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Calcined Bone Ash Filled Polyethylene Composites

Khemchai HEMACHANDRA

Department of Materials Science, Faculty of Science, Chulalongkorn University

Abstract

Calcine bone ash (CBA) particulates and high density polyethylene (HDPE) were compounded to produce composites with volume fractions up to 0.50 for clinical uses. The production consisted of blending, compounding, and injection moulding. Tensile and microhardness tests were used to evaluate mechanical properties of the composites. The composite fracture surfaces and microstructures were studied by scanning electron microscopy.

Introduction

Various composite materials have been developed and used as implants for bone replacements (Bonfield, 1993). But the modulus values are not high enough as compared with that of human cortical bone. The closer stiffness matching hydroxyapatite - high density polyethylene (HA-HDPE) composites show promise in solving the bone resorption encountered with implants of conventional materials (Bonfield, *et al.* 1984). Bioactivity tests on HA-HDPE composites with high HA volume fractions reveal that new bone formation is promoted on the composite implants (Bonfield, *et al.* 1986). Calcined bone ash (CBA) is used to substitute hydroxyapatite due to availability and lower cost. In this study, the mechanical properties of calcined bone ash filled polyethylene (CBA-HDPE) composite are reported.

Materials and Methods

The matrix material in this study was a blow moulding grade high-density polyethylene and the filler was calcined bone ash particles with mean particle size 6.00 micrometre. The composites were produced by premixing polyethylene and calcium carbonate with volume fraction up to 0.50 by manual mixing and then compounding in a co-rotating twin screw extruder. Composite chips from the extruder were injection molded into thick sheets. Tensile specimens conforming to ISO/DIS 6239/1 were cut from the plaques, and the machined surfaces were smoothed using fine abrasive papers. All surfaces at the test specimens were free from visible flaws, scratches and other imperfections.

Results and Discussion

The data of the CBA-HDPE composites are shown in Figures 1 and 2 which plots volume fraction of CBA against Young's modulus and strain to failure. It can be seen that the Young's modulus increases with volume fraction. It should be noted that 0.5 volume fraction of CBA corresponds to about 0.75 weight fraction. i.e. CBA is the major constituent of the composite. The stiffening produced is accompanied by a decrease in the strain to failure, but the composite retains ductility until a CBA volume fraction of ~0.4. Hence composites with volume fractions less than 0.4 deliver the ceramic in a fracture tough condition.

The experimental data shown in Figures 1 and 2 cannot be modelled on the basis of simple rule-of-mixture calculations combining polyethylene (E= 1 GPa) with calcine bone ash (E = 80 GPa) , an approach which gives theoretical values much higher than the measured values. The theoretical approach needs to be modified but the curves shown in Figures 1 and 2 may be used as predictors for the experimental behavior of other similar ceramic- polymer composites.

Effects of strain rate on Young's modulus and strain to failure values of CBA-HDPE composites can be seen in both Figures. At higher strain rates, the composites show higher higher mechanical values. Also, this phenomenon is a factor affecting the mechanical values of the natural bone (Cowin, *et al.* 1987). As indicated, bone is stiffer and stronger at high strain rates. These relationships were said to hold for all bone in the skeleton.

From the SEM micrographs of fracture surfaces reveal voids around the calcined bone ash particles and strands of stretched polyethylene thinning continuously towards their ends. This indicates that is plastically deformed and stretched away from the parts of the calcined bone ash particles which lie perpendicular to the applied stress, as shown in Figure 3. The micrograph of a distinct form of micro-failure is seen and is denoted as craze. The craze is characterized by a void-fibril structure with dominant dimension, perpendicular to the applied stress. Associated with the craze is seen large dewetted calcined bone ash particle. The mode is seen more clearly in the Figure and the fibrillated nature of the craze is more distinctly revealed. Fully dewetted particle is evident in the midsection of the craze.

A possible explanation for crazing in filled thermoplastics is discussed by Chacko, *et al.* (1983) and Friedrich and Karsch (1981) and this is shown in Figures 4 (a), (b), and (c). Only a small amount of plastic strain is needed for the first step of damage. With further plastic strain the voids usually growing the stress direction, forming dimple-like holes around the particles (step II).

Subsequently, the stress concentrations would shift to the equatorial region since the void is not reinforcing. A profile of the void would accelerate the formation of craze (step III).

The strain levels in Figures 4 (b), (c) are continuously reduced with increasing filler fraction. This will lead to a transition from a more ductile mode of failure to brittle fracture behaviour. In the ductile mode, the polyethylene matrix is not restricted in it ability to stretch to support a load and therefore would be expected to provide greater strength. In the brittle mode, the coalescence of holes, which is the dominant mechanism of crack formation and fracture, will occur even before the ultimate dimple length is reached.

When the calcined bone ash volume fractions are low, all the mechanisms of void formation, dimple growth and plastic deformation of the polyethylene matrix can become effective in front of a crack, so a large plastic zone can be expected. It is a ductile mode of failure. In the case of higher calcined bone ash fraction, the polyethylene matrix is not able to develop its resistance to crack growth because so much energy release is obtained. In addition the maximum amount of strain at break is reduced by the coalescence of holes around the particles. This leads to a reduction of the plastic zone and a transition from ductile to brittle fracture behaviour.

