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Predictive modelling of the mechanical properties and failure processes in hydroxyapatite-polyethylene (Hapex^{TM}) composite

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The development of a wide range of hydroxyapatite polyethylene composites for medical applications is increasing the need for accurate predictive modelling. The objective of this work was to elucidate the observed mechanical processes and failure processes in this material using the finite element analysis method. The need for full three-dimensional modelling of this material has been shown. The results from this predictive model lead to accurate predictions of measured mechanical properties, and allow deduction of possible routes to improved ductility at high volume fractions.

1. Introduction

The development of a wide range of hydroxyapatite-reinforced polyethylene composites for medical applications is increasing the need for accurate predictive modelling. Theoretical investigations can make a substantial contribution to the further development of this important class of biomedical materials. One such composite based on a hydroxyapatite volume percentage of 40 was commercialized in 1995 for clinical application in middle ear implants involving replacement of diseased or damaged ossicular bones. By 1997, Hapex^{TM} had been incorporated into 22 different designs and has been utilized for several thousand patients.

The further application of this and related composites will involve their potential in major, as well as minor, weight-bearing applications. For such applications, the modulus of the replacement material should match that of bone, the ultimate strength properties must be adequate to prevent fracture, and the fracture processes must be essentially ductile. Both experimental measurements [1] and preliminary predictive results [2] indicate that the required stiffness of the material should be obtained at around 50%–60% volume fraction of hydroxyapatite particles. However, a sudden reduction of ductility is observed when the volume fraction of hydroxyapatite exceeds around 40% volume fraction. The required stiffness cannot be achieved with the required failure properties.

The objective of the predictive modelling presented in this paper is to elucidate the cause of this reduction in ductility and allow the deduction of a route to improve the failure processes at the required volume fraction of hydroxyapatite. A new three-dimensional finite element model describing this material has been developed. The results from this predictive model lead to accurate predictions of measured mechanical properties; the failure processes can be described and a route to improved ductility at the high volume fractions is deduced.

2. The finite element analysis

2.1. Material modelling

The finite element analysis method is a powerful tool for predictive modelling of composite materials. However, such an analysis cannot be properly carried out without due attention to the deduction of the material model. The finite element analysis method is based on the analysis of a representative cell of the materials. The representative cell is deduced from an assumption made regarding the distribution of the filler material. The deduction of the representative cell may be based on the assumption of either a fixed, regular distribution of the filler particles (e.g. [3]) or on the assumption of a random distribution (e.g. [4, 5]).

Previous modelling of this material has been performed using the assumption of a random distribution [2]. This material model is based on the concept that the interaction of neighbouring particles on the given particle is not directional; the overall effect is an "average" arising from all the neighbouring particles. Thus, the overall material can be divided into cells, each containing a single filler sphere with surrounding matrix. The boundary of a given cell is the region of the matrix closer to that particle than any other. These cells are the Voronoi cells; assuming a random distribution of particles, the distribution functions describing the interparticle distances have been calculated [6].

It is apparent that on an "average", the correct shape of the Voronoi cell is spherical. Thus, the correct overall material model for the assumption of a random distribution is a collection of spherical cells of different sizes, each containing a single sphere. However, previous work based on the assumption of a random distribution has been based on the assumption that the material consists of a collection of cylinders, each containing a sphere at its centre [2, 4]. Such an analysis is relatively straightforward, because the constraints required to model the interactions of surrounding particles may be simply applied by forcing the sides of the cylinder to remain straight. However, as described above, this material model is inherently inaccurate; it does not reflect either the assumption of a regular distribution or the assumption of a random distribution.

The method of analysis required for the more complex spherical material model has now been developed [7]. This method of modelling has been applied to the hydroxyapatite–polyethylene composite. Comparison of the finite element predictions of Young's modulus with experimental values is shown in Fig. 1. (Note that the results shown in Fig. 1 are for the injection-moulded composite.) Very poor agreement between the predicted and experimental values of Young's modulus is found, particularly at high volume fractions. It was deduced that this discrepancy must arise from the assumption inherent in this method of modelling; namely the lack of directionality in the interactions between particles. These interactions are expected to be very substantial in this material owing to the large difference in mechanical properties of the constituent materials. Indeed, better agreement between predicted and experimental values of stiffness had been obtained using the cylindrical material model [2]. It was concluded that this material should be modelled assuming a regular distribution; such modelling requires full three-dimensional finite element analysis. The cubic packing model was chosen because the representative cell for this material model is a simple geometric shape, namely a cube containing a sphere at its centre.

