

A New Approach for the Young's Modulus–Porosity Correlation of Ceramic Materials

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Abstract: A new approach for determining the Young's modulus of porous ceramics is presented. The approach is based on a previous equation derived for two-phase composites on the basis of a microstructural description that uses the contiguity and continuous volume as topological parameters. The derived equation shows very good agreement with experimental data drawn from the literature on porous glasses and ceramics for a wide range of porosity volume fractions. As a result of the limited geometrical and topological information on the porosity structure supplied in the original studies, however, the full potential of the model for Young's modulus prediction could not be demonstrated. For a rigorous verification of the equation, therefore, experimental data on materials with well-described porosity structures is required. © 1997 Elsevier Science Limited and Techna S.r.l. All rights reserved

1 INTRODUCTION

There has been much research effort during the last 30 years dedicated to the understanding of the porosity dependence of the elastic constants of ceramic materials. Theoretical and experimental work has been conducted and numerous relationships have been put forward.^{1–14} Most relationships give the variation of the elastic constants, usually Young's modulus, bulk and shear modulus or Poisson's ratio, in terms of the porosity content or volume fraction of pores (P). In Table 1 the most commonly used equations for the dependence of the Young's modulus on porosity are presented. Examining the equations in Table 1, it becomes clear that the influence of the porosity structure and arrangement in the material, for example pore shape, orientation, distribution, homogeneity, etc., has been neglected in almost all approaches. In recent communications, however, the need for taking into account such parameters describing the porosity

structure has been emphasized.^{15,16} In addition, the equations that did consider porosity structure parameters such as the shape and orientation of the pores, for example the equations by Janowski and Rossi¹⁰ [eqn (5)] and by Boccaccini *et al.*⁵ [eqn (6)], are based in a spheroidal model to describe the porosity shape and therefore they are strictly valid only at low volume fractions of porosity or for materials containing solely closed isolated pores.

For high volume fractions of the second phase in a two-phase microstructure not only geometrical but also topological parameters must be considered to characterize the microstructure, as pointed out recently by Fan and co-workers.^{17,18} One useful approach for the topological characterisation of such microstructures has been developed¹⁷ based on the concept of contiguity, first proposed by Gurland,¹⁹ which describes the degree of particle contact of one phase in a two-phase microstructure. It has been found that many physical and mechanical properties, such as electrical resistivity,¹⁸ fracture toughness,²⁰ Vickers hardness²¹ and tensile properties²² can be related closely to this contiguity parameter. As far as the authors are

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aware, however, topological parameters based on the concept of contiguity and continuous volume have not been used specifically to determine the elastic constants of porous ceramics.

In the present study a recently derived equation for calculating the effective properties of multi-phase composite materials, incorporating topological parameters in the calculations, has been adapted to predict the Young's modulus of porous materials. The equation is verified by comparison with the predictions by other approaches and by comparing calculated values with experimental data on porous ceramics and glasses drawn from the literature.

2 MICROSTRUCTURAL CHARACTERIZATION

The quantitative characterization of a two-phase microstructure involves analysis of both geometrical and topological quantities. The geometrical characterisation is well established and involves the use of stereological parameters, such as particle (inclusion) size, particle axial ratio and orientation, volume fraction, particle spacing, etc., as reviewed by Underwood.²³ The use of these parameters in microstructure-property correlations has been the subject of much research in the past.^{24,25} However, a major limitation of using only geometrical parameters for microstructural description arises;

namely, the microstructure-property equations are valid only for matrix-type microstructures, i.e. microstructures in which the second phase is discontinuously embedded in a continuous matrix.²⁵ For high concentrations of the second phase, or when both phases become continuous (at least partially), topological parameters are necessary for the proper characterisation of the microstructure,¹⁷ the topological characterization of a two-phase microstructure being inherently more difficult than the geometrical characterization, as demonstrated recently.^{17,18} A number of topological parameters, such as separation, separated volume, degree of contiguity and degree of separation, based on the topological characterization of contiguity and continuous volume in two-phase microstructures, have been proposed recently by Fan and co-workers.^{17,18} These topological parameters can be either measured experimentally or calculated theoretically under certain simplified assumptions about the real microstructure.¹⁷ The combination of such topological parameters can offer an effective description of the phase distribution in any two-phase microstructure.