In the case of bone, there are many points of crack tip instability in fracture propagation because bone is a nonhomogeneous, anisotropic and nonlinearly viscoelastic material. The discontinuous structure of bone such as blood vessel, lamellae, osteons is formed as the weak crosstinks between them. When a fiber breaks, there is a discrete drop in the stress at the crack tip and each fiber fail catastrophically because there is nothing to allow any changes in the stress. Hence, the discontinuous structure of bone leads to its discontinuous type of fracture, increasing the robustness of bone rather than increase its tendency for brittle fracture (Piekarski, 1970)

By comparison, the calcined bone ash reinforced polyethylene is isotropic material because there is continuously bound through the polyethylene matrix due to intermolecular forces. Then, at the tip of a propagating crack material can take up much of the stress by deforming plastically for some distance ahead of the crack tip. When material of the crack tip breaks, the stress is zero. The yield stress will be reach by material some distance in front of the crack tip. The reason of this continuity is the intermolecular attraction of polyethylene matrix, causing plastic deformation. Piekarski (1970) found that no bone has been stretched or drawn out into thin strands in the same way as the polyethylene.

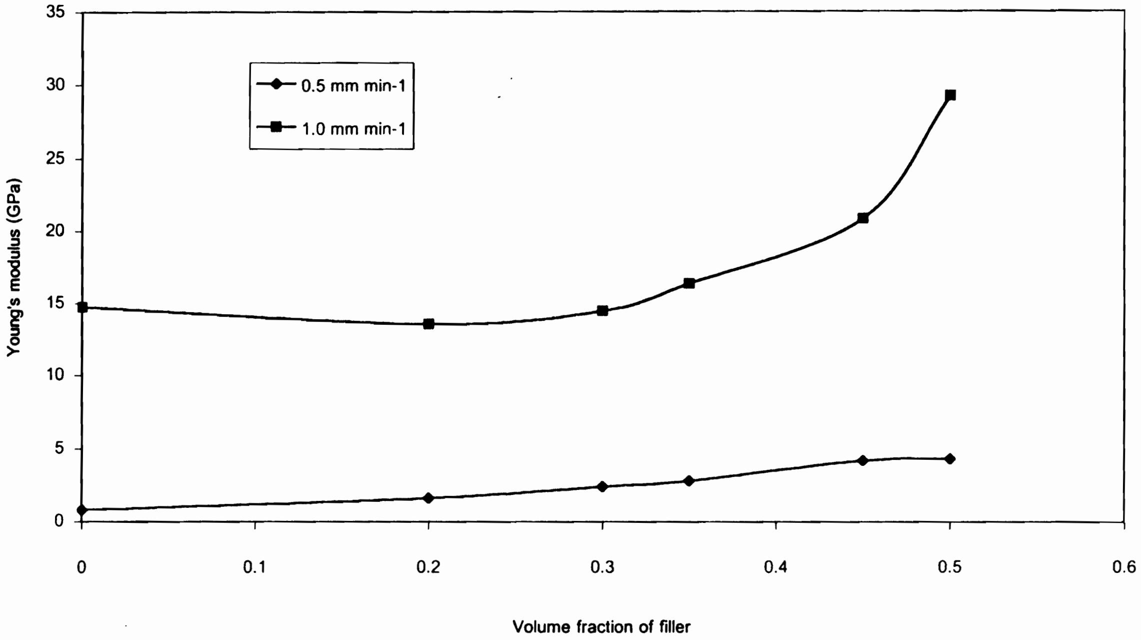

Figure1 The difference of the Young's modulus for two strain rates

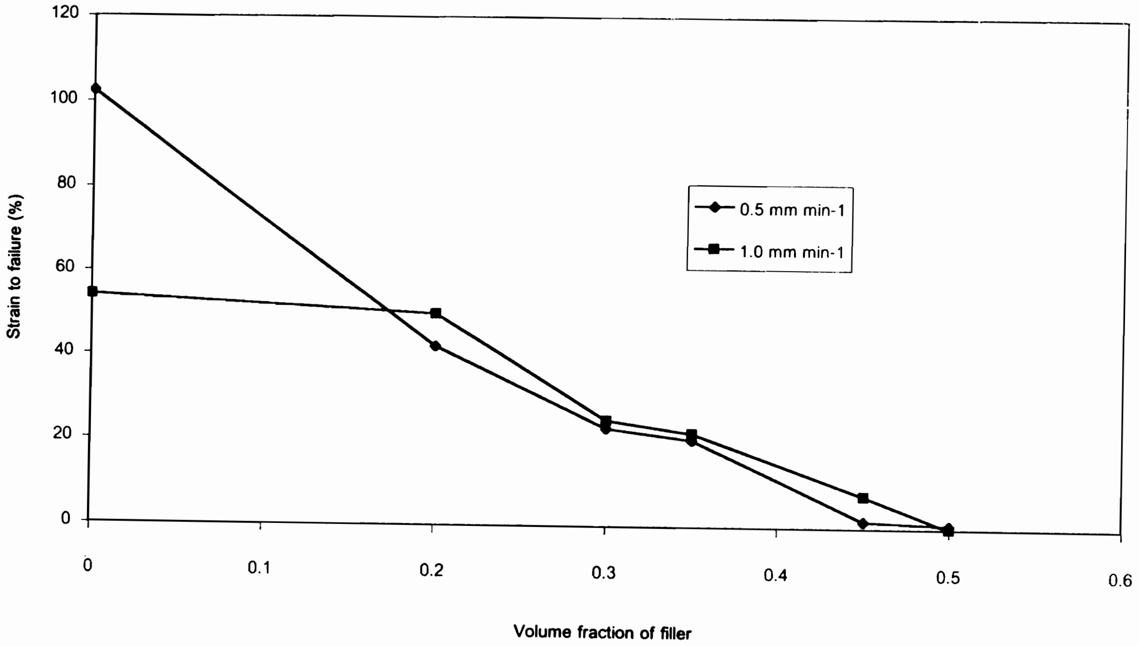

Figure 2 The difference of the strain to failure for two strain rates

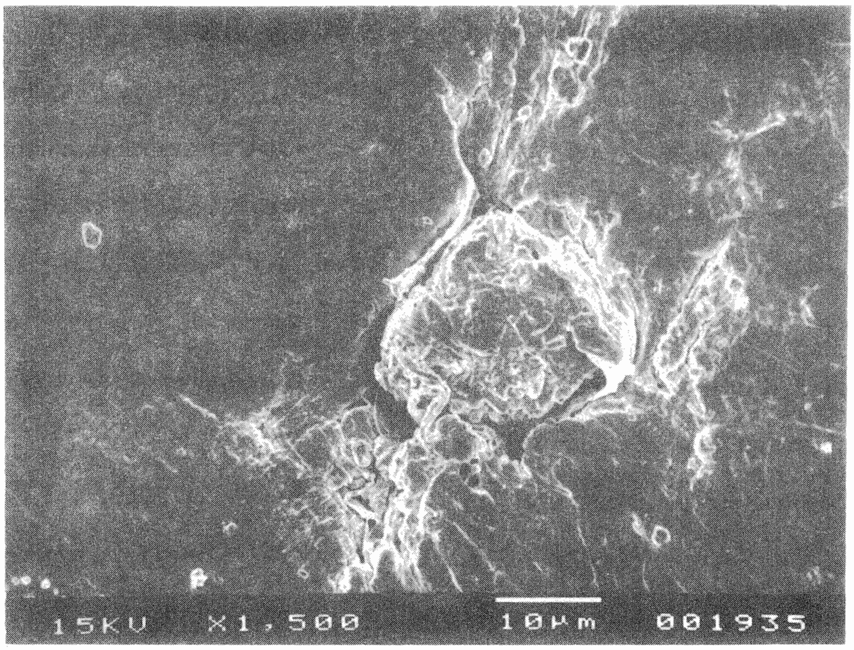

Figure 3 SEM images of a single calcined bone ash particle during test

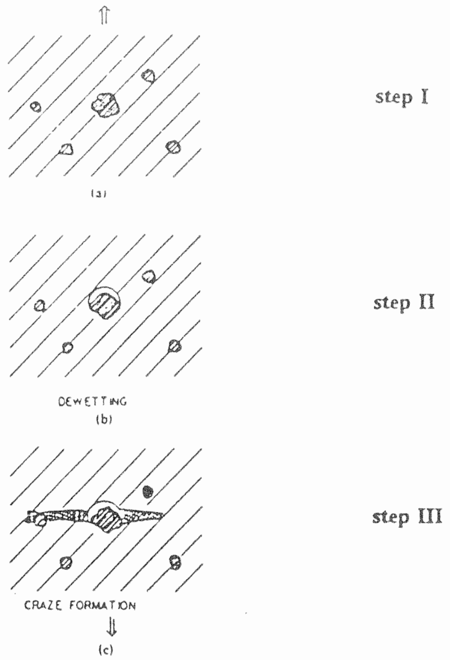

Figure 4 Schematic diagramme for purposed craze formation process in filled polyethylene : (a) initial confriguration; (b) dewetted particle; (c) craze deformation normal to the stress direction (from Chacko, Farris and Karasz, 1983)

Conclusions

The Young's modulus of calcined bone ash reinforced polyethylene composite increased with calcined bone ash content while tensile strength, strain to failure, energy to failure and toughness decreased over the same range. At higher strain rate, the Young's modulus values of the composite were higher than the low one. The micro-hardness of the composite increased with increasing volume fraction of calcined bone ash particles and it is a good predictor for the Young's modulus of the composite. Fractography of the composites revealed that the plastic deformation was strongly influenced by the amount of calcined bone ash leading to transition from ductile to brittle mode.

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