NUMERICAL TESTING ON THE EFFECT OF PHASE CONTIGUITY IN BI-CONTINUOUS COMPOSITES

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ABSTRACT

Numerical modelling of two-phase composites with bi-continuous microstructures is presented in this paper. The mechanical and thermal loading behaviours were simulated by a finite element technique and the optimal property of the microstructure was explained using the concept of phase contiguity. The results show that the elastic modulus was not only a function of volume fraction, but also increased with the increasing contiguity of the reinforcing phase. Further extending to the multifunctional application, phase contiguity plays an important role on the properties combinations in the bi-continuous composites. These results are of significant importance in the design and optimization of the microstructures of bi-continuous composite especially when a property combination is sought after.

1. INTRODUCTION

In a bi-continuous composite, both phases are 3D continuous and interpenetrating throughout the microstructure. They are also called co-continuous, interpenetrating or multi-continuous composites and have many advantages over the conventional discontinuous composites. For example, the properties of bi-continuous composites such as elastic modulus [1, 2] may increase significantly towards the optimal Hashin-Shtrikeman (HS) upper bound [3] or even higher when the composites are created from stiff and compliant materials and the stiff phase becomes continuous. Molchanov et al. [4] demonstrated the benefit of bi-continuous composites—a considerable increase in strength of a bi-continuous alumina/aluminium composite at elevated temperatures, due to the enhanced mismatch in the mechanical properties of the two phases. Daehn et al. [5] found in their experimental observation that there were no significant microcracking in a bi-continuous composite (70% alumina and 30% aluminium) with higher volume fraction of stiff phase and no measurable degradation in the elastic modulus after apparent plastic flow. The phenomenon suggests that the elastic deformation in the ceramic was accommodated by plastic deformation in the metal phase.

These results indicate that such an attribute of bi-continuous composites that the phases can mechanically constrain one another offers opportunities for optimization of the phases/structures in terms of the volume fraction and phase connectivity dependence of transport properties. In other words, it is expected that mutual interconnectivity allows each phase to contribute its desirable properties in an optimal way, so that the composites may provide an optimal combination of properties for a given combination of phases. Thus, the use of numerical techniques to automatically optimise or search for microstructures with prescribed properties is of much interest and numerous efforts [5-10] have been made to achieve this. Although more complex models and the better

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understanding of the link between the microstructure and the macroscopic properties are still needed, these efforts provided examples to tailor the microstructure with desirable properties [7, 10]. It has been shown in principle that a microstructure with an interface as a triply periodic minimal surfaces is optimal single-scale bi-continuous structure [11, 12], the interface is a surface that is locally area minimizing and necessarily has zero mean curvature. Such a microstructure at the volume fraction of each phase of 50% is extremal not only for effective thermal and electrical conductivities [7, 13,14] but also for the effective bulk modulus and thermal conductivity [10]. In these research works, however, a geometrical parameter— the overall volume fraction was used to characterize the material and hence lack of the microstructural information. The present work appears to be the first attempt to try to relate the properties of bi-continuous composites to an important topological parameter— phase contiguity, and to provide useful insight into this specific group of composites.

The concept of contiguity was firstly proposed by Gurland [15] for the description of the extent of particle contact in dual phase structures. In other words, it can be used to quantify the connected nature of the phases in a composite. This concept has been accepted and used to characterise composite materials by some researchers [2, 16]. Gurland [15] defined contiguity as the fraction of the internal surface of a phase (A) shared with other phase (A) particles in an A–B two-phase mixture. The mathematical expression was as the following equation [15,16]:

$$C_A = \frac{2S_{AA}}{2S_{AA} + S_{AB}} \tag{1}$$

where C_A is contiguity of A phase, S_{AA} is the surface area between A phase, S_{AB} is the surface area between A and B phase per unit volume. This equation is valid for any particle size, shape, and distribution.

In the present work, we explore the mechanical and thermal loading behaviours of bi-continuous metal – ceramic composites with varying phase interconnectivity and relate the composite properties to the microstructure using the concept of contiguity.