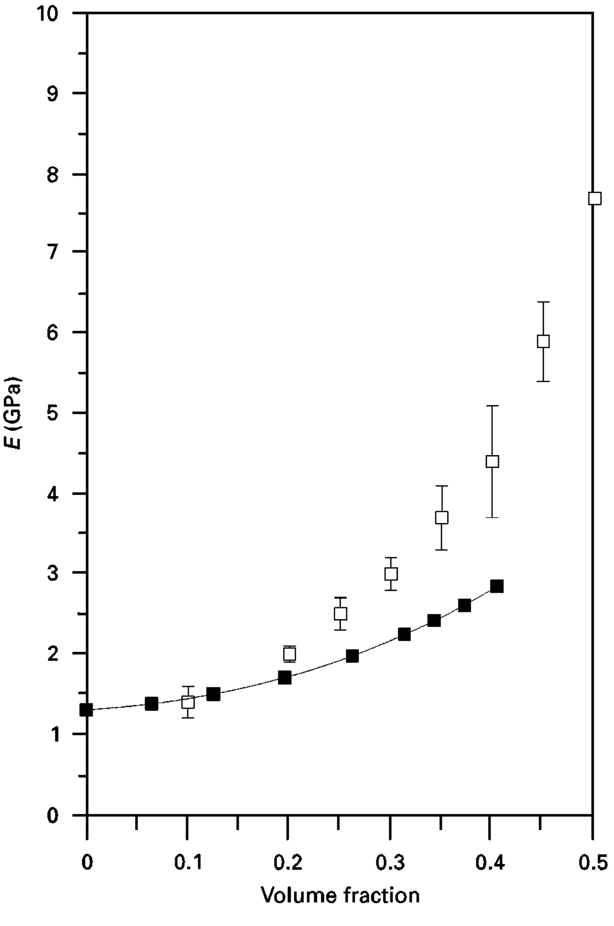

Figure 1 Comparison of (□) experimental and (■) predicted values, using the spherical material model, of Young's modulus, *E*.

2.2. The finite element grid

The representative cell is a cube containing a sphere at its centre. The symmetry of this cell allows the analysis to be carried out on one-eighth of the cell. The outline of the grid showing the segment of sphere is shown in Fig. 2. On deformation from unidirectional load applied in the 2-direction, the shape of the deformed grid must be a cuboid, with the faces of the cube within the whole representative cell remaining stationary and the remaining faces remaining parallel to their original directions. An original grid and the deformed shape are compared in Fig. 3. The constraints required to force this deformed shape model the interactions between the particles.

The grid was drawn using the mesh generator FEMGEN [8]; a separate grid is required for each volume fraction analysed. The grid was drawn using 20-noded brick elements. A typical grid contained 1025 elements and 4920 nodes. The finite element package used was ABAQUS version 5.5, running on a Convex 3860 computer.

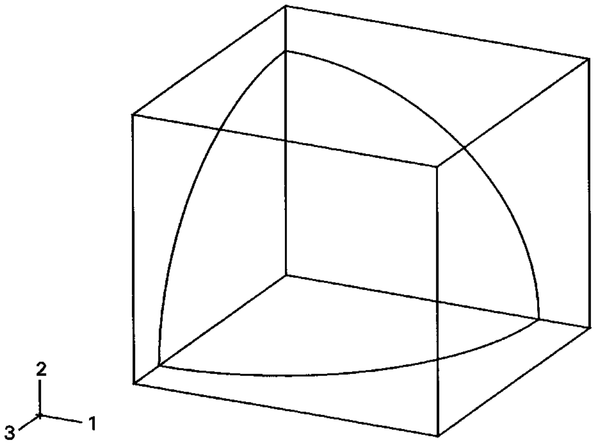

Figure 2 The geometry of the finite element grid.

