JOURNAL OF MATERIAL SCIENCE 28 (1993) 5265–5268

Porosity dependence of material elastic moduli

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A new equation $E = E_0(1 + aP + bP^2)/(1 + cP)$, where E and E_0 are Young's moduli at porosity P and zero, respectively, and a, b, c are constants, has been derived. Our theoretical derivation is based on the dependence of sound velocity on the Young's modulus of the material.

1. Introduction

Ceramic systems are materials with high potential for engineering applications. Ceramics are usually prepared by sintering and so porous materials can occur if this process is incomplete, but they can be used in their own right in applications. Therefore, investigation of the effect of porosity on the mechanical properties of solids is of broad scientific as well as technological interest. A number of expressions for the relation between Young's modulus and porosity have been proposed. For example

$$E = E_0 \exp(-bP) \tag{1}$$

$$E = E_0 \exp[-(bP + cP^2)] \tag{2a}$$

$$E = E_0 \exp[-(bP + cP^2 + dP^3 + \dots)] \tag{2b}$$

$$E = E_0(1 - f_1 P + f_2 P^2) \tag{3}$$

$$E = E_0(1 - hP) \tag{4}$$

$$E = E_0(1 - aP)^n \tag{5}$$

$$E = E_0\left(1 - \frac{AP}{1 - (A + 1)P}\right) \tag{6}$$

The number of each equation here is the same as the number of the corresponding reference [1–6]. Here E and E_0 are Young's moduli at porosity P and $P = 0$, respectively, and other letters represent empirical constants. Among the above-mentioned expressions there are empirical ones [1, 4], semi-empirical [2, 5] and more or less theoretically based relations [3, 6].

The direct relation between stress and deformation was used for derivation of the E–P relation in the majority of the above-mentioned publications. Our derivation is based on the connection between the sound velocity and elastic moduli. We have calculated the sound velocity as a function of porosity and it enabled us to obtain the relation between Young's modulus and porosity on the form

$$E = E_0 \frac{1 + aP + bP^2}{1 + cP} \tag{7}$$

where a, b, c are constants independent of porosity, but dependent on the shape and size of the average pore as well as on the properties of the investigated material without pores. For illustration, we compare our Equation with the exact Wang's curve [2] obtained numerically for his theoretical model. These two curves agree very well over nearly the whole interval of porosity. Deviation of our curve from that of Wang occurs near the top of the porosity interval, when our treatment is not applicable.

2. Calculation of sound velocity for simple model of porous material

Because sound velocities provide an important constraint on the elastic moduli of materials, we have theoretically derived the sound velocity as a function of porosity. The sound velocity can be determined if we know the dispersion relation of acoustic phonons in the long-wave region (this is $\omega = \omega(\mathbf{k})$, for $\mathbf{k} \to 0$, where ω is the frequency of sound and $\mathbf{k}$ is the wave vector) [7]. For small $\mathbf{k}$, the dispersion relation of acoustic phonons has the form

$$\omega = \mathbf{c}_i \cdot \mathbf{k} \tag{8}$$

where $\mathbf{c}_i$ is the sound velocity. The subscript i determines the polarization of the sound wave (longitudinal or transversal).

Let

$$\mathbf{u}(\mathbf{n}) = \begin{pmatrix} u_1(\mathbf{n}) \\ u_2(\mathbf{n}) \\ u_3(\mathbf{n}) \end{pmatrix}$$

be the vector of deviation from equilibrium of an atom in lattice site $\mathbf{n}$ (in the case that it is a material with one atom in the elementary cell). In the case of more complicated materials with more atoms in the elementary cell, the vector $\mathbf{u}(\mathbf{r})$ describes the deviation from equilibrium of the centre of mass of this elementary cell.

We will study acoustic long-wave phonons (sound) and for this we are interested in movement of the entire elementary cell. This means that we are suggesting that the mutual positions of atoms in one elementary cell are unchanging. Change of these mutual positions is connected with high-energy optical phonons, which are not of interest in this case.

