PROCESS FOR ORIENTING DISCONTINUOUS FIBERS IN THERMOPLASTICS

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SUMMARY: Equal channel angular extrusion (ECAE) can affect the alignment of materials at the molecular level. This process method is used to create novel metallic and polymer materials. The present work looks at ECAE as a method of producing high-value recycled short-fiber composites. Commercial glass fiber/polyacetal rods processed with ECAE were inspected to determine the effect of the process on fiber length and orientation at both room temperature and at 73 °C. The initial fiber orientation had a uniform distribution function about the major axis of the rods in the range of 11.8 to 56.0 degrees and the fibers were 105.6 ± 35.6 μm long. Processing at 73 °C did not change the length of the fibers, but it aligned them to a loglogistic distribution with a mean angle of 23.5 ± 18.2°. At room temperature the process both aligned the fibers and reduced their length by 23%. The authors conclude that temperature adjustments control the strength of the interface, which has an impact on the fiber fracture process, but not the flow mechanics, which control the fiber orientation.

KEYWORDS: ECAE, extrusion, recycling, short fiber, fiber fracture, fiber orientation.

INTRODUCTION

Equal channel angular extrusion (ECAE or ECAP) is a novel process that affects microstructures through extreme, uniform simple-shear [2]. Although the total strain history is large the material regains its initial shape when the process cycle is complete; it can be processed again if greater plastic deformation is beneficial. This extreme strain generates novel properties at the molecular level [3, 4]. Prior research with ECAE shows that the process aligns inclusions in metals and crystallites in semicrystalline polymers. Metal powders processed with the method can be consolidated and given specific textures. These effects might lead to benefits in new and

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[2] Z.Y. Liu et al., The effect of cumulative large plastic strain on the structure and properties of a cu–zn alloy, Materials Science and Engineering A, 242 (1- 2) (1998) 137-140

[3] Z.Y. Xia, H.J. Sue and T.P. Rieker, Morphological evolution of poly(ethylene terephthalate) during equal channel angular extrusion process, Macromolecules, 33 (23) (2000) 8746-8755

[4] Z. Xia et al., Dynamic mechanical behavior of oriented semicrystalline polyethylene terephthalate, Journal of Polymer Science, Part B, 39 (12) (2001) 1394-1403

recycled composites with a thermoplastic matrix polymer because ECAE can produce alignment in a bulk material.

Equal channel angular extrusion (ECAE or ECAP [5]) is a novel process that applies extreme strain to a material through uniform, simple shear [6]. Segal describes the process as rigid body motion of the material except for a moving thin layer that receives severe plastic deformation [6]. A finite element method (FEM) model of the process appears in Fig. 1; the darkest regions represent the steel die and the shaded portion is a model of a nylon billet subjected to the process. The top of the billet is forced downward until it plastically deforms in the shearing plane. The shearing plane appears as the shaded region 45 degrees from the vertical.

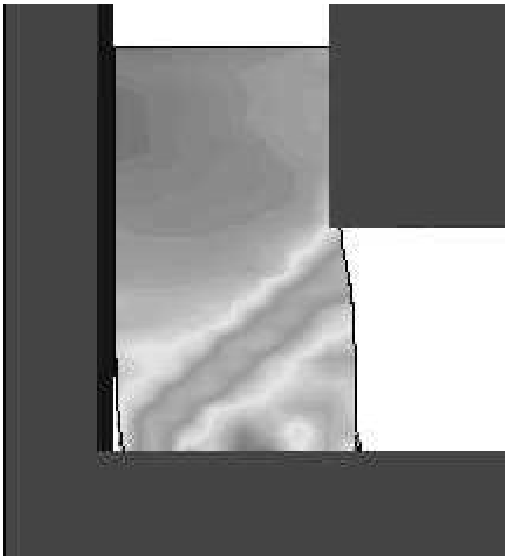

Fig. 1. FEM model of ECAE. The shaded region oriented 45 degrees from the vertical direction is the high shear strain plane. Portions of the billet above this plane have a negative vertical velocity and portions to the right of this plane have a rigid body velocity to the right.

