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Creep in polyethylene and hydroxyapatite reinforced polyethylene composites

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Isochronous stress–strain relationships and long term creep performance for unfilled and hydroxyapatite filled polyethylene composites have been studied. The tests were carried out in a buffered (pH = 7.5) Ringer's solution at 37 °C. It was observed that the inclusion of hydroxyapatite does not remove the non-linear viscoelasticity of polyethylene. The creep resistance is found to increase with increase in volume fraction of hydroxyapatite. The creep failure of composites at long times can occur due to debonding of the interface.

1. Introduction

An important design consideration in long term load-bearing applications of plastics is their integrity under applied load. Plastics are viscoelastic in nature and their deformation properties are time and stress dependent. Creep describes the deformation of materials with time under constant stress and provides useful information on the long term integrity of the materials. In the clinical application of polyethylene for the bearing surfaces of joint replacements, creep is a problem as it alters its mechanical performance and can lead to wear of the material causing adverse tissue response and eventually to revision [1]. The addition of reinforcing particles of hydroxyapatite has been shown to increase the modulus and the bioactivity of the composite material [2, 3]. The optimum filler volume fraction for both mechanical and bioactive properties is 0.40 volume fraction where the modulus of the composite reached the lower band of the modulus of bone, while the fracture toughness was higher than that of bone and the tensile failure was ductile. The objective of the work reported here is to characterize the creep performance of these composites in order to understand the mechanisms and to aid design using hydroxyapatite reinforced polyethylene composites in clinical applications.

2. Materials and specimen preparation

The matrix material was a high density polyethylene (Rigidex HM4560EP, B.P. Chemicals Ltd, UK). The filler was hydroxyapatite particles, grade P88, obtained from Plasma Biotal Ltd, UK. The materials were mechanically premixed by a commercial mixer and then compounded using a twin screw extruder (Betol BTS40L) producing 0.00 (denoted HDPE), 0.20 (denoted 20 HA/PE) and 0.40 (denoted 40 HA/PE) filler volume fraction composites. Four millimetre thick sheets of these materials were prepared by compression moulding. Tensile specimens conforming to ISO 527 [5] were then milled from the plaques using a pantograph. The machined surface was smoothed using fine abrasive paper. All the specimens were gamma irradiated at a dose of 2.5 Mrad before testing.

3. Experimental methods

3.1. Creep apparatus

The tests were performed on a purpose built three station tensile creep machine (Stephenson Engineering plc, UK) (Fig. 1). Each station has a lever arm which produces a mechanical advantage of 10:1. The specimen was positioned between two hooks in an environmental chamber containing buffered (pH = 7.5) Ringer's solution pumped from a common reservoir. The temperature in each chamber was monitored using a Pt-100 resistance thermocouple and was maintained at 37 °C throughout the test. To ensure axial loading without interference with free movement, a low-friction linear bearing, integral with the upper hook and the machine frame, guides the axial movement of the hook. The load was applied via a non-linear dashpot which enables the releasing mechanism to be rapid, but without vibration. The strain is measured by a Linear Variable Differential Transducer (LVDT). Two adapters were attached to the ends of the gauge sections of the specimen and extended out of the liquid chamber by mean of two metallic arms. These arms were then connected to the LVDT and the relative movement of the specimens could thus be measured. The signals received from the LVDTs were logged by Schlumberger Isolated Measurement Pods (IMPs) which were then transferred to be stored in a personal computer (Compaq 386) in a controlled stepwise manner.

3.2. Isochronous experiment

Isochronous experiments were carried out by loading and unloading the specimens in a repeated manner.

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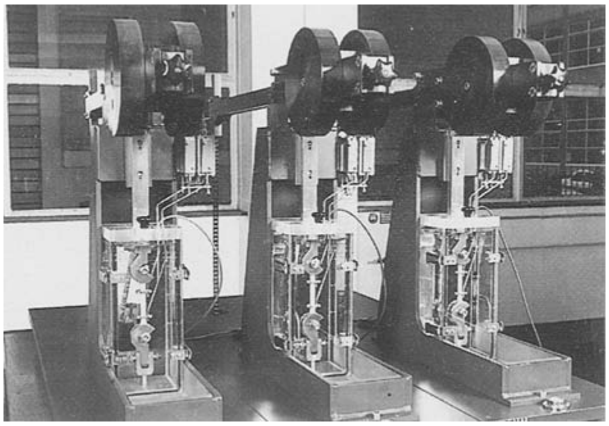

Figure 1 Creep apparatus.

The original gauge length (L_0) was assumed to be 50 mm for all the tests. The sample was stressed for 100 s, at a low load, and the deformation at that time was recorded (L_{100}). Then the load was removed and the specimen allowed to recover towards its original state for 400 s. The strain (ε_{100}) was calculated as follows

$$\varepsilon_{100} = \frac{L_{100} - L_0}{L_0} \tag{1}$$

The procedure was then carried out in this manner successively with the extensometer zeroed before each load step. The stress was increased at each load step as shown in Fig. 2, according to BS 4618 [6].