2. Microstructural geometry modelling

To examine different microstructures and their loading behaviours, four FE models were created representing four types of general microstructures. (see Fig. 1 for the RVEs of FE model). First two represent anisotropic scaffold structures which consisted of alternatively layered filament in 0/90° pattern along horizontal direction. By changing the shape of the filament, a discontinuous composite with round shaped filament (Type I, RF) and a continuous composite with square shaped filament (Type II, SF) can be obtained. These two are used to test the phase connectivity along the vertical (Z) direction as the filaments are arranged in parallel along the X and Y directions in a similar manner. The third RVE is triply periodic structures with a cubic-square-shaped connected phase (Type III, SQ) which enables us to test the variation of phase contiguity at a given overall phase volume fraction; and finally, the fourth model is the structure with a so-called triply periodic minimal 'P' interfaces between two phases (Type IV, PS) which will be used to test the property combinations and also provide comparison with other models.

These regular models are chosen so that the phase contiguity can be mathematically calculated to given meaningful comparisons.

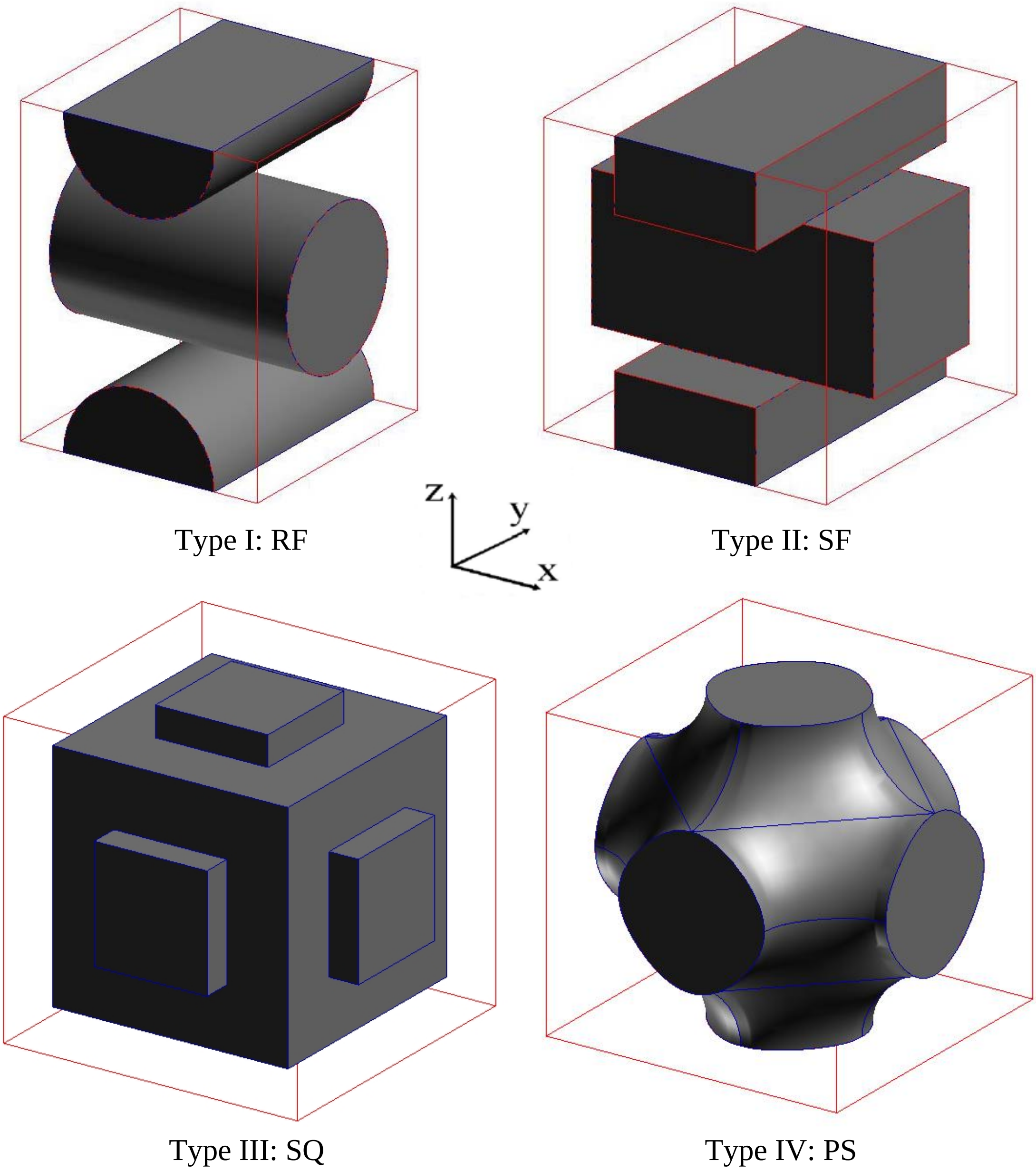


Figure1: Three-dimensional illustration of the RVEs consisted of four different types of microstructures with 50% volume fraction of reinforcing phase in this study

3. Finite element simulations

According to energy equivalence principle for the actual and equivalent homogenized materials, the homogenized effective material properties E^{H}_{ijkh} can be obtained by volume averaging of the variables E_{ijkh} in the RVE, following the definition:

$$E^{H}_{ijkh} = \frac{1}{|Y|}\int_{Y} E_{ijkh}\, dY \text{, where } |Y| = \int_{Y} dY \text{,} \tag{2}$$

which is subjected to the homogeneous stress/strain and the periodic boundary conditions applied to the RVE domain *Y*:

uniform stress/traction $\mathbf{T}^b = \boldsymbol{\sigma} \cdot \mathbf{n}(y)$ on ∂Y ,

or uniform strain/displacement $\mathbf{u}^b = \boldsymbol{\varepsilon} \cdot \mathbf{y}$ on ∂Y , (3)

and *Y*-periodicity $\mathbf{u}^b = \boldsymbol{\varepsilon} \cdot \mathbf{y} + \hat{\mathbf{u}}(y)$ on ∂Y , (4)

where $\hat{\mathbf{u}}$ is the periodic part of the displacement (local fluctuation) on the boundary surfaces and is dependent on the applied macroscopic loads, $\mathbf{y}$ is the position of the microscopic points.