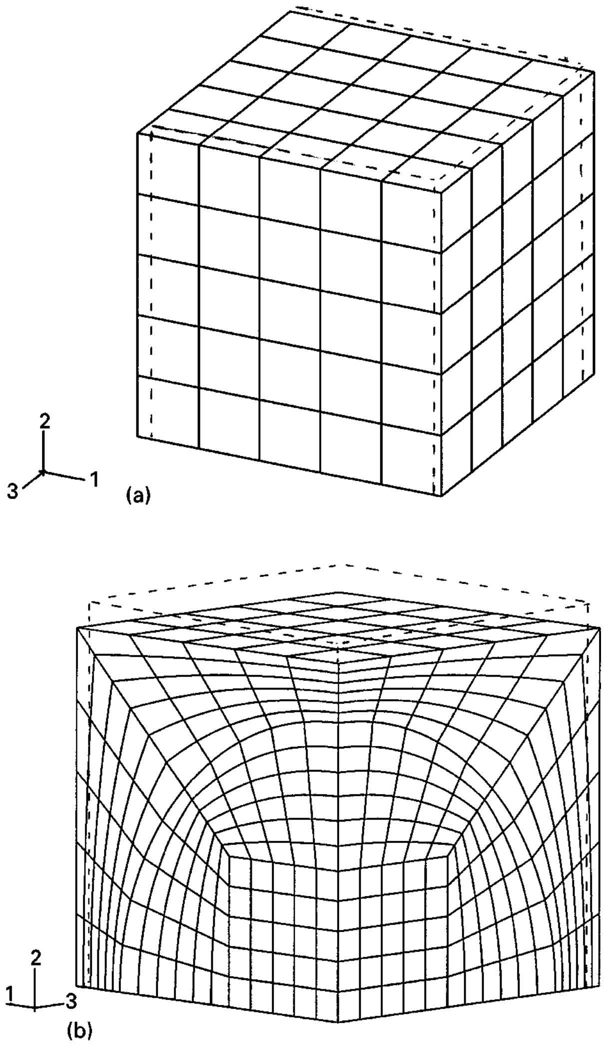

Figure 3 Comparison of original grid and deformed shape (- - -). (a) viewed from (+ 1, + 2, + 3) direction; (b) viewed from (− 1, + 2, − 3) direction, showing the segment of the sphere.

3. Material properties

All analyses reported here have been carried out assuming linear elastic behaviour for both the hydroxyapatite and polyethylene. The values required for the finite element analysis are Young's modulus, E, and Poisson's ratio, ν. The value of Young's modulus for this polyethylene has been measured experimentally. However, the value of Poisson's ratio is not well known. Although the value of Poisson's ratio can be calculated from the experimentally measured values of Young's modulus and shear modulus, an accurate value cannot be attained due to small experimental errors, which lead to obvious large errors in the values of Poisson's ratio. Therefore, the values of Poisson's ratio for the polyethylene matrix were calculated from the value of the bulk modulus, K. The value of the bulk modulus of polyethylene is expected to be in the range of 5–10 GPa [9]. Simulations were carried out using the calculated values of Poisson's ratio at either end of this range. The values used are shown in Table I; the analyses with the different properties are designated analyses 1 and 2.

TABLE I Material properties

	E (p)	ν	K (p)
Polyethylene matrix (analysis 1)	0.65	0.4783	5.0
Polyethylene matrix (analysis 2)	0.65	0.4892	10.0
Hydroxyapatite sphere	80	0.3	