According to the proposed topological transformation, a two-phase microstructure (denoted as α - β hereafter) with any grain size, grain shape and phase distribution, as illustrated schematically in Fig. 1 (a), can be transformed topologically into a body with three parallel aligned microstructural elements, as illustrated schematically in Fig. 1 (b). Element I (EI) consists of continuous α -phase with a volume fraction of $f_{\alpha c}$ (the continuous volume of the α -phase); Element II (EII) consists of the continuous β -phase with a volume fraction $f_{\beta c}$ (the continuous volume of the β -phase); Element III (EIII) consists of the long-range α - β chains. Therefore, there are only phase boundaries in EIII. The volume fraction of EIII is defined by the degree of separation, F_s . The volume fractions of α -phase and β -phase in EIII body, $f_{\alpha III}$ and $f_{\beta III}$, respectively, can be calculated by the following equations:

$$f_{\alpha III} = \frac{f_{\alpha} - f_{\alpha c}}{F_s} \quad (10)$$

$$f_{\beta III} = 1 - f_{\alpha III} \quad (11)$$

where f_{α} is the total volume fraction of the α -phase.

As has been discussed in detail elsewhere,¹⁷ microstructures A and B (see Fig. 1) are mechanically equivalent along the aligned direction of microstructure B. Thus, the determination of the effective mechanical properties of a complicated two-phase microstructure, such as that shown in Fig. 1 (a), can be replaced by an analysis of the

Table 1. Summary of Young's modulus-porosity correlations

Equation		Reference
$E = E_0 e^{-bP} P \leq 0.5$	(1)	1,9
$E = E_0 e^{-b'(1-P)} P \geq 0.5$	(2)	1
$E = E_0 \frac{(1-P)}{1+kP}$	(3)	14
$k = \frac{(1+\nu_0)(13-15\nu_0)}{2(7-5\nu_0)}$		
$E = E_0 (1 - aP^{2/3})$	(4)	12
$E = E_0 (1 - mP) P \leq 0.5$	(5)	10
$m = f(z/x)$		
$E = E_0 (1 - P^{2/3})^{1.21S} P \leq 0.5$	(6)	5
$s = g(z/x)$		
$E = E_0 \left(1 - \frac{uP}{1+ u-1 P}\right)$	(7)	8
$E = E_0 \frac{(1-P)^2}{(1+k_E P)}$	(8)	7
$k_E = 2 - 3\nu_0$		
$E = E_0 (1 - pP)^q$	(9)	2,3,6

(E : Young's modulus of the porous material, E_0 : Young's modulus of the pore-free matrix, P : volume fraction of porosity, ν_0 : Poisson's ratio of the pore-free matrix; b, b', k, u, p, q : are fitting parameters without a clear correlation to the porosity structure, m and s are parameters related to the shape and orientation of spheroidal pores, z/x is the axial ratio of spheroidal pores)

simpler but equivalent microstructure with three well-defined microstructural elements, Fig. 1 (b).

In relation to the proposed microstructural characterization the following aspects must be emphasized:

1. The topological parameters developed by Fan *et al.*¹⁷ were derived by means of statistics and probability theory, and hence they have to be treated in terms of average quantities. They reflect implicitly the change in size, shape, orientation and distribution of the second phase.
2. In the general case, the topological parameters have to be measured experimentally by applying standard metallographic methods and stereological relationships, as described by Underwood.²³ However, under the assumption of equiaxed particle shape and random distribution, they can also be calculated, for example by means of the following relationships:

$$f_{\alpha c} = \frac{f_{\alpha}^2 d_{\beta}}{f_{\beta} d_{\alpha} + f_{\alpha} d_{\beta}} = \frac{f_{\alpha}^2 R}{f_{\beta} + f_{\alpha} R} \quad (12)$$

$$f_{\beta c} = \frac{f_{\beta}^2 d_{\alpha}}{f_{\beta} d_{\alpha} + f_{\alpha} d_{\beta}} = \frac{f_{\beta}^2}{f_{\beta} + f_{\alpha} R} \quad (13)$$

$$F_s = 1 - f_{\alpha c} - f_{\beta c} \quad (14)$$

where R is the particulate size ratio defined as $R = d_{\beta}/d_{\alpha}$. d_{α} and d_{β} are the grain sizes of the phases α and β , respectively, and they can be determined by the mean intercept length of each phase.¹⁷

3. In the cases where experimentally measured topological parameters are not available, the following power law can be used to approximate the continuous volumes:¹⁸

$$f_{\alpha c} = f_{\alpha}^v \quad (15)$$

$$f_{\beta c} = f_{\beta}^w \quad (16)$$

For example, the experimentally measured continuous volume of the WC phase in Co-WC composites²¹ can be adequately represented by $w = 4$.²⁶

4. Topological parameters are directional.¹⁷ The measurement of all the topological parameters of a given two-phase microstructure with specific microstructural features must be made along the direction of interest (e.g. field or stress direction).