For heat transfer problems, a uniform temperature gradient is applied, hence the temperature gradient on ∂Y is ,

$$\Delta \mathbf{T} = -\frac{\mathbf{q} \cdot \mathbf{n}(y)}{\mathbf{k}} \tag{5}$$

where $\mathbf{q}$ is the heat fluxes of the RVE, $\mathbf{k}$ is the coefficient of thermal conductivity.

To represent accurately the microscopic geometry of each model including the particles and matrix, 10-node tetrahedral finite element was used and there were around maximum 60,000 elements and 100,000 nodes for the models in this study. The finite element simulations were performed with the commercial code MSC-PATRAN/NASTRAN. The included phase shown in Fig.1 were regarded as elastic isotropic Alumina and the other phase (matrix—the opaque phase shown in Fig.1) was modelled as an isotropic elasto-plastic aluminium alloy. The flow stress was formulated [17] as $\sigma_{\mathrm{pl}} = A\varepsilon_{\mathrm{pl}}^{n}$, where $\varepsilon_{\mathrm{pl}}$ is the accumulated plastic strain *A* = 390 MPa and *n* = 0.15. The material properties used in the simulations are listed in Table 1.

Table 1: The properties of the Al_2O_3 and Al phases used in simulations

	Alumina phase	Aluminium phase
Young's moduous (*E*, GPa)	390	69
Poisson's ratio (ν)	0.2	0.33
Thermal conductivity (W/mm/K)	0.03	0.21
Specific heat (J/Kg/K)	850	900
Density (Kg/mm3)	3.96e-6	2.7e-6

4. RESULTS AND DISCUSSION

4.1 From discontinuous to continuous reinforcing phase

The FEM predicted elastic modulus of model RF (i.e., discontinuous along the Z-direction) and SF (i.e., continuous along the Z-direction) with various alumina volume fraction where the elastic modulus increased with the increasing volume fraction of alumina phase, seen in Fig. 2. This is applicable in both discontinuously and continuously reinforced composites that the effective elastic modulus is a property in associated with the overall phase volume fraction. Another interesting observation in this study is that the elastic modulus of composites is not only dependent upon the phase overall volume fraction but also on the phase contiguity. Fig.3 plotted the strengthening effect (the change of the elastic modulus) in relation to the contiguity of the alumina phase between the RF model and SF models at the given alumina volume fraction. It should be noted that the reinforcing filaments in RF model is considered as disconnected, therefore the corresponding contiguity of the alumina is zero. Generally the higher contiguity of stiffer phase, the higher elastic modulus was obtained.