4. Results

4.1. Elastic properties

Values of Young's modulus, E, were obtained from the finite element results from the sum of the 2-reactions-to-earth for the displaced nodes and the applied displacement. Values of Poisson's ratio ν, were found from the ratio of the resulting lateral (1 or 3) displacement to the applied (2) displacement. Because the overall material is isotropic, values of shear modulus, G, and bulk modulus, K, can be calculated using the well-known relationships for isotropic materials

$$K = E/[3(1 - 2\nu)] \quad (1)$$

$$G = E/[2(1 + \nu)] \quad (2)$$

Predicted values of Young's modulus are compared with experimental values in Fig. 4. There is good agreement between experimental and predicted values; the predicted values are higher for analysis 2, assuming the higher value of bulk modulus, as expected. Predicted values of shear modulus are compared with experimental values in Fig. 5. The predicted values are all in reasonable agreement with the experimental values; the agreement appears more accurate for analysis 1, using the lower value of bulk modulus, particularly at higher volume fractions. It is important to note that the value of shear modulus for the pure polyethylene used in the analyses is not identical to the measured value, although the value used is within the experimental error (see Fig. 5). The predicted value for pure polyethylene has been calculated from the material properties shown in Table I. As described above, the calculated value of Poisson's ratio from the measured values of Young's modulus and shear modulus is in obvious error; the value calculated is 0.157. The more proper procedure, as used here, must utilize the expected value of bulk modulus to gain the value of Poisson's ratio.

The predicted values of the bulk modulus, K, were calculated using Equation 1. The values are shown in Fig. 6. These values are hard to measure experimentally, but may be important for an optimized bone replacement material, as described in Section 5 below. As shown in Table I, the values are highly sensitive to the precise value of Poisson's ratio used for the polyethylene matrix. For both sets of material properties, the value of bulk modulus increases with increasing volume fraction of hydroxyapatite; the rate of increase

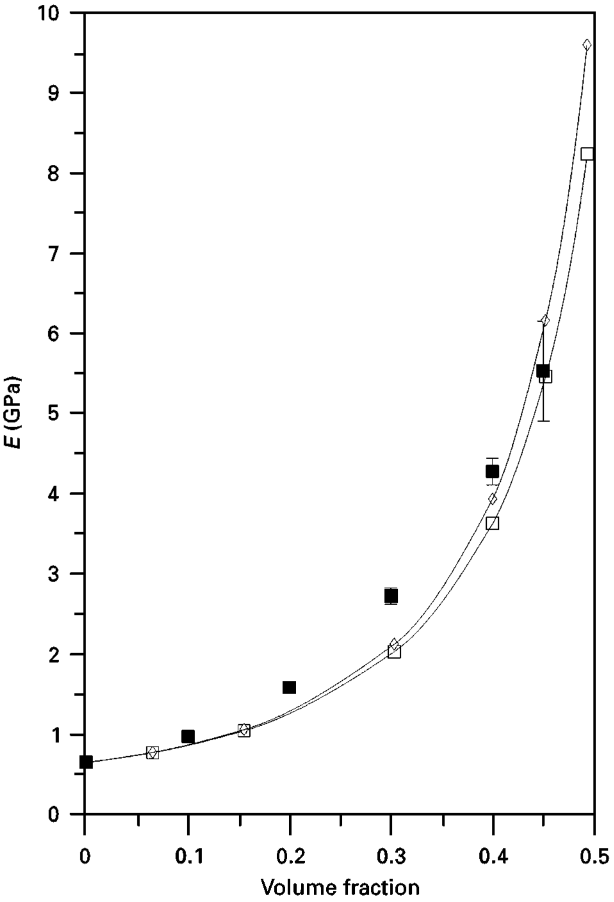

Figure 4 Variation with volume fraction of hydroxyapatite particles of experimental and predicted values of Young's modulus, *E* (□) FE predictions 1, (◇) FE predictions 2, (■) experimental values.