The equation for small deviations in an ideal crystal is [8]

$$m\ddot{\mathbf{u}}(\mathbf{n}, t) = -\sum_{\mathbf{n}'} \bar{\bar{A}}(\mathbf{n} - \mathbf{n}')\mathbf{u}(\mathbf{n}', t) \tag{9}$$

where $\bar{\bar{A}}(\mathbf{n} - \mathbf{n}')$ is a 3×3 matrix, called the dynamic power matrix, which describes the power with which an atom (or cell) deviated at point $\mathbf{n}'$ about $\mathbf{u}(\mathbf{n}')$ acts on the atom (cell) at point $\mathbf{n}$ deviated about $\mathbf{u}(\mathbf{n})$; m is the mass of the atom (elementary cell). We are interested in stationary oscillations, which means that the solution is obtained in the form

$$\mathbf{u}(\mathbf{n}, t) = \mathbf{u}(\mathbf{n})\exp(-\mathrm{i}\omega t) \tag{10}$$

Hence, introducing Equation 10 into Equation 9 we obtain

$$\omega^2 m\mathbf{u}(\mathbf{n}) = \sum_{\mathbf{n}'} \bar{\bar{A}}(\mathbf{n} - \mathbf{n}')\mathbf{u}(\mathbf{n}') \tag{11}$$

Now, we assume that there are N_p pores in the material (system) centres (centres of mass) which are at positions $\mathbf{R}_1, \mathbf{R}_2, \ldots \mathbf{R}_n$. These pores cause the following perturbations to Equation 11:

(i) they remove material (atoms) from the volume of pores and

(ii) they change the dynamic matrix in their environment. Because of this fact, new terms are added to Equation 11 to give

$$\begin{aligned}\omega^2\mathbf{u}(\mathbf{n}) - \frac{1}{m}\sum_{\mathbf{n}'} \bar{\bar{A}}(\mathbf{n} - \mathbf{n}')\omega(\mathbf{n}') &= \omega^2 \sum_{\mathbf{R}_j}\sum_{\xi} \delta(\mathbf{n} - \mathbf{R}_j - \xi)\mathbf{u}(\mathbf{n}) \\ &\quad + \sum_{\mathbf{R}_j}\sum_{\mathbf{n}'} \bar{\bar{U}}(\mathbf{n} - \mathbf{R}_j; \mathbf{n}' - \mathbf{R}_j)\mathbf{u}(\mathbf{n}')\end{aligned} \tag{12}$$

The first term on the right describes the influence of removing atoms from the pore volume (ξ = position of lattice sites in the pore with regard to its centre of mass), while the second term describes the correction of the dynamic matrix. Green's function for Equation 12 then obeys the equation

$$\begin{aligned}&\omega^2 \bar{\bar{G}}(\mathbf{n}, \mathbf{n}') - \frac{1}{m}\sum_{\mathbf{n}''} \bar{\bar{A}}(\mathbf{n} - \mathbf{n}')\bar{\bar{G}}(\mathbf{n}'', \mathbf{n}') \\ &= \delta(\mathbf{n} - \mathbf{n}')\bar{\bar{1}} + \omega^2 \sum_{\mathbf{R}_j}\sum_{\xi} \delta(\mathbf{n} - \mathbf{R}_j - \xi)\bar{\bar{G}}(\mathbf{n}, \mathbf{n}') \\ &+ \sum_{\mathbf{R}_j}\sum_{\mathbf{n}''} \bar{\bar{\mathrm{U}}}(\mathbf{n} - \mathbf{R}_j; \mathbf{n}'' - \mathbf{R}_j)\bar{\bar{G}}(\mathbf{n}'', \mathbf{n}')\end{aligned} \tag{13}$$

Writing $\bar{\bar{G}}(\mathbf{n}, \mathbf{n}')$ as

$$\bar{\bar{G}}(\mathbf{n}, \mathbf{n}') = \bar{\bar{G}}_0(\mathbf{n}, \mathbf{n}') + \bar{\bar{G}}_z(\mathbf{n}, \mathbf{n}') \tag{14}$$

where $\bar{\bar{G}}_0(\mathbf{n}, \mathbf{n}')$ is Green's function for an ideal crystal (without pores), we obtain from Equation 13

$$\begin{aligned}\bar{\bar{G}}(\mathbf{n}, \mathbf{n}') = \bar{\bar{G}}_0(\mathbf{n} - \mathbf{n}') &+ \sum_{\mathbf{n}''}\sum_{\mathbf{n}'''} \bar{\bar{G}}(\mathbf{n}, \mathbf{n}'') \\ &\times [\omega^2 \sum_{\mathbf{R}_j}\sum_{\xi} \delta(\mathbf{n} - \mathbf{R}_j - \xi)\bar{\bar{G}}_0(\mathbf{n}''' - \mathbf{n}') \\ &\times \delta(\mathbf{n}'' - \mathbf{n}''') + \sum_{\mathbf{R}_j} \bar{\bar{U}}(\mathbf{n}'' - \mathbf{R}_j; \mathbf{n}''' - \mathbf{R}_j) \\ &\times \bar{\bar{G}}_0(\mathbf{n}''' - \mathbf{n}')]\end{aligned} \tag{15}$$