3.3. Creep testing

Creep tests were performed at various stress levels for each composition to establish similar 100 s creep strains. To produce a strain of 0.2% at 100 s, stresses of 2, 4 and 8 MPa were used for 0.00, 0.20 and 0.40 volume fraction composites respectively while stress levels of 4 and 8 MPa were employed to yield 0.6 percent creep strain for 0.00 and 0.20 volume fraction composites respectively. Creep strain was calculated using equation (1).

4. Results and discussion

4.1. Isochronous stress-strain relationship

Fig. 3 shows typical isochronous stress-strain curves of 0.00, 0.20 and 0.40 volume fraction hydroxyapatite polyethylene composites tested at 37 °C demonstrating nonlinear viscoelasticity. The inclusion of hydroxyapatite particles does not overcome the non-linear viscoelasticity of the polyethylene. An increase of hydroxyapatite content results in steeper stress-strain curves signifying an increase in modulus for the filled materials. Table I shows the relation between the volume fraction of the filler and moduli of the composites at various strain levels ranging from 0.1% to 0.5%. It can be seen that higher moduli were achieved with increasing hydroxyapatite content and that the moduli decreased as the strain level increased, which is non-linear viscoelastic behaviour. This trend is consistent with the relations reported earlier [3], but with different values due to the different test conditions.

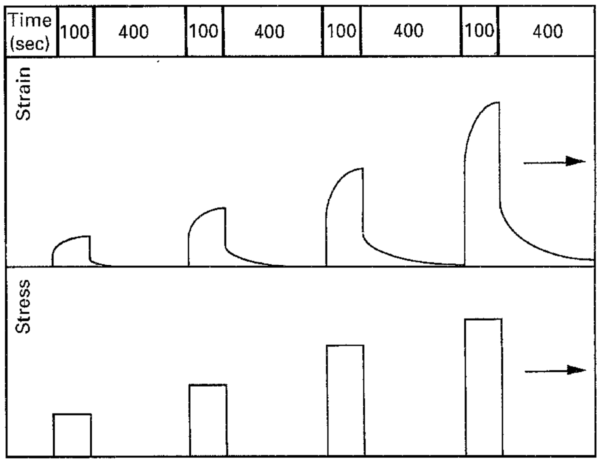

Figure 2 Diagram of isochronous experiment showing stresses applied with time and the resultant strains.

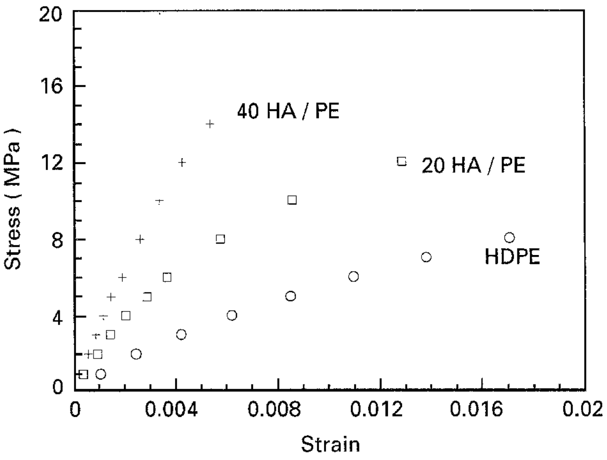

Figure 3 Isochronous stress–strain curves of 0.00, 0.20 and 0.40 volume fraction composites.

TABLE I Variation of creep moduli at various strain levels with filler volume fraction

Modulus (GPa)	Volume fraction		
	0.00	0.20	0.40
At 0.1% strain	0.962	2.155	3.420
At 0.2% strain	0.825	1.959	3.072
At 0.4% strain	0.716	1.643	2.813
At 0.5% strain	0.646	1.390	2.607

4.2. Creep behaviour

Fig. 4 shows the log–log plot of creep strain versus time for unfilled and filled materials under loads which produce 0.2% strain at 100 s. The use of the log–log plot has reduced the data to nearly linear relationships. Creep performance of the two filled samples was superior to the unfilled polyethylene and increasing filler volume fraction increased creep resistance as observed by the higher stresses required to obtain