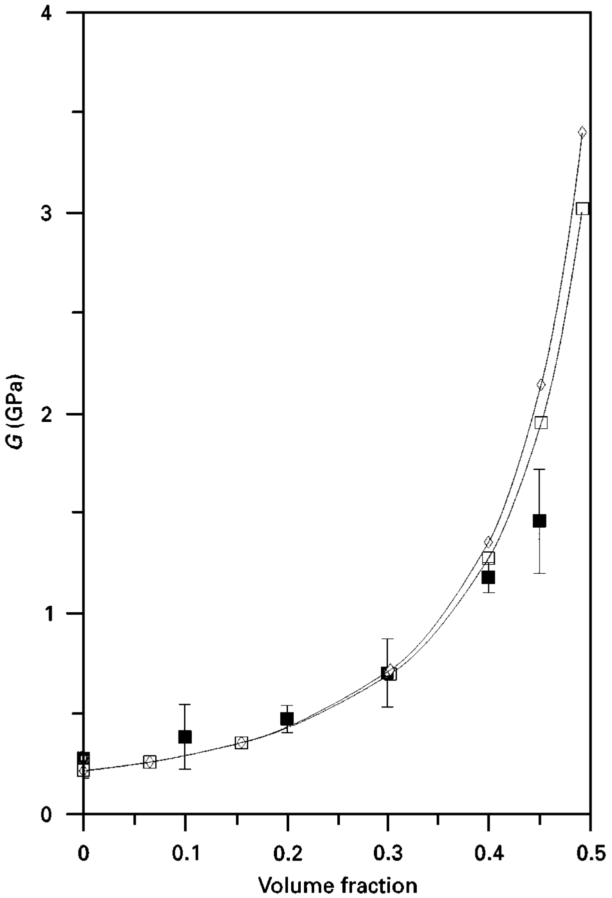

Figure 5 Variation with volume fraction of hydroxyapatite particles of experimental and predicted values of shear modulus, *G* (□) FE predictions 1, (◇) FE predictions 2, (■) experimental values.

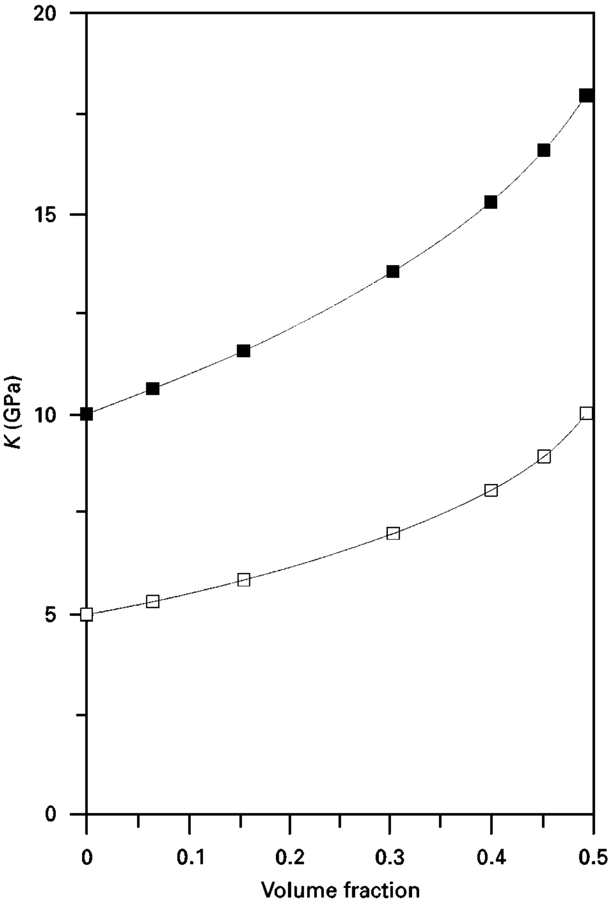

Figure 6 Variation with volume fraction of hydroxyapatite particles of predicted values of bulk modulus, *K*. (□) FE predictions 1, (■) FE predictions 2.

is slightly lower for the lower initial value for polyethylene.

The predicted values of Young's and shear moduli for the two sets of material properties are not very different in the range of volume fraction of the experimental results. Comparing the predicted results for the shear modulus, at the high value fractions measured, the experimental measurements are closer to the values predicted from analysis 1, using the lower value of bulk modulus. The comparison is less clear for the predicted values of Young's modulus, where the discrepancy between the experimental and predicted values occurs at the lower values of volume fraction, where there is little difference between the two sets of predicted results. There is, however, an indication of better agreement for analysis 1 for the highest value of volume fraction measured. Following careful consideration of these comparisons, detailed investigation of stress distributions has been carried out using the properties derived for the assumed lower value of bulk modulus of polyethylene (analysis 1 in Table I).