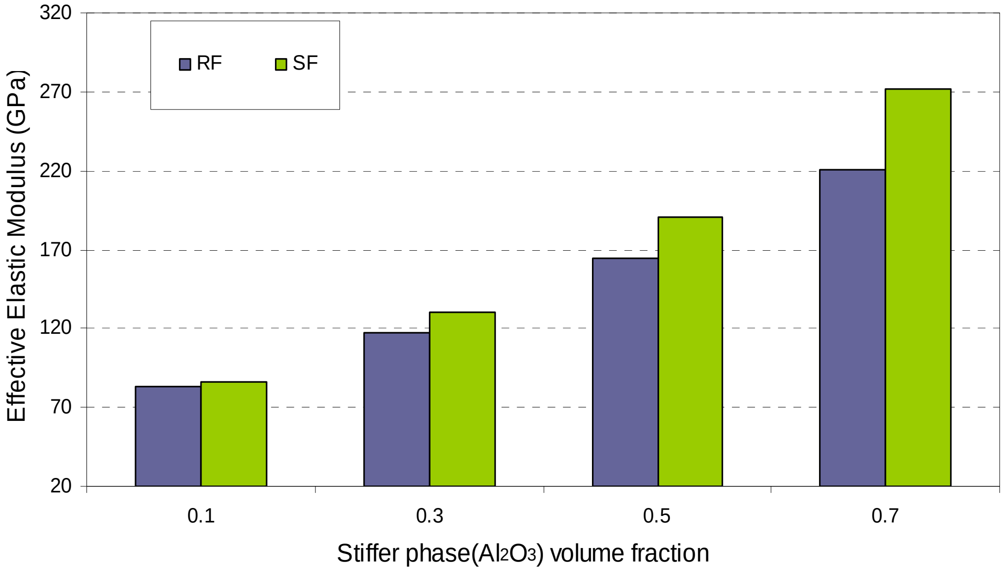


Figure2: Comparison of effective modulus along the Z-direction between the round filament (RF) and square filament (SF) models with varying overall volume fraction.

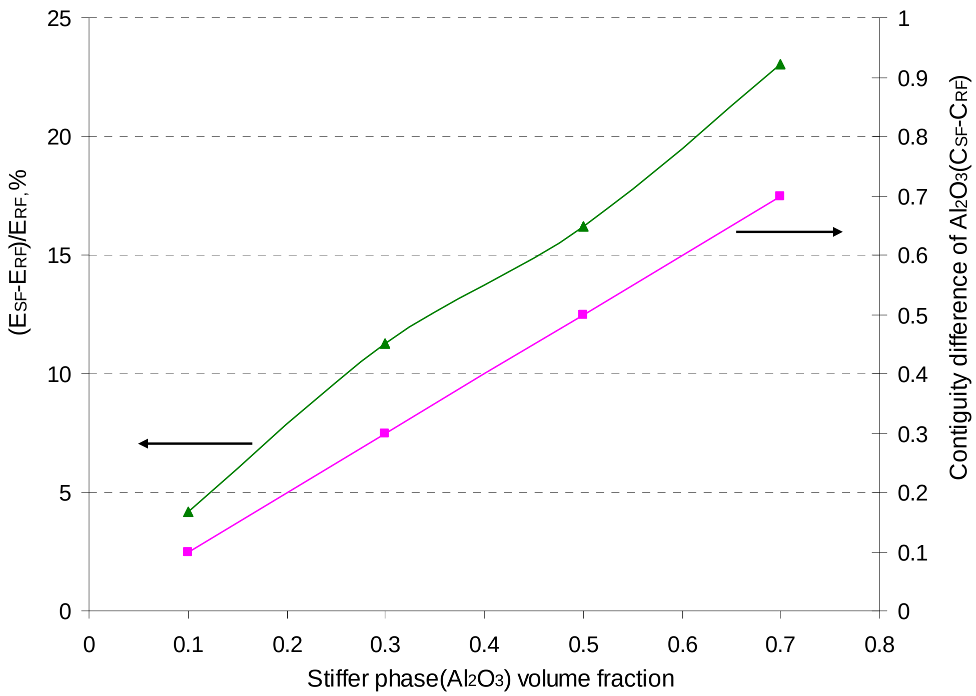


Figure 3: Percentage change of the elastic modulus in relation to the phase contiguity difference between the RF model and the SF model.[b]

[b] The phase contiguity of Al_2O_3 in RF models is zero.