3 THE YOUNG'S MODULUS-POROSITY EQUATION

In previous work, Fan and co-workers¹⁸ have derived an equation to predict the effective field properties of two-phase composite as a function of the properties of the constituent phases and the topological parameters of the composite described in the previous section ($f_{\alpha c}$, $f_{\beta c}$, F_s , $f_{\alpha III}$, $f_{\beta III}$). When applied for the effective Young's modulus (E_{eff}), the equation takes the form:

$$E_{eff} = E_{\alpha} f_{\alpha c} + E_{\beta} f_{\beta c} + \frac{E_{\alpha} E_{\beta} F_s}{E_{\beta} f_{\alpha III} + E_{\alpha} f_{\beta III}} \quad (17)$$

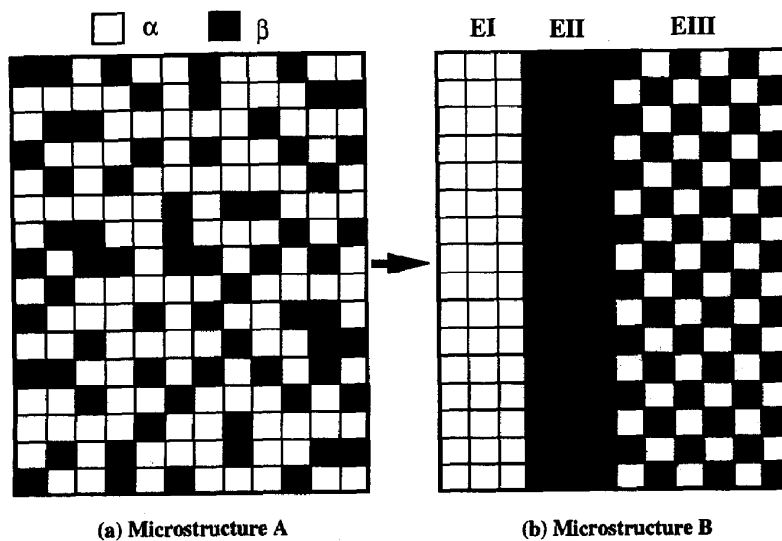


Fig. 1. Schematic illustration of the topological transformation from microstructure A to microstructure B after Fan *et al.*¹⁷ It should be emphasized that this graph is just a schematic illustration of the topological transformation and it does not represent any quantitative information such as volume fraction or particle size and shape.

where E_α and E_β are the Young's moduli of the constituent phases. Equation (17) is applicable to a two-phase composite with any combination of volume fraction, particle size, shape and distribution of the second phase. It has been shown that the predictions by eqn (17) are in good agreement with experimental data for different composites.²⁶ A porous material can be treated as a special case of a two-phase composite in which the Young's modulus of one phase (pore) is equal to zero, i.e. $E_\beta = 0$. Therefore, according to eqn (17), the effective Young's modulus of porous materials (E_P) will take the following form:

$$E_P = E_M f_{ac} \quad (18)$$

where E_M represents the Young's modulus of the pore-free matrix. It is interesting to see from eqn (18) that the normalized Young's modulus of a porous material (E_P/E_M) is simply equal to the continuous volume of the matrix (f_{ac}). By assuming that the pore phase has an equiaxed shape and random distribution inside the matrix phase, the continuous volume of the matrix phase can be calculated from eqn (12), if the particle size ratio R is known. Noting that f_α can be written as $f_\alpha = (1-P)$, and $f_\beta = P$, with P : volume fraction of porosity, then eqn (12) can be rewritten for the porous case as:

