm{m}}/2r^3\right)\left(3\cos^2\theta - 1\right), \quad \text{for } r \geqslant a, \tag{28a}$$

$$T_1^{\mathrm{s}} = D_{0\mathrm{s}} + D_{1\mathrm{s}}r\cos\theta + \left(D_{2\mathrm{s}}r^2 + E_{2\mathrm{s}}/r^3\right) \times \left(3\cos^2\theta - 1\right)/2, \quad \text{for } r < a, \tag{28b}$$

with $A_{1\mathrm{m}}$, $B_{1\mathrm{m}}$, $A_{1\mathrm{s}}$, $D_{0\mathrm{m}}$, $D_{1\mathrm{m}}$, $E_{1\mathrm{m}}$, $E_{2\mathrm{m}}$, $D_{0\mathrm{s}}$, $D_{1\mathrm{s}}$, $D_{2\mathrm{s}}$, and $E_{2\mathrm{s}}$ being coefficients to be determined from the following boundary conditions:

$$e_z^{\mathrm{m}} = e_0, \quad \text{for } z \to \infty, \tag{29a}$$

$$\mu^{\mathrm{m}} = \mu^{\mathrm{s}}, \quad \text{for } r = a, \tag{29b}$$

$$e_r^{\mathrm{m}} = e_r^{\mathrm{s}}, \quad \text{for } r = a, \tag{29c}$$

$$u_z^{\mathrm{m}} = u_0, \quad \text{for } z \to \infty, \tag{30a}$$
$$T^{\mathrm{m}} = T^{\mathrm{s}}, \quad \text{for } r = a, \tag{30b}$$
$$u_r^{\mathrm{m}} = u_r^{\mathrm{s}}, \quad \text{for } r = a. \tag{30c}$$

From Eqs. (29a) and (30a), the electric current density e_z^{m} and energy density u_z^{m} should be identical to the applied electric current density e_0 and applied energy density u_0 at a distance away from the spherical inclusion, which gives

$$A_{1\mathrm{m}} = -e_0/\sigma^{\mathrm{m}}, \tag{31a}$$
$$D_{1\mathrm{m}} = -u_0/k^{\mathrm{m}}. \tag{31b}$$

Making use of the interface boundary conditions between the spherical inclusion and the thermoelectric material matrix, the other coefficients are obtained from Eqs. (29b), (29c), (30b), and (30c) as follows:

$$A_{1\mathrm{s}} = -\frac{3e_0}{2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}} + \frac{6\left(\alpha^{\mathrm{m}}-\alpha^{\mathrm{s}}\right)\sigma^{\mathrm{m}}u_0}{\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right)\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right)}, \tag{32a}$$
$$B_{1\mathrm{m}} = -\frac{\left(\sigma^{\mathrm{m}}-\sigma^{\mathrm{s}}\right)a^3e_0}{\sigma^{\mathrm{m}}\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right)} - \frac{3\left(\alpha^{\mathrm{m}}-\alpha^{\mathrm{s}}\right)\sigma^{\mathrm{s}}a^3u_0}{\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right)\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right)}, \tag{32b}$$
$$D_{0\mathrm{s}} = \frac{3a^2\sigma^{\mathrm{s}}}{8k^{\mathrm{s}}\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right)^2}\left[e_0 - \frac{8\left(\alpha^{\mathrm{m}}-\alpha^{\mathrm{s}}\right)\sigma^{\mathrm{m}}u_0}{2k^{\mathrm{m}}+k^{\mathrm{s}}}\right]^2, \tag{32c}$$
$$D_{0\mathrm{m}} = \frac{3a\sigma^{\mathrm{m}}}{2k^{\mathrm{m}}\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right)^2}\left[e_0 + \frac{\left(\alpha^{\mathrm{m}}-\alpha^{\mathrm{s}}\right)\sigma^{\mathrm{s}}u_0}{2k^{\mathrm{m}}+k^{\mathrm{s}}}\right]^2, \tag{32d}$$
$$D_{1c} = -\frac{3u_0}{2k^{\mathrm{m}}+k^{\mathrm{s}}}, \tag{32e}$$
$$E_{1\mathrm{m}} = -\frac{\left(k^{\mathrm{m}}-k^{\mathrm{s}}\right)a^3u_0}{k^{\mathrm{m}}\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right)}, \tag{32f}$$
$$E_{2\mathrm{m}} = \frac{3a^5\sigma^{\mathrm{m}}}{k^{\mathrm{m}}\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right)^2}\left[e_0 + \frac{\left(\alpha^{\mathrm{m}}-\alpha^{\mathrm{s}}\right)\sigma^{\mathrm{s}}u_0}{2k^{\mathrm{m}}+k^{\mathrm{s}}}\right]^2, \tag{32g}$$
$$D_{2\mathrm{s}} = \frac{6}{5a^2}D_{0\mathrm{s}} - \frac{3k^{\mathrm{m}}}{5a^5k^{\mathrm{s}}}E_{2\mathrm{m}}, \tag{32h}$$
$$E_{2\mathrm{s}} = \frac{4a^3}{5}D_{0\mathrm{s}} + \frac{3k^{\mathrm{m}}}{5k^{\mathrm{s}}}E_{2\mathrm{m}}. \tag{32i}$$