4.2 Extended effect of the phase contiguity on the multifunctional properties

In order further to investigate the effect of contiguity on the material properties, the volume fraction of alumina was fixed at 50% in type III (SQ) microstructure. The variation of contiguity was realized by changing the dimensional ratio between the extruded square solid and the cubic block in centre. It was found that the yield strength of the composites increased with the increasing contiguity of the alumina at the same phase volume fraction, seen in Fig.4. It is proposed that the larger connectivity between stiffer phases can improve the load bearing ability during deformation. Further detailed work is being carried out to focus on the plastic deformation and fracture properties.

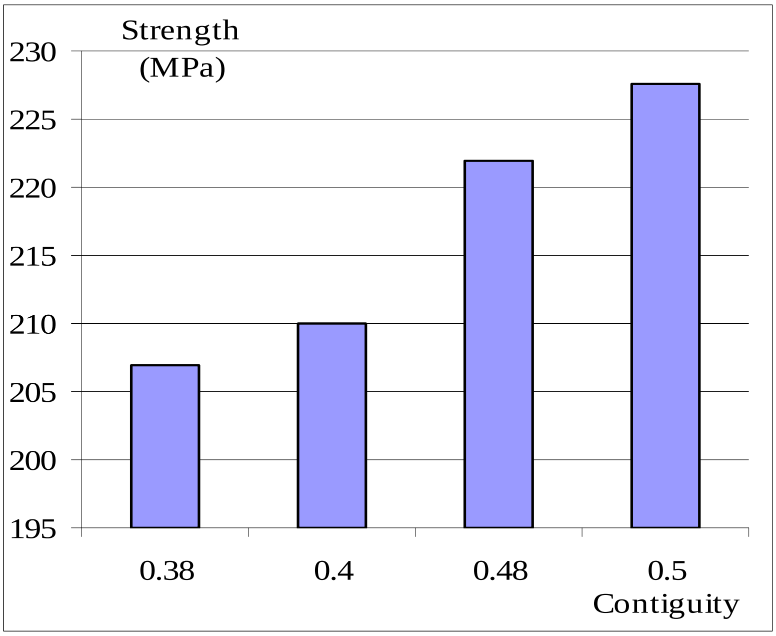
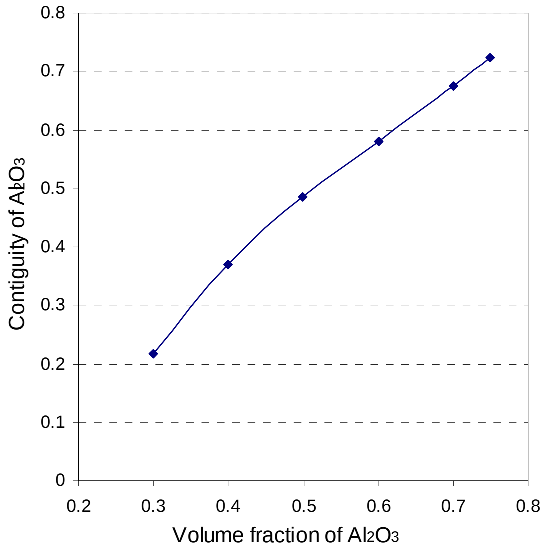