4.2. Stress distributions

The stress values studied are those which may be directly related to observed failure processes, as discussed in Section 5 below. The stresses considered are those within the polyethylene matrix, because failure of the hydroxyapatite particles is not observed. The

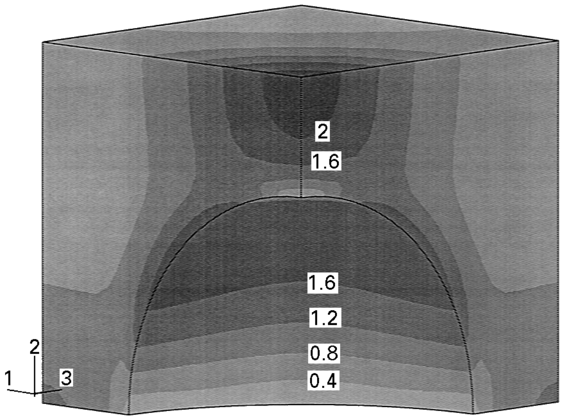

Figure 7 Contours of stress concentration factor of von Mises stress in the polyethylene matrix (viewed from (− 1, + 2, − 3) direction, with the segment of sphere removed).

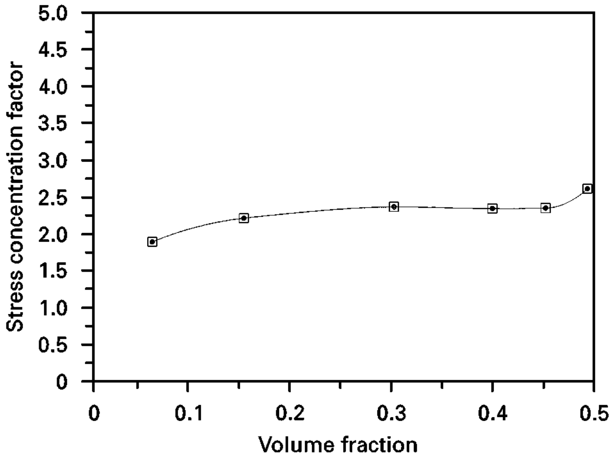

Figure 8 Variation with volume fraction of hydroxyapatite particles of predicted values of maximum stress concentration factor of von Mises stress.

variation of values of maximum stress concentration with volume fraction are presented as values of maximum stress concentration factor; this is defined as the ratio of the stress at the maximum position to the average applied stress.

The distribution of the von Mises equivalent stress, leading to shear yielding of the polyethylene, was examined. The positions of maximum stress found are similar to those found in the previous axisymmetric modelling [2]. There are two regions of stress concentration; above the pole of the particle, and at the interface. The positions are indicated by the contours shown in Fig. 7. These are contours of von Mises stress drawn on the surface of the grid shown; the grid is drawn with the hydroxyapatite particle removed. The direction of the view is similar to the view shown in Fig. 3b. At higher volume fractions, the position at the interface moves closer to the position at the pole; at around 40% volume fraction, the positions merge. The predicted variation of values of stress concentration factor of maximum von Mises stress with volume fraction is shown in Fig. 8. The maximum stress concentration factor is not sensitive to volume fraction. Shear yielding of the polyethylene will initiate at an approximately constant value of applied stress throughout the range of volume fraction.

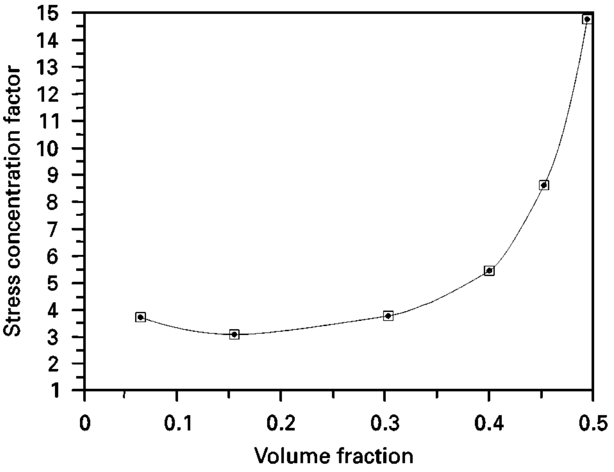

Figure 9 Variation with volume fraction of hydroxyapatite particles of predicted values of maximum stress concentration factor of direct stress.