Referring to Eqs. (3a), (15a), (27), and (32), the $\bar{H}'$ function is immediately obtained when the applied energy density u_0 is equal to zero, and given as

$$\bar{H}' = \frac{3\sigma^{\mathrm{s}}}{2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}}. \tag{33}$$

Similarly, the $\bar{G}'$ function is obtained from Eqs. (3b), (15b), (28), and (32) as

$$\bar{G}' = \frac{3k^{\mathrm{s}}}{2k^{\mathrm{m}}+k^{\mathrm{s}}}. \tag{34}$$

For the case of spherical inclusions, the results from Eq. (25) are obtained as follows:

$$\bar{H} = \bar{H}'/3 = \sigma^{\mathrm{s}}/\left(2\sigma^{\mathrm{m}}+\sigma^{\mathrm{s}}\right), \tag{35a}$$
$$\bar{G} = \bar{G}'/3 = k^{\mathrm{s}}/\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right). \tag{35b}$$

4. Effective thermoelectric properties

From Eqs. (20), (22b), and (35), the effective thermoelectric properties of thermoelectric material with spherical thermoelectric inclusions are described by

$$\bar{\alpha} = \alpha^{\mathrm{m}} - \frac{fk^{\mathrm{m}}\left(\alpha^{\mathrm{m}}-\alpha^{\mathrm{s}}\right)}{(2-f)k^{\mathrm{m}}+(1-f)k^{\mathrm{s}}}, \tag{36a}$$
$$\bar{\sigma} = \sigma^{\mathrm{m}} - \frac{f\sigma^{\mathrm{m}}\left(\sigma^{\mathrm{m}}-\sigma^{\mathrm{s}}\right)}{(2-f)\sigma^{\mathrm{m}}+(1-f)\sigma^{\mathrm{s}}}, \tag{36b}$$
$$\bar{k} = k^{\mathrm{m}} - \frac{fk^{\mathrm{m}}\left(k^{\mathrm{m}}-k^{\mathrm{s}}\right)}{(2-f)k^{\mathrm{m}}+(1-f)k^{\mathrm{s}}}, \tag{36c}$$
$$\bar{Z} = \frac{\left\{1-f+fk^{\mathrm{m}}\alpha^{\mathrm{s}}/\left[\alpha^{\mathrm{m}}\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right)\right]\right\}^2\left\{1-f\left(\sigma^{\mathrm{m}}-\sigma^{\mathrm{s}}\right)/\left[(2-f)\sigma^{\mathrm{m}}+(1-f)\sigma^{\mathrm{s}}\right]\right\}}{(1-f/2)\left[1-f+fk^{\mathrm{m}}/\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right)\right]\left[1-f+fk^{\mathrm{s}}/\left(2k^{\mathrm{m}}+k^{\mathrm{s}}\right)\right]}Z^{\mathrm{m}}. \tag{36d}$$

The influence of the type of spherical inclusion on the effective properties of thermoelectric composite material will be studied in this section. The following dimensionless parameters such as $\alpha_0 = \alpha^{\mathrm{s}}/\alpha^{\mathrm{m}}$, $\sigma_0 = \sigma^{\mathrm{s}}/\sigma^{\mathrm{m}}$, $k_0 = k^{\mathrm{s}}/k^{\mathrm{m}}$ are introduced for the convenience of discussion.