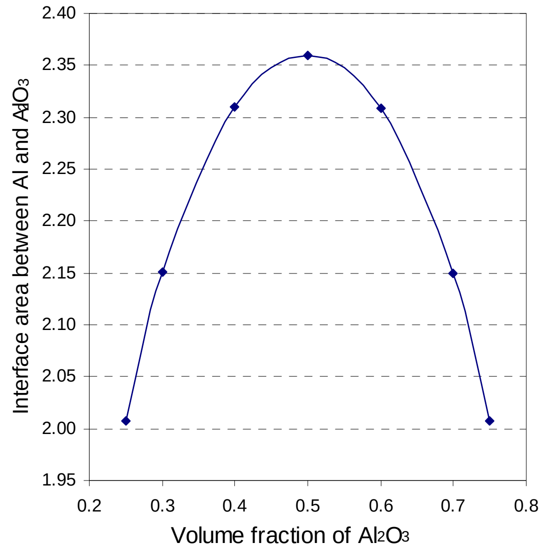
Figure 4: Effect of phase contiguity on composite yield strength of Type III- SQ model at fixed overall volume fraction of 50%.

According to the Equation (1), Fig. 5 shows the variation of the geometry properties (contiguity of the Al_2O_3 phase and the interface area between Al and Al_2O_3), elastic modulous and thermal conductivities of the type IV microstructure with different volume fraction. It can be seen from Fig.5 (a) and (b) that the contiguity values increase with the increase of the volume fraction, while the interface area between alumina and aluminium phase reaches the maximum value when the volume fraction is 50%. In Fig.5 (c) and (d), it shows that with the increase of the volume fraction, the elastic modulus increases while the thermal conductivity decreases.

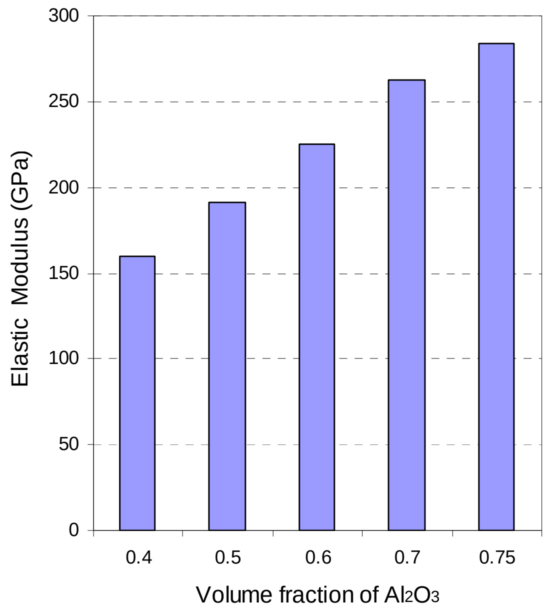
Indeed, it was demonstrated by the researchers [7] that this type of microstructure is not only geometrically extremal, but also extremal for both the effective thermal conductivity and elastic modulus when the volume fraction is 50%. This particularly makes sense in such a case where the properties of two phases are competing each other, i.e., one phase (e.g., aluminium) is a good thermal conductor but has lower stiffness while the other phase (e.g., alumina) is a poor thermal conductor but has a higher stiffness. Further investigations, including non-linear properties, are being carried out on more bi-continuous composites with different types of microstructure.



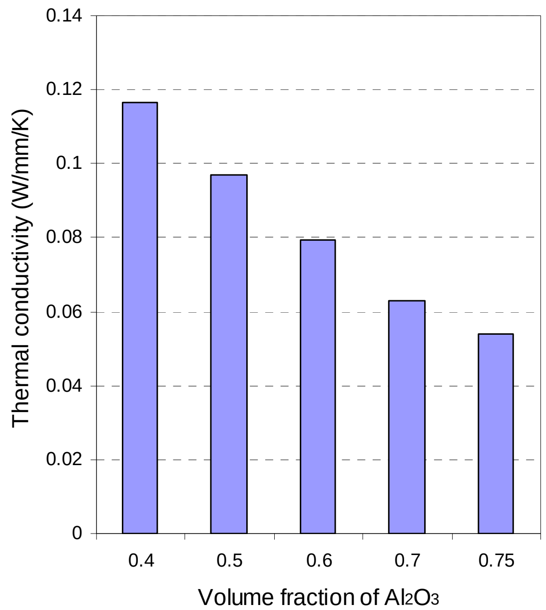
(a) Contiguity of Al_2O_3 versus its volume fraction



(a) Interface area between Al and Al_2O_3 versus volume fraction of Al_2O_3



(c) Variation of elastic modulus of composite with volume fraction of Al_2O_3



(d) Variation of thermal conductivity of composite with volume fraction of Al_2O_3

Figure 5: Variation of the properties of the Type IV composite with 'P' interface.