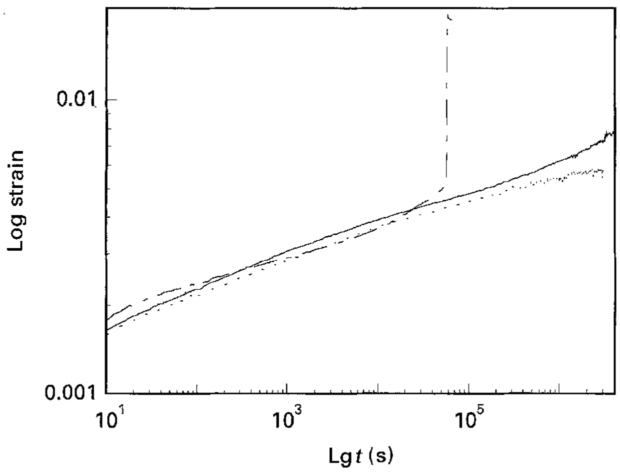

Figure 4. Log–log plot of strain versus time for 0.00, 0.20 and 0.40 volume fraction composites at the stresses of 2, 4 and 8 MPa respectively at 37 °C in Ringer's solution, (- - -), HDPE; (—), 20 HA/PE; (– – –), 40 HA/PE.

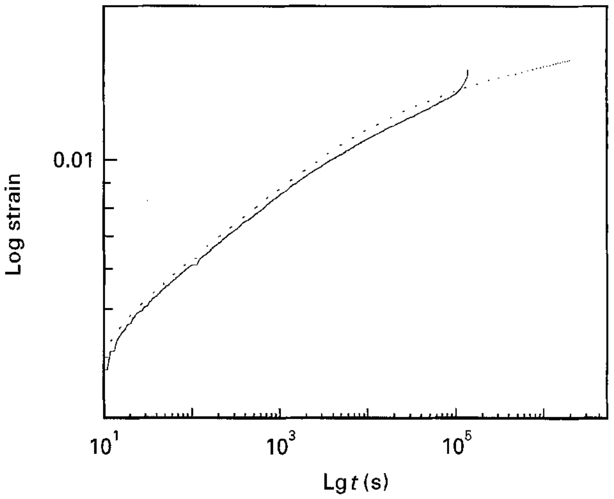

Figure 5 Log–log plot of strain versus time for 0.00 and 0.20 volume fraction composites at the stresses of 4 and 8 MPa respectively at 37 °C in Ringer's solution, (- - -), HDPE; (—), 20 HA/PE.

similar strain levels. The stresses applied for 0.20 and 0.40 volume fraction composites are around 2 and 4 times, respectively, the stresses applied to unfilled polyethylene. Although the initial responses of all three materials are quite similar, yielding nearly parallel straight lines, both composites later display an acceleration in creep strain which eventually leads to the failure of the material as indicated by the 90 degree upward divergence of the curve for the 0.40 HA/PE in this test. It should be noted that the stress levels of the filled materials were significant fractions of the ultimate tensile strengths of the materials, being 17.9 MPa, 17.8 MPa and 22.7 MPa respectively for HDPE, 20 HA/PE and 40 HA/PE [7] at room temperature and therefore lower at 37 °C. This divergence is in contrast to polyethylene which shows no sign of acceleration in creep strain up to the maximum period of the test, around 1 month. On the other hand, the behaviour follows the normal polyethylene pattern in that the slope of the curve decreases after passing a pseudo-equilibrium point and continues along in a second straight line.

It has been reported earlier that the second constant slope of a log strain–log time curve can continue as a straight line for at least a year for low density polyethylene [4] and this deviation is thought to be due to a separation of the fillers from the matrix. The interface adhesion in these types of composites is a mechanical bond, caused by the shrinkage of the polyethylene matrix onto the hydroxyapatite particles during the cooling after processing. The interfacial integrity depends on the friction force between the filler particles and matrix. When the stress is applied to the specimen, high local stress concentrations can occur as radial stresses at the hydroxyapatite polyethylene interface [8]. The maximum values is above the pole in the direction of the applied stress and this stress tends to pull the interface apart. 40 HA/PE exhibits a shorter acceleration time than 20 HA/PE. This result is probably because 40 HA/PE contains more filler and there are more possible debonding sites. Fig. 5 shows the log–log plot of strain and time at stresses producing 0.60% strain at 100 s. The creep behaviour of the materials at this stress level is similar to that at the lower stress level, but the acceleration time at higher stresses is shorter.

5. Conclusions

The inclusion of hydroxyapatite particles in polyethylene matrix improves the short-term creep resistance when specimens are subjected to similar stresses and increased filler volume fraction increases creep resistance. The increase of creep resistance is associated with the increase in moduli of the composites. However, creep failure of composites can occur at long times due to decoupling at the hydroxyapatite polyethylene interface. The marked divergence in the creep curve of sample 40 HA/PE that occurs after less than a day under a stress of 8 MPa can be expected after longer times at lower stresses. The quantitative relationships between applied stress versus log failure time (defining failure as the onset of the divergence) has not yet been established and requires accurate long-term creep tests at low levels of applied stress. Such tests are obviously essential for prospective implant materials.

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