4.1. Insulated spherical cavities

The electric and heat properties of cavities filled with air or vacuum are always neglected in the theoretical analysis because air is a very good insulator. Therefore α_0, σ_0, and k_0 are all equal to zero, and the effective thermoelectric properties from Eq. (36) are reduced to

$$\bar{\alpha} = \frac{1-f}{1-f/2}\alpha^{\mathrm{m}}, \tag{37a}$$
$$\bar{\sigma} = \frac{1-f}{1-f/2}\sigma^{\mathrm{m}}, \tag{37b}$$
$$\bar{k} = \frac{1-f}{1-f/2}k^{\mathrm{m}}, \tag{37c}$$
$$\bar{Z} = \left(\frac{1-f}{1-f/2}\right)^2 Z^{\mathrm{m}}. \tag{37d}$$

From Eq. (37), it can be seen that the effective Seebeck coefficient $\bar{\alpha}$, effective electric conductivity $\bar{\sigma}$, and effective

heat conductivity $\bar{k}$ have the same variation trend with the volume fraction f for the case of insulated spherical cavities.

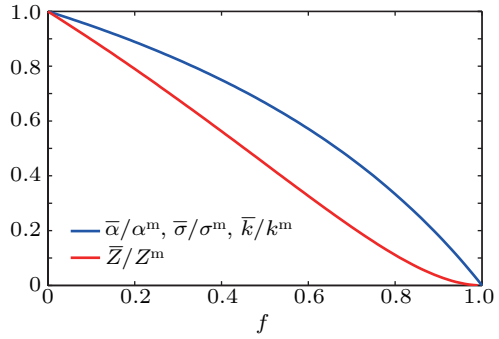


Fig. 2. Effective properties of thermoelectric materials containing spherical cavities.

The dependence of the effective properties of thermoelectric material containing insulated spherical cavities on f is plotted in Fig. 2, the $\bar{Z}/Z^{\mathrm{m}}$ is almost linearly varied from 1 to 0 as the f increases.

4.2. Spherical inclusions subjected to conductive electric and heat exchange

The conductive electric and heat transfer across the spherical inclusion are considered to examine the influence of inclusions on the effective thermoelectric properties. For this case, $\alpha_0 = 0$, σ_0 and k_0 are no longer equal to zero, and thus the effective Seebeck coefficient, electric conductivity, heat conductivity, and figure of merit are given as

$$\bar{\alpha} = \left[1 - \frac{f}{(2-f)+(1-f)k_0}\right]\alpha^{\mathrm{m}}, \tag{38a}$$

$$\bar{\sigma} = \left[1 - \frac{f(1-\sigma_0)}{(2-f)+(1-f)\sigma_0}\right]\sigma^{\mathrm{m}}, \tag{38b}$$

$$\bar{k} = \left[1 - \frac{f(1-k_0)}{(2-f)+(1-f)k_0}\right]k^{\mathrm{m}}, \tag{38c}$$

$$\bar{Z} = \frac{(1-f)^2[2(1-f)+\sigma_0]}{[1-f+f/(2+k_0)][1-f+fk_0/(2+k_0)][(2-f)+(1-f)\sigma_0]}Z^{\mathrm{m}}. \tag{38d}$$

Figures 3–6 show that the numerical results for effective properties of the thermoelectric material containing spherical inclusions ($\alpha_0 = 0$) with the different values of volume fraction, electric and heat conductivity. The volume fraction f is shown to have a major influence on the effective properties. From Figs. 3, 4, and 6, we can see that the effective figure of merit $\bar{Z}/Z^{\mathrm{m}}$ is improved when the electric conductivity is significantly larger than that of thermoelectric material if the inclusions are thermal insulations or their heat conductivity is small. However, for the electrical insulated inclusions, the effective figure of merit $\bar{Z}/Z^{\mathrm{m}}$ is always smaller than 1 (Fig. 5). It can be found that the optimized volume fraction corresponding to the maximum effective figure of merit is significantly dependent on the electric and heat properties of inclusions, and the performance of thermoelectric materials containing inclusions can be enhanced when the inclusions are properly introduced.