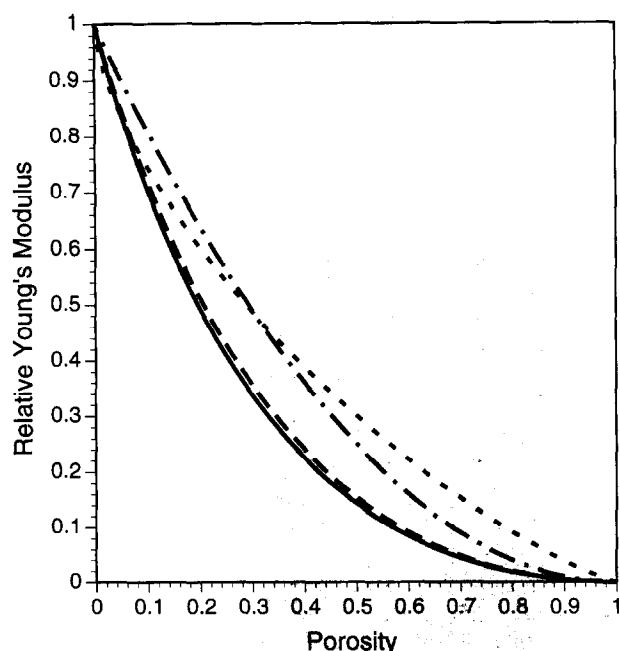


Fig. 2. Young's modulus–porosity relationship as predicted by several theoretical approaches: (—) R.K. model [eqn (8)]⁷ for $v_0 = 0.25$, (---) Boccaccini *et al.* model [eqn (6) with $S = 1$]⁵ for spherical porosity, (—) this model [eqns (18) and (19)] with $R = 1.0$, (---) this model [eqns (18) and (19)] with $R = 0.4$.

$$f_{ac} = \frac{(1-P)^2 R}{P + (1-P)R} \quad (19)$$

The utility of eqns (18) and (19) to be used for the prediction of the Young's modulus of porous materials is because only the parameter R , besides the pore volume fraction, which can be obtained by quantitative microstructural analysis, remains influencing the effective property. Thus, no fitting parameters are necessary and the equations become predictive having a great practical significance.

Alternatively, the power law given by eqn (15) can be used if measured topological parameters are not available. In such case the exponent v will act as a fitting parameter unless a direct correlation between v and the porosity structure can be found.²⁶

4 EVALUATION OF THE MODEL

4.1 Comparison with other equations

Figure 2 shows the theoretical variation of the effective Young's modulus as a function of porosity as predicted by eqns (18) and (19). $R = 0.4$ and $R = 1.0$ were used in the calculations. Also shown

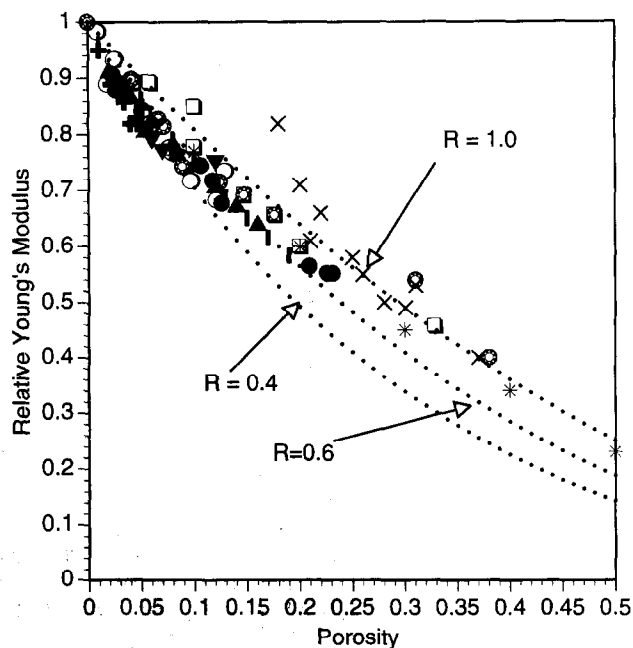


Fig. 3. A comparison of the theoretically predicted normalized effective Young's modulus as a function of porosity (E_P/E_M) with experimental results from the literature for different porous ceramics: (●) Gd_2O_3 ,¹¹ (▼) $CaTiO_3$,²⁹ (*) Al_2O_3 ,²⁸ (○) ZrO_2 ,³⁴ (■) Dy_2O_3 ,³³ (X) Si_3N_4 ,³¹ (⊕) Hf_2O_7 ,³⁴ (+) mullite,³⁰ (□) UO_2 ,³² (▲) Y_2O_3 .³³ The theoretical curves were obtained from eqns (18) and (19), using the shown values for R .