It is convenient to work in the $\mathbf{k}$ space, so we will work with Fourier representation of Green's function. Using the relations

$$X(\mathbf{n}, \mathbf{n}') = \frac{1}{N^2}\sum_{\mathbf{q}}\sum_{\mathbf{q}'} X(\mathbf{q}, \mathbf{q}')\mathrm{e}^{i\mathbf{q}\cdot\mathbf{n}}\mathrm{e}^{-i\mathbf{q}'\cdot\mathbf{n}'}$$

$$\sum_{\mathbf{n}} \mathrm{e}^{i\mathbf{q}\cdot\mathbf{n}} = N\delta(\mathbf{q})$$

where N is the number of elementary cells in the whole crystal and $\delta(\mathbf{q})$ is the Kronecker delta, we have

$$\begin{aligned}\bar{\bar{G}}(\mathbf{k}, \mathbf{k}') = N\bar{\bar{G}}_0(\mathbf{k})\delta(\mathbf{k} - \mathbf{k}') &+ \sum_{\mathbf{q}} \bar{\bar{G}}(\mathbf{k}, \mathbf{k}' + \mathbf{q}) \\ &\times \bar{\bar{G}}_0(\mathbf{k}')\omega^2 \sum_{j}\sum_{\xi} \frac{\exp[-\mathrm{i}\mathbf{q}\cdot(\mathbf{R}_j + \xi)]}{N} \\ &+ \sum_{\mathbf{q}} \bar{\bar{G}}(\mathbf{k}, \mathbf{k}' + \mathbf{q})\bar{\bar{U}}(\mathbf{k}' + \mathbf{q}) \\ &\times \sum_{j} \frac{\exp[-\mathrm{i}\mathbf{q}\cdot(\mathbf{R}_j + \xi)]}{N}\bar{\bar{G}}_0(\mathbf{k}')\end{aligned} \tag{16}$$

$\bar{\bar{G}}(\mathbf{k}, \mathbf{k}')$ is a function of the concrete positions of centres (centres of mass) of pores $\mathbf{R}_1, \mathbf{R}_2, \ldots, \mathbf{R}_{N_\mathrm{p}}$. We are interested in the average Green's function $\langle\bar{\bar{G}}(\mathbf{k}, \mathbf{k}')\rangle$ [9]

$$\langle\bar{\bar{G}}(\mathbf{k}, \mathbf{k}')\rangle = \frac{\sum_{\{\mathbf{R}_j\}} \bar{\bar{G}}(\mathbf{k}, \mathbf{k}', \{\mathbf{R}_j\})}{N\{\mathbf{R}_j\}}$$

where summation over all possible configurations of centres of mass pores is implied and $N\{\mathbf{R}_j\}$ is the number of these configurations.

If we assume that each pore is uncorrelated with any other, then

$$\langle\bar{\bar{G}}(\mathbf{k}, \mathbf{k}')\rangle = \sum_{\mathbf{R}_1}, \sum_{\mathbf{R}_2}, \ldots, \sum_{\mathbf{R}_{N_\mathrm{p}}} \frac{1}{(N)^{N_\mathrm{p}}}\bar{\bar{G}}(\mathbf{k}, \mathbf{k}', \mathbf{R}_1, \ldots, \mathbf{R}_{N\mathrm{p}}) \tag{17}$$

where summation is now implied for each $\mathbf{R}_i$ over all lattice positions.

We can see that in the iteration series for $\bar{\bar{G}}(\mathbf{k}, \mathbf{k}')$ created by Equation 16, we must average the products of type

$$\left(\frac{\sum_j \mathrm{e}^{\mathbf{i}\mathbf{q}\cdot\mathbf{R}_j}}{N}\right)\left(\frac{\sum_k \mathrm{e}^{\mathbf{i}\mathbf{q}'\cdot\mathbf{R}_k}}{N}\right)\cdots\left(\frac{\sum_p \mathrm{e}^{\mathbf{i}\mathbf{q}''\cdot\mathbf{R}_p}}{N}\right) \tag{18}$$

where the number of these terms is M. The average value of the term $\mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_j}$ is $\langle\mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_j}\rangle = \delta(\mathbf{q}, 0)$.

The most important contribution to the sum of averaged products of terms $\mathrm{e}^{i\mathbf{q}\cdot\mathbf{R}_j}$ in Equation 18 is provided by the term in which every $\mathbf{R}_j$ is different (or where are the most different $\mathbf{R}_j$, in the case that $M > N_p$). We suppose that N_p is large enough but $N_p \ll N$. The contribution is proportional to $(1/N)^M$, so we are interested only in the members with small M ($M < N_p$) which give the largest contributions.