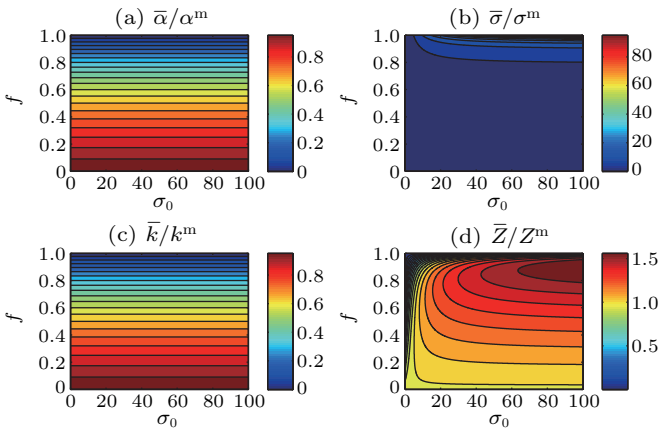


Fig. 3. Effective properties of thermoelectric material containing thermal-insulated spherical inclusions ($k_0 = 0$): (a) Seebeck coefficient $\bar{\alpha}/\alpha^{\mathrm{m}}$, (b) electric conductivity $\bar{\sigma}/\sigma^{\mathrm{m}}$, and (c) thermal conductivity $\bar{k}/k^{\mathrm{m}}$, and (d) figure of merit $\bar{Z}/Z^{\mathrm{m}}$.

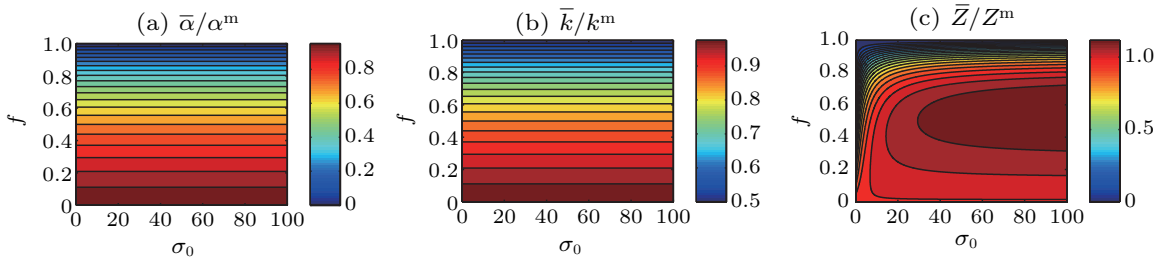


Fig. 4. Effective properties of thermoelectric material containing spherical-inclusions ($k_0 = 0.5$): (a) Seebeck coefficient $\bar{\alpha}/\alpha^{\mathrm{m}}$, (b) thermal conductivity $\bar{k}/k^{\mathrm{m}}$, and (c) figure of merit $\bar{Z}/Z^{\mathrm{m}}$.

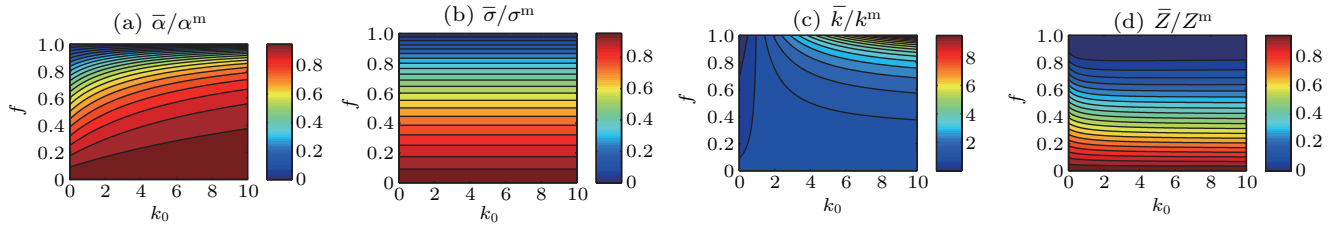


Fig. 5. Effective properties of thermoelectric material containing electrical-insulated spherical inclusions ($\sigma_0 = 0$): (a) Seebeck coefficient $\bar{\alpha}/\alpha^{\mathrm{m}}$, (b) electric conductivity $\bar{\sigma}/\sigma^{\mathrm{m}}$, (c) thermal conductivity $\bar{k}/k^{\mathrm{m}}$, and (d) figure of merit $\bar{Z}/Z^{\mathrm{m}}$.