in the figure are the curves calculated using the model of Boccaccini *et al.*⁵ for spherical pores [eqn (6) with $S=1$] and the model of Ramakrishnan–Arunachalam (R–K)⁷ for a material with Poisson's ratio $\nu_0=0.25$ [eqn (8)]. It is interesting to note that the present equation closely matches the model of R–K when $R\approx 0.4$ in the entire porosity range. Using $R=1$ results in higher values for the Young's modulus and the calculated values are closer to those calculated for spherical porosity.⁵

If alternatively one applies the power law [eqn (15)] to obtain f_{∞} , eqn (18) reduces to the equation proposed in the literature independently by Phani and co-workers^{2,3} and by Wagh *et al.*⁶ [eqn (9) in Table 1]. This power equation has also been suggested for the Young's modulus of foam-like materials.²⁷

4.2 Comparison with experimental data on porous ceramics and glasses

Most work in the literature dealing with the elastic constants of porous ceramics and glasses report very little information about the porosity structure and even less studies can be found that report quantitative parameters characterising the porosity structure. Thus the present verification of the model will have to be limited to a study of the fitting capability of the equations to the available experimental data and no attempt will be made to quantitatively assess the effect of the porosity

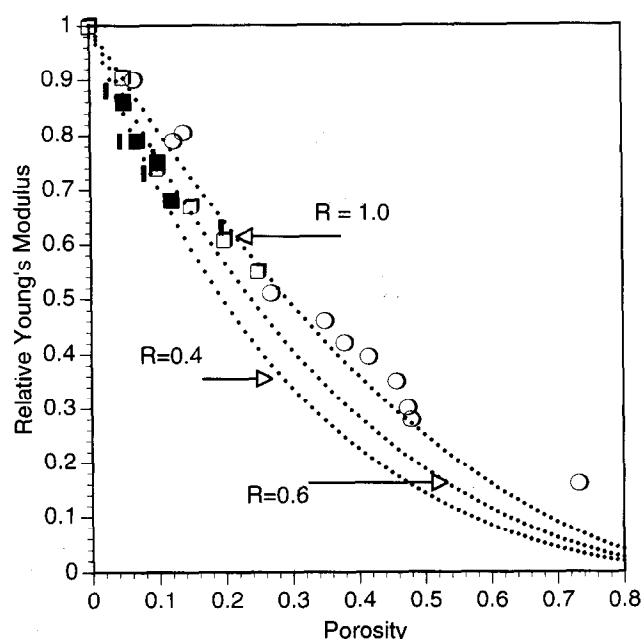


Fig. 4. A comparison of the theoretically predicted normalized effective Young's modulus as a function of porosity (E_p/E_M) with experimental results from the literature for different porous glasses: (■) borosilicate glass,²⁹ (●) DURAN borosilicate glass,³⁵ (○) P-311 glass,⁴ (□) D-glass.³⁶ The theoretical curves were obtained from eqns (18) and (19), using the shown values for R .

parameters on the Young's modulus–porosity relationship. Figure 3 shows, for example, the comparison between calculated and experimental values for a variety of porous ceramics with $P\leq 0.5$.^{11,28–34} The curves were obtained assuming equiaxed pore morphology and random porosity distribution, i.e. using eqns (18) and (19) with different values of R . It seems that $R=1$ overestimates the Young's modulus at low porosity content ($P\leq 0.25$). However, for intermediate porosity levels ($0.25\leq P\leq 0.5$) using $R=1$ seems to better predict the experimental data than when lower values of R were used. On the contrary, for the low-porosity range, R values between 0.4 and 0.6 show better correlation with the experimental data. Similar conclusions can be drawn from the comparison between theory and experiment for porous glasses,^{4,29,35,36} as shown in Fig. 4. Extensive data on the Young's modulus of porous Si_3N_4 ³⁷ are considered in Fig. 5. Here eqn (18), with eqn (19) for obtaining f_{∞} , correlates with the trend of the experimental data fairly well using $R=1.0$. Finally, the high-porosity range ($P>0.6$) is investigated in Fig. 6. Data for gypsum,³ cellular alumina,³⁸ silica gel³⁹ and a polymere foam²⁷ are considered. Using eqn (19) with $R=0.4$ provides an excellent fit of the data.

