The effect of the processing parameters on the fabrication of auxetic polyethylene

Part I  The effect of compaction conditions

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A novel fibrillated particulate microstructure has been fabricated in ultra high molecular weight polyethylene (UHMWPE) that produces a negative Poisson’s ratio (auxetic) material. The processing route involves compaction, sintering and extrusion of a UHMWPE fine powder. The first, compaction stage is examined in this paper in detail in order to ascertain the compaction conditions required to produce, as an end-product, an auxetic polymer and to assess the importance of this stage in the processing route. It was found that while part of the function of the compaction stage was to impart structural integrity to the processed polymers, the conditions for optimizing the production of auxetic UHMWPE were not identical to those for optimum structural integrity of the compact. Both sets of conditions were examined, with compaction pressure and temperature being the most important of the variables examined.

1. Introduction
The fabrication of a microporous form of ultra high molecular weight polyethylene (UHMWPE) which possesses a negative Poisson’s ratio (v) was first reported in 1992 [1]. This auxetic material, in common with a microporous form of polytetrafluoroethylene (PTFE) [2, 3], achieves a negative Poisson’s ratio by means of its microstructure which consists of an interconnected network of nodules and fibrils. When the material is deformed in tension, the nodules and fibrils react co-operatively to produce an expansion in the transverse direction with the fibrils causing the nodules to be pushed apart. In order to produce the auxetic form of UHMWPE, a novel thermal processing route was developed [1, 4] consisting of 3 distinct stages: compaction, sintering and extrusion of a fine powder of UHMWPE.

An initial set of conditions for the production of UHMWPE which combines an acceptable structural integrity (i.e. a modulus of at least 0.2 GPa [5]) and the property of being auxetic have been determined [1, 4]. The process commences with the compaction stage, which takes place within the barrel of a specially designed rig fitted with a blank die (see Fig. 1(a)). The internal dimensions of the barrel were a bore diameter of 10 mm and a length of approximately 90 mm. For convenience, these will be referred to as the standard conditions. Finely divided UHMWPE powder (supplied by Hoechst [6]) was poured into the barrel, which was at a preset temperature of 110 °C and left to equilibrate for a period of 10 min (referred to as the stand time). After this, compaction was achieved by lowering the brass tipped ram into the powder at a rate of 20 mm min⁻¹ (the loading rate) until an applied pressure of 0.04 GPa had been achieved, with the load being applied via the ram which was attached to the load cell of a Schenck-Trebel electro-mechanical testing machine for 20 min, with fluctuations between 0.041 and 0.037 GPa as the powder settled. After 20 min loading time, the load was removed and the compacted rod pushed out of the barrel.

After it had been allowed to slowly cool to room temperature, the compacted rod was reinserted into the barrel, which was now at a preset temperature of 160 °C. The same rig was used as in the compaction stage of the processing route, except that the blank die was replaced with a die of exit diameter 5 mm, cone semi-angle of 30° and capillary of 3.4 mm (see Fig. 1(b)). After 20 min sintering, extrusion took place at a rate of 500 mm min⁻¹, again at a temperature of 160 °C.

In this new work, the conditions necessary to make auxetic UHMWPE of larger dimensions (i.e. the diameter of the barrel was increased to 15 mm, the barrel length to 165 mm and the exit diameter of the die to 7.5 mm) and the effects of varying the processing conditions away from the standard case were explored. In particular, interest centred on achieving structural...
integrity of the extrudate and obtaining a range of Poisson's ratio. This paper forms part of a series [7, 8] which studies in detail the effects of varying the processing parameters within each stage of the fabrication route and concentrates on providing a greater understanding of the first stage of the processing route, i.e. compaction of finely divided UHMWPE powder to produce rods for further processing.

The room temperature (or cold) compaction process for conventional polymers is well documented and understood [9-11]. Crawford and Paul [9] explain the compaction process as occurring in a number of distinct stages. On filling a chamber, the powder particles fall into a random arrangement and hold each other in position by the formation of arches and bridges between adjacent particles and by friction between particles and the chamber wall. The density of the powder at this point is the lowest achievable and is referred to as the bulk density. As the powder is compacted, the applied pressure causes the bridging to be destroyed and the particles move to a lower position. From powder metallurgy, this stage is known to have an influence on the subsequent transmission of pressure through the powder. Further compaction results in the elastic deformation of particles at their contact points with other particles or the walls of the container. This elastic deformation allows further rearrangement of particles, with smaller particles moving to fill the gaps between larger particles. As the load increases, plastic deformation occurs, leading to a large reduction in porosity.

This investigation examines the effects of varying the compaction conditions on the microstructure, density, mechanical properties and ability of the rods produced to form an auxetic polymer. The primary function of compaction in this processing route is to impart some degree of structural integrity to the rods without (unlike conventional compaction) excessively reducing porosity. Hot compaction is necessary to aid structural integrity, normally obtained by the higher pressures of cold compaction. So it is expected that the requirements for the compaction stage of this process will be very different from those of previous work where the aim was a highly compacted end-product.

2. Experimental methods

The polymer used in this study was GUR 415 UHMWPE powder supplied by Hoechst [6] and its powder morphology is shown in Fig. 2. The barrel of the processing rig was heated by an external band heater, with the temperature being controlled by a thermocouple placed within the barrel wall (see Fig. 1(a)). Precise calibration of the rig (and indeed of all barrel and bore sizes used to date) has been carried out by placing a second thermocouple in the powder in the bore and comparing the temperature it records with that displayed by the thermocouple sited in the barrel wall. It was found that the temperature of the powder was within 2% of that recorded by the thermocouple in the barrel wall up to the required sintering temperature of 160 °C and beyond for all combinations of barrel and bore size used to date. In the example shown in Fig. 3, for the barrel and bore size used in this part of the investigation (i.e. a barrel of length 165 mm and bore diameter 15 mm), agreement within 0.1 °C was seen between the two thermocouples.

A typical schematic load against time trace obtained during compaction is shown in Fig. 4. This shows the stand time (under zero load), the compaction stage (where the load gradually increases at first followed by a rapid increase) and the loading time (where the load is maintained around the desired value).

2.1. Variables investigated in the compaction stage of the fabrication route

The starting point for this work was the set of standard conditions previously found to achieve auxetic properties in a smaller barrel geometry [1, 4] as detailed in the Section 1. In order to investigate the effects of each of the five variables in turn, one variable at a time was changed with the other four remaining