5. CONCLUSIONS

Finite element simulations were performed to study the effect of phase contiguity on mechanical behaviour of bi-continuous composite materials with different phase volume fraction. Four types of 3D periodic microstructures with varying phase contiguity were constructed in these studies.

Modelling results indicated that the elastic modulus was not only a function of volume fraction, but also increased with the increasing phase contiguity. The research concluded that the microstructure with larger contiguity in 3D can not only increase the elastic modulus but also to achieve better combined transport properties in the bi-continuous composites. These results can provide insights into the optimization and design of bi-continuous composites.

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REFERENCES

1. Clarke, D.R., “Interpenetrating Phase Composites”. J. Am. Cerm. Soc., 1992. 75(4): 739-759.
2. Peng, H.X., Fan Z., and Evans, J.R.G., “Bi-Continuous Metal Matrix Composites”, Materials Science and Engineering. Mat. Sci. Eng. A, 2001. 303: 37-45.
3. Hashin, Z. and Shtrikman S., “A Variational Approach to the Theory of the Elastic Behavior of Multiphase Materials”. J. Mech. Phys. Solids, 1963. 11: 127-140.
4. Daehn, G.S., Starck B., and Xu L., “Elastic and Plastic Behaviour of a Co-Continuous Alumina/Aluminium Composite”. Acta Mater., 1996. 44: 249-261.
5. Kouzeli, M. and Dunand D.C., “Effect of Reinforcement Connectivity on the Elastic-Plastic Behaviour of Aluminium Composites Containing Sub-Micron Alumina Particles”. Acta Mater., 2003. 51: 6105-6121.
6. Molchanov, I.S., Chiu S.N., and Zuyev S.A., “Design of Inhomogeneous Materials with Given Structural Properties”. PHYSICAL REVIEW E, 2000. 62(4):4544-4552.
7. Brighenti, R., “Fibre Distribution Optimisation in Fibre-Reinforced Composites by a Genetic Algorithm”. Compo. Struc., 2005. 71:1-15.
8. Torquato, S., Hyun S., and Donev A., “Multifunctional Composites: Optimizing Microstructures for Simultaneous Transport of Heat and Electricity”. Phys. Rev. Lett., 2002. 89(26):1-4.
9. Hyun, S. and Torquatoa S., “Designing Composite Microstructures with Targeted Properties”. J. Mater. Res., 2001. 16(1):280-285.
10. Hyun, S. and Torquatoa S., “Optimal and Manufacturable Two-Dimensional, Kagome-Like Cellular Solids”. J. Mater. Res., 2002. 17(1):137-144.
11. Torquato, S. and Donev A.. “Minimal Surfaces and Multifunctionality”. in R. Soc. London. 2004.
12. Michielsen, K. and Kole J.S., “Photonic Band Gaps in Materials with Triply Periodic Surfaces and Related Tubular Structures”. Phys. Rev. B, 2003. 68: 115107.
13. Wohlgemuth, M., et al., “Triply Periodic Bicontinuous Cubic Microdomain Morphologies by Symmetries”. Macromolecules, 2001. 34: 6083-6089.
14. Torquato, S., Hyun S., and Donev A., “Optimal Design of Manufacturable Three-Dimensional Composites with Multifunctional Characteristics”. J. Appl. Phys., 2003. 94(9): 5748.
15. Gurland, J., “The Measurement of Grain Contiguity in 2-Phase Alloys” TRANS. AMERICAN INSTITUTE OF MINING AND METALLURGICAL ENGINEERS 1958. 212: 452-455.
16. Aldrich, D.E. and Fan Z., “Microstructural Characterisation of Interpenetrating Nickel/Alumina Composites”. Materials Characterization, 2001. 47:167-173.
17. Segurado, J., Gonza´lez C., and LLorca J., “A Numerical Investigation of the Effect of Particle Clustering on the Mechanical Properties of Composites”. Acta Mater., 2003. 51: 2355–2369.