In summary, observation of Figs 3–6 reveals that the present approach fits the experimental values for porous ceramics and glasses in the entire porosity range fairly well, if the appropriate parameter R is used in the calculations. However, no clear relationship between R and the amount of porosity

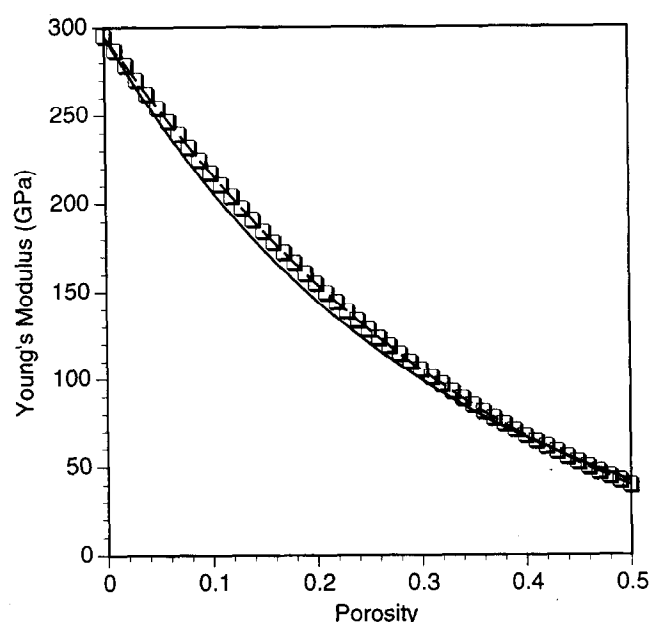


Fig. 5. Comparison of the theoretically predicted effective Young's modulus of porous Si_3N_4 with the trend of experimental data,³⁷ (□□□) experimental trend,³⁷ (—) theoretical curve obtained from eqns (18) and (19), using $R=1.0$.

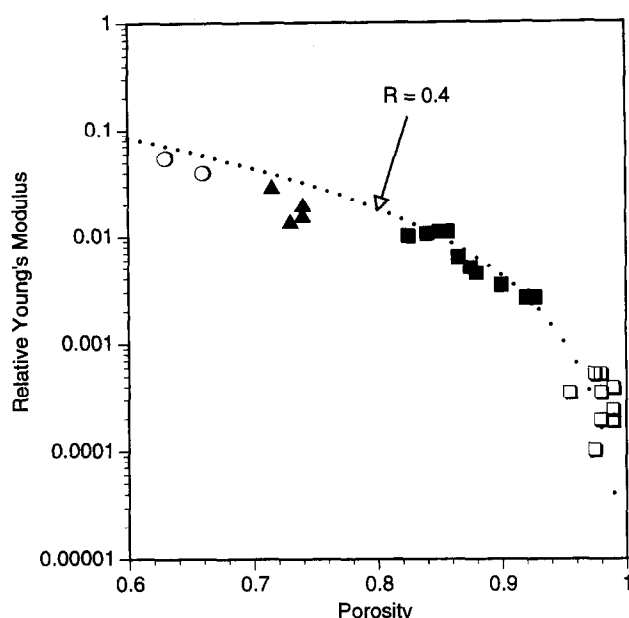


Fig. 6. A comparison of the theoretically predicted normalized effective Young's modulus as a function of porosity (E_p/E_M) with experimental results from the literature for different materials with very high porosity: (■) cellular alumina,³⁸ (○) gypsum,³ (▲) silica gel,³⁹ (□) polymere foam.²⁷ The theoretical curve was obtained from eqns (18) and (19), using $R=0.4$.

could be drawn, indicating that R should be determined in each case from metallographic measurements. This highlights, moreover, the necessity of considering a structure-related parameter besides the porosity volume fraction for a proper description of the experimental trends, as noted in the introduction. In the present case, because of the limited geometrical and topological information on the porosity structure supplied in the original studies, the full potential of the present approach for Young's modulus prediction could not be demonstrated with the available experimental data.

5 CONCLUSIONS

A new approach has been presented for determining the Young's modulus of porous materials. The approach is based on a previous equation derived for two-phase composites on the basis of a microstructural description using the contiguity and continuous volume as topological parameters. The derived equation shows very good agreement with experimental data on porous glasses and ceramics for a wide range of porosity volume fractions. For a rigorous verification of the equation, however, experimental data on materials with well-described porosity structures is required. As there is limited experimental data on such well-characterized porous materials, the necessity of further experimental work on this subject must be emphasized.

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