For large N (i.e. a large sample) we can then write

$$\begin{aligned}&\left\langle\left(\frac{\sum_j \mathrm{e}^{\mathbf{i}\mathbf{q}\cdot\mathbf{R}_j}}{N}\right)\left(\frac{\sum_k \mathrm{e}^{\mathbf{i}\mathbf{q}'\cdot\mathbf{R}_k}}{N}\right)\cdots\left(\frac{\sum_p \mathrm{e}^{\mathbf{i}\mathbf{q}''\cdot\mathbf{R}_j}}{N}\right)\right\rangle \\ &\approx \frac{N_\mathrm{p}}{N}\delta(\mathbf{q})\frac{N_\mathrm{p}}{N}\delta(\mathbf{q}')\ldots\frac{N_\mathrm{p}}{N}\delta(\mathbf{q}'') \\ &= P'\delta(\mathbf{q})P'\delta(\mathbf{q}')\ldots P'\delta(\mathbf{q}'')\end{aligned} \tag{19}$$

After averaging, we can again sum iteration series and we obtain a closed equation for the averaged Green's function $\langle \bar{\bar{G}}(\mathbf{k}, \mathbf{k}') \rangle$:

$$\langle \bar{\bar{G}}(\mathbf{k}, \mathbf{k}') \rangle = N\delta(\mathbf{k} - \mathbf{k}') \times \left[\bar{\bar{G}}_0^{-1} - \left(\omega^2 P + \frac{\bar{\bar{U}}(\mathbf{k}, \mathbf{k}')}{N_\xi} P \right) \right]^{-1} \tag{20}$$

Here P is the conventional porosity.

Taking into account the symmetry of $\bar{\bar{U}}(\mathbf{n}, \mathbf{n}')$ and the fact that we are interested in the long-wave ($\mathbf{k} \to 0$) we obtain for $\bar{\bar{U}}(\mathbf{k}, \mathbf{k})$ the expression

$$\frac{\bar{\bar{U}}(\mathbf{k}, \mathbf{k})}{N_\xi} = -\frac{1}{2}\left(\sum_n \sum_{n'} \frac{\bar{\bar{U}}(\mathbf{n}, \mathbf{n}')}{N_\xi} [\mathbf{e}_k \cdot (\mathbf{n} - \mathbf{n}')]^2 \right) k^2$$
$$= + \bar{\bar{\alpha}}\{\mathbf{e}_k\} k^2$$

where $\mathbf{e}_k$ is a unit vector in the direction of wave propagation and N_ξ is the number of elementary cells in one pore. We suppose that the size of the pore is much less than the wavelength of sound ($\mathbf{k} \to 0$), and that the polarization of wave motion is unchanged in pore's environment in comparison with the matrix without pores. We can then use the scalar model. Hence Equation 20 is reduced to

$$\langle \bar{\bar{G}}(\mathbf{k}, \mathbf{k}') \rangle = \frac{N\delta(\mathbf{k} - \mathbf{k}')}{\omega^2 - c_i(0)^2 k^2 - \omega^2 P - \alpha_i P k^2} \tag{21}$$

where the subscript i specifies the polarization of sound waves (longitudinal, transversal) and $c_i(0)$ is the velocity of this wave in the environment without pores. Finally, the poles of Equation 21 now correspond to the dispersion relation

$$\omega = \left(\frac{1 + a_i P}{1 - P} \right)^{1/2} c_i(0) k$$

where $a_i = \alpha_i / c_i^2(0)$. For sound velocity in the porous environment it follows ($i = \mathrm{L}, \mathrm{T}$) that

$$c_i(P) = \left(\frac{1 + a_i P}{1 - P} \right)^{1/2} c_i(0) \tag{22}$$

For concrete calculation of $c_i(P)$ it is important to know the perturbation of the dynamic matrix $\bar{\bar{U}}(\mathbf{n}, \mathbf{n}')$ caused by the pore with its centre (centre of mass) at $\mathbf{R} = 0$. Here the parameter a_i depends on the shape and size of pores, but it is independent of their number. If we are interested only in the relations between the sound velocity and porosity we can consider a_i as a constant. If we know the sound velocity we can obtain the elastic moduli of material [10]

$$\sigma = \frac{(a_\mathrm{L} - a_\mathrm{T})P + \sigma_0[1 + (2a_\mathrm{T} - a_\mathrm{L})P]}{1 + (2a_\mathrm{L} - a_\mathrm{T})P + 2\sigma_0(a_\mathrm{T} - a_\mathrm{L})P} \tag{23}$$

$$E = E_0 \frac{(1 + a_\mathrm{T} P)\left(1 + \dfrac{3a_\mathrm{L}(1 - \sigma_0) - 2a_\mathrm{T}(1 - 2\sigma_0)}{1 + \sigma_0} P \right)}{1 + [2a_\mathrm{L}(1 - \sigma_0) - a_\mathrm{T}(1 - 2\sigma_0)]P}$$

where E_0 and σ_0 (E and σ) are Young's modulus and Poisson's ratio at $P = 0$ and some volume fraction of porosity P, respectively. Equation 24 can be written in the simple form of Equation 7.