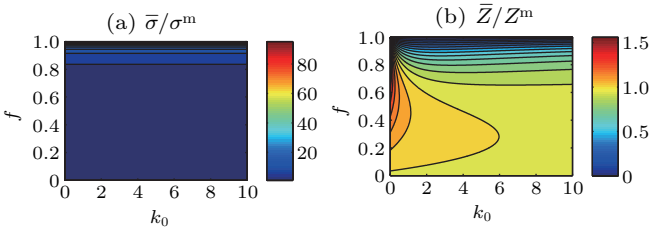


Fig. 6. Effective properties of thermoelectric material containing spherical inclusions ($\sigma_0 = 100$): (a) electric conductivity $\bar{\sigma}/\sigma^{\mathrm{m}}$ and (b) figure of merit $\bar{Z}/Z^{\mathrm{m}}$.

4.3. Thermoelectric spherical inclusions

The thermoelectric effect of spherical inclusions will be considered based on the analysis in Subsection 4.2, for $\alpha_0 \neq 0$, $\sigma_0 \neq 0$, and $k_0 \neq 0$. In this case, the effective Seebeck coefficient is given as

$$\bar{\alpha} = \left[1 - \frac{f(1-\alpha_0)}{(2-f)+(1-f)k_0}\right]\alpha^{\mathrm{m}}. \tag{39}$$

The effective electric conductivity and thermal conductivity are in accordance with the Eqs. (38b) and (38c), respectively. The effective figure of merit is

$$\bar{Z} = \frac{[1-f+f\alpha_0/(2+k_0)]^2\,[2(1-f)+\sigma_0]}{[1-f+f/(2+k_0)]\,[1-f+fk_0/(2+k_0)]\,[(2-f)+(1-f)\,\sigma_0]}Z^{\mathrm{m}}. \tag{40}$$

From Eq. (40), we can also see that the effective figure of merit $\bar{Z}$ of thermoelectric composites with spherical thermoelectric inclusions increases with the values of α_0 and σ_0 increasing, but decreases with the value of k_0 increasing. Figures 7 and 8 show that the variations of effective properties of thermoelectric materials containing thermoelectric spherical inclusions with volume fraction and Seebeck coefficient. The effective figure of merit is improved by introducing thermoelectric inclusions with higher figure of merit than that of thermoelectric material matrix.

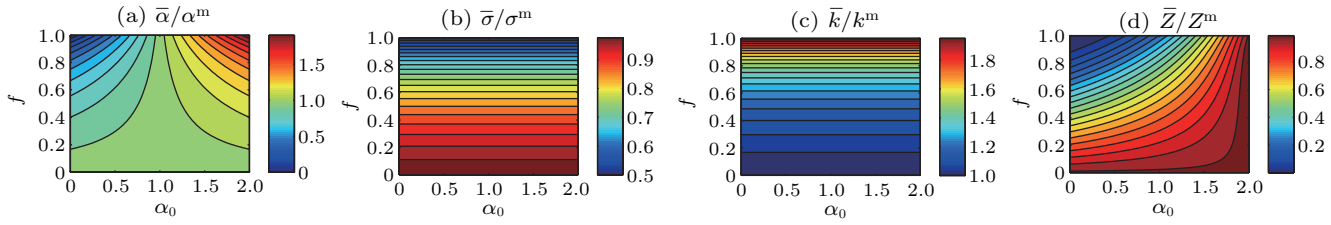


Fig. 7. Effective properties of thermoelectric material containing thermoelectric spherical inclusions ($\sigma_0 = 0.5$, $k_0 = 2$): (a) Seebeck coefficient $\bar{\alpha}/\alpha^{\mathrm{m}}$, (b) electric conductivity $\bar{\sigma}/\sigma^{\mathrm{m}}$, (c) thermal conductivity $\bar{k}/k^{\mathrm{m}}$, and (d) figure of merit $\bar{Z}/Z^{\mathrm{m}}$.