The fracture of brittle fibers in polymer melt flow has been studied in detail [7, 8]. ECAE provides the novel opportunity to process in the solid state and have a useable material with new capabilities after extreme deformation. Before it reaches the shearing plane, the material undergoing ECAE is moving with solid body motion in the direction of the entrance channel. After passing the shear plane, the particles of the body are moving with the same magnitude of velocity, but directed at an angle of to the initial solid body motion. During the transition---the passing through the shear plane---the fiber can encounter bending and tensile stresses as its

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[5] Liu Z. Y., et al. (1998) The Effect of Cumulative Large Plastic Strain on the Structure and Properties of a Cu–Zn Alloy, Materials Science and Engineering A, A242(1-2):137-140

[6] Segal V.M. (1995) Materials Processing by Simple Shear, Materials Science and Engineering A, A197(2):157-164

[7] S.Y. Fu and B. Lauke, Effects of fiber length and fiber orientation distributions on the tensile strength of short-fiber-reinforced polymers, Composites Science and Technology, 56 (10) (1996) 1179-1190

[8] T.S. Creasy, S.G. Advani and R.K. Okine, Non-linear response of a long, discontinuous fiber/melt system in elongational flows, Rheologica Acta 35 (4) (1996) 347-355

trailing portion attempts to move with the initial sold body velocity and its leading portion attempts to move with the new velocity vector. A brittle, solid, body in a flow with a velocity gradient or sharp transition must either move with the velocity of its centroid or fracture into two fibers [8, 9]. The gradient or transition in velocity introduces strain in the fiber through the stress transfer at the fiber-matrix interface. The effect of this transition region is found from the experiments described below.

EXPERIMENTS

Material Studied

The specimens were commercially extruded cylindrical rods of polyacetal (Delrin™ 570), which is a semicrystalline thermoplastic. Polyacetal has a glass transition temperature (Tg) of -30°C, a thermal deflection temperature (TDT) under an 0.5-MPa load of 167°C and a melting point (Tm) of 178°C. Short glass fiber filler at a loading of up to 20% weight was added to the polymer by compounding and conventional extrusion. The dimensions of the rods were 12.7-mm diameter by 121 mm in length. Three pieces were processed via ECAE and one piece was analyzed as the 'as-received' condition of the composite.

Experimental Methods

The fiber mass fraction and mean fiber length were measured after the polymer was burned away [10]. Fiber orientation was measured by optical microscopy of cross-sections [11]. The data were processed using a rigorous statistical approach; at least 60 samples of each measurement were taken. Processing conditions included one pass at 73 °C and two passes at room temperature (RT).

RESULTS

Fiber Length

The fiber length distributions appear in Fig. 2. Although the initial fiber length distribution narrows—the shorter fibers are not affected, but the longer fibers are shortened—when the material receives plastic deformation at 73 °C, the change in mean fiber length is not statistically significant. The colder process at room temperature did significantly reduce the mean fiber length from 105.6 ± 35.6 µm to 81.0 ± 32.2 µm after a single pass through the die. This was a significant reduction in the fiber length. However, a second pass did not reduce the fiber length to a significant degree.

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[9] S.F. Shuler, D.M. Binding, R.B. Pipes, Rheological behavior of 2-phase and 3-phase fiber suspensions, Polymer Composites, 15 (6) (1994) 427-435

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[11] creasy kang j thermoplastic composites

Fiber Orientation

ECAE aligned the fibers when the material was processed at either temperature. The statistical distribution of the fiber angles changed from a uniform probability density function to a loglogistic distribution after a single pass. All three process conditions brought the mean fiber angle to about 23°. The second pass at room temperature did not shift the mean fiber angle farther.

CONCLUSIONS

Based on the measured effects of ECAE processing on the short fiber composites, the authors state these conclusions:

1) Fiber length may be a strong function of the processing temperature. If the matrix is softer—weak interface—the fiber cannot be loaded to its fracture stress.
2) Fiber orientation is not a strong function of the processing temperature. The same mean fiber angle was obtained with each process condition.
3) Number 1 above implies that using a specific processing temperature might control fiber length.

Conclusion number 2 has not been noted for ductile inclusions. Ductile inclusions become fibrous after multiple ECAE processing passes and they continue to align with the major axis of the extrudate. The results here are novel because they indicate that brittle, fiber shaped inclusions approach a fixed angle that is away from the extrusion axis.