3. Discussion and conclusion

We have derived here the relation between Young's modulus and the porosity of a material (E–P relation) using a theoretical approach. Our result is based on a microscopical model and on corresponding mathematical methods. In contrast to the majority of existing more or less theoretical derivations of the E–P relation, which are based on defining an expression for Young's modulus (relation between stress and deformation), a dependence of sound velocity on Young's modulus is used in our case.

The sound velocity as a function of porosity and other model parameters was obtained from the dispersion relation (dependence of frequency of sound waves on their wave vectors) in the long-wave region, and the required dispersion relation was provided by the poles of Green's function for a corresponding microscopical equation of motion for small deviations from equilibrium positions of material structural units. We assumed that pores are mutually uncorrelated, that they have the same size and form and that they are randomly distributed in a homogeneous matrix. A change of porosity is assumed to be a change of the number of pores, while their form and size remain unchanged.

The resulting relation between Young's modulus and porosity (Equation 7) is

$$E = E_0 \frac{1 + aP + bP^2}{1 + cP}$$

where a, b and c are quantities independent of porosity, but dependent not only on material parameters but also on the size and form of the average pore.

Our E–P relation (Equation 7) becomes identical with some relations proposed by other authors (see Section 1) for special choices of the parameters a, b and c; but in general (if no constraints are set on a, b, c), our result is different.

To obtain a first checking of the applicability of our result, we compare Equation 7 with the exact Wang's curve [2] obtained numerically for his theoretical model. In Wang's model pores are identical and they are arranged in a simple cubic lattice. As the porosity increases, the size of pores increases too (their number remains unchanged). This model is also able to describe the transition from isolated pores to connected ones.

The behaviour of pores with change of porosity is different in Wang's model and in our model, but nevertheless our curve fits Wang's result very well over nearly the whole porosity range (ever better than Wang's Equation 2a with three parameters) (Fig.1, curve 1). Deviations occur near the top of the porosity interval, e.g. for porosity corresponding to interconnected pores.

However, comparison with experimental results is important. Therefore, the aim of our further investigation will be to compare the theoretical E–P dependence (Equation 7) with experimental data for systems obeying convenient conditions for the behaviour of porosity (changes of the number of pores, without change of their shape and size).

From the theoretical point of view, further investigation is needed for calculation of concrete value of the quantities a, b, c in Equation 7 for a given shape and size of average pore, or for some distribution of pore size and shape.

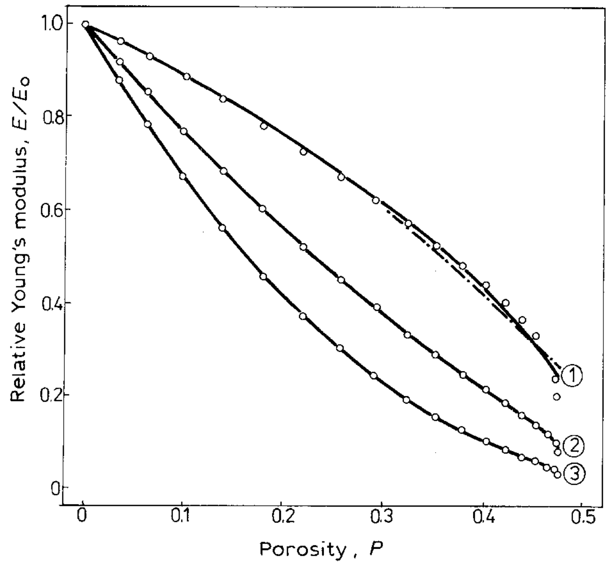

Figure 1 Young's modulus as a function of porosity: (1) ideal case, (2) non-ideal case (shear effect), (3) non-ideal case (combined shear and hinge effect). (○) Exact solution obtained numerically from Wang's theory; solid curves obtained from Equation 7 with three constants a, b, c; and for comparison the broken curve obtained from Equation 2b with three parameters b, c, d.

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Received 27 March 1992
and accepted 24 February 1993