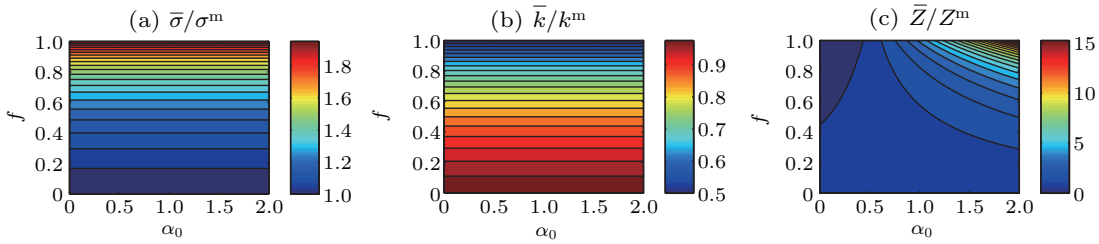


Fig. 8. Effective properties of thermoelectric material containing thermoelectric spherical inclusions ($\sigma_0 = 2$, $k_0 = 0.5$): (a) electric conductivity $\bar{\sigma}/\sigma^{\mathrm{m}}$, (b) thermal conductivity $\bar{k}/k^{\mathrm{m}}$, and (c) figure of merit $\bar{Z}/Z^{\mathrm{m}}$.

It seems that the figure of merit of two-phase thermoelectric composite is bounded from above by the larger one of the constituent phases. However, what we calculate is the effective figure of merit of two groups of Bi_2Te_3-based thermoelectric composites, the inclusion phases are $Bi_{0.5}Sb_{1.5}Te_3$ and BiSbTe, respectively. The material constants used in the calculation are listed in Table 1.[42] Figure 9 shows that the effective Seebeck coefficient, electric conductivity, and heat conductiv-

ity increase or decrease monotonically from the properties of Bi_2Te_3 to those of inclusion phase as the volume fraction of inclusions increases. It should be noted that there is a peak in the effective figure of merit, which exceeds both constituents'. The maximum figure of merit $\bar{Z}/Z^{\rm m} = 1.0955$ is attained for the thermoelectric composite material $Bi_2Te_3/Bi_{0.5}Sb_{1.5}Te_3$ with a corresponding volume fraction of inclusions $f = 0.80$. The results may illustrate that the effective figure of merit of thermoelectric composites can be higher than that of each constituent. The similar conclusion is obtained when the effective properties of layered heterogeneous medium is studied.[30,31]

Table 1. Material properties of thermoelectric materials.[42]

Materials	$\alpha/(\mu V/K)$	$\sigma/(S/m)$	$k/(W/mK)$
Bi_2Te_3	200	110000	1.6
BiSbTe	213	27700	0.7
$Bi_{0.5}Sb_{1.5}Te_3$	250	38000	0.85

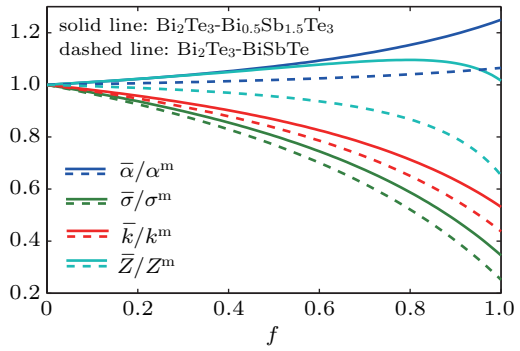


Fig. 9. Effective properties of Bi_2Te_3-based thermoelectric composites with spherical inclusions.

Finally, we point out that it is very difficult to analytically determine the optimized relationship between f and $(\alpha_0, \sigma_0, k_0)$ under the condition of $\bar{Z} > \max(Z^{\rm m}, Z^{\rm s})$ from Eq. (40). However, in this paper we provide a new validation method to estimate what properties of (thermoelectric) inclusions can be introduced to obtain the higher effective figure of merit of thermoelectric composites than that of its constituent without considering the size and interface effects.

5. Conclusions

In this paper, we proposed a micro-mechanical model to estimate the effective properties of the thermoelectric material containing spherical inclusions based on the Mori–Tanaka method. The general framework of homogenization for thermoelectric composites is developed by following the thermodynamic principle. Solutions of a spherical thermoelectric inclusion embedded in a thermoelectric material matrix subjected to uniform electric current density and energy flux density are derived, and the closed-form expressions of effective Seebeck coefficient, electric conductivity, heat conductivity, and figure of merit are obtained. Numerical results show that the effective properties are significantly degraded when the spherical cavities are produced inside the thermoelectric material. The effective figure of merit of thermoelectric composites can be improved by appropriately introducing (thermoelectric) inclusions, and it can be higher than that of each constituent.

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