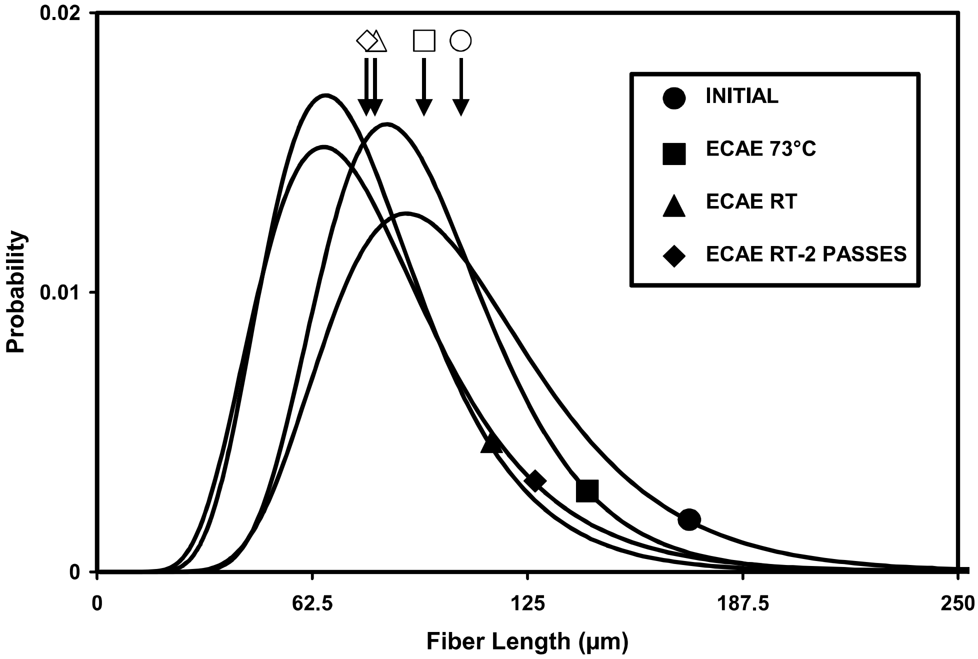

Fig. 2. The initial fiber length is not significantly reduced when the composite is processed at 73 °C, but the process at RT does reduce the fiber length during the first pass. A second pass produces no further reduction in length. [10]

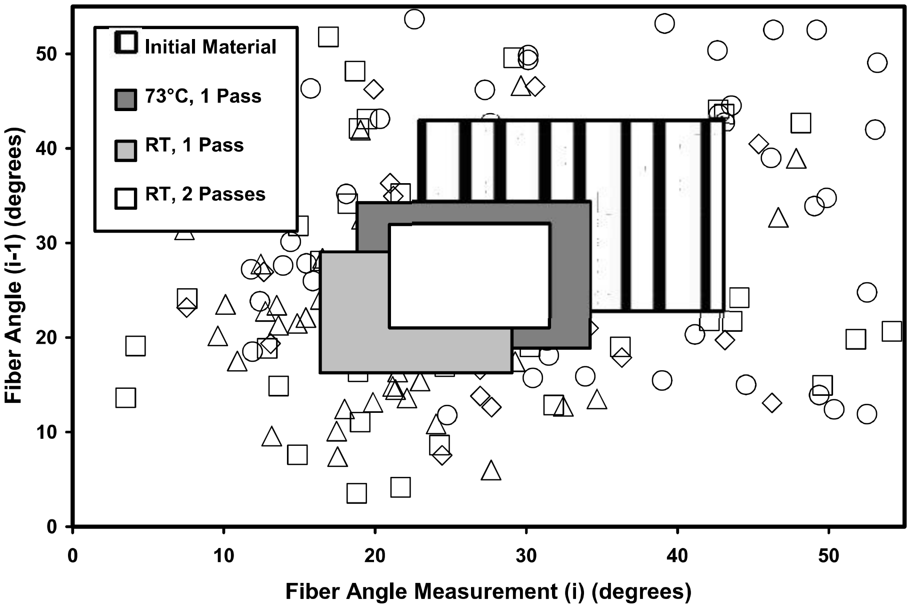

Fig. 3. The lag chart shows the range of fiber angles found in initial and processed short fiber composite. Each rectangle encloses the interquartile range—middle 50%--of the fiber angles measured for the condition. ECAE moves the center of each rectangle toward a mean fiber angle of 22°.

ACKNOWLEDGEMENTS

The authors are grateful to Mr. Yoosin Kang and Mr. Robert Barber, who processed the materials and to Dr. K. T. Hartwig for the use of his ECAE apparatus.

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