The contours of distribution of maximum direct stress, leading to interface failure, were examined. For all values of volume fraction, the position of maximum direct stress was found at the interface at the pole of the fibre. The direction of this maximum direct stress was the direction of the applied stress. The predicted variation of values of stress concentration factor of maximum direct stress with volume fraction is shown in Fig. 9. The value of maximum stress concentration factor increases very steeply at values of volume fraction above about 40% volume fraction. For high values of volume fraction, the stress at the interface may reach a high value at low value of applied stress.

5. Discussion

Matching of stiffness between replacement bone material and natural bone is considered an essential requirement for effective long-term performance of the prosthesis. Reasonable agreement is found between experimentally measured values and predicted values of both Young's modulus and shear modulus. It is clear that, to match the tensile stiffness of bone, at least 50% volume fraction of hydroxyapatite is required. Values of bulk modulus have also been presented; this property is hard to measure experimentally. The predicted values are very sensitive to the precise material properties used (see Fig. 6). Measured values of bulk modulus for bone are not available in the literature. However, because the stresses applied to replacement bone are generally triaxial, the mechanical response may be dependent on the value of bulk modulus in addition to the values of tensile and shear moduli. For optimum performance, it may be important to match all elastic properties of the prosthesis to those of bone.

The predicted stress distributions can be correlated with observed failure processes in the material [1]. The ductile failure processes, observed for lower values of volume fraction, arise from ductile flow of the polyethylene leading to the formation of "fibrils" on the fracture surface. The fracture surface arising from the more brittle failure process, occurring for high values of volume fraction, show exposed hydroxyapatite particles, indicating interface failure. Failure of the particles is not observed. The material ductility at high volume fractions was not improved by increased adhesion at the interface using silane coupling [10]. The predicted stress distributions and values of stress concentration factor presented here are in reasonable agreement with these experimental observations. Ductile failure of this composite material may be described as the ability of the polyethylene to yield before the occurrence of any other critical failure process. The predictions show that yield of the polyethylene is expected at approximately the same applied stress for all values of volume fraction. However, at high values of volume fraction, above around 40%, there is a very high stress concentration of direct stress at the interface, at the pole of the particle. Such a high stress would be expected to lead to interface failure; for such high volume fraction of particles, with the particles in close proximity, this would be expected to be a critical failure process leading to failure of the material prior to extensive ductile yielding of the polyethylene. These are the failure processes observed, with a change of failure process at around 40% volume fraction of hydroxyapatite. Hence, it is not surprising that improvement of the adhesion at the interface using silane coupling failed to improve the behaviour.

The high concentration of direct stress arises from the interactions between the particles at high volume fractions, and the large difference between the material properties of the filler and the matrix (see Table I). Improvement in the ductility could only be achieved by modification of this difference in properties. This result could be achieved by increasing the stiffness of the matrix and this effect has been demonstrated experimentally [11]. Alternatively, the stress concentration could be reduced by incorporation of an intermediate modulus layer around the particle; this approach has been successfully demonstrated for reduction in stress concentrations in fibre-reinforced composite materials [12]. A possible means of introduction of such a layer could be via plasma coating [13]. The results presented here clearly demonstrate that such modification of the surface of the ceramic particles should be investigated.

6. Conclusion

Predictive modelling of this composite material has demonstrated the power of this technique in describing the observed material properties and failure mechanisms. The importance of the accurate knowledge of the properties of the constituent materials has been demonstrated. The source of the lack of ductility at the required high volume fraction of filler has been identified, and possible routes to an improvement in this critical material property have been deducted.

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