EQUAL-CHANNEL ANGULAR EXTRUSION OF THERMOPLASTIC MATRIX COMPOSITES FOR SHEET FORMING AND RECYCLING

T. S. Creasy and Y.S. Kim
Polymer Technology Center
Texas A&M University

Abstract

Equal channel angular extrusion creates novel properties in metal and polymer materials. Recently the authors investigated the effects of this process on commercial short fiber composites. Experiments show that ECAE provides a means for controlling fiber length and orientation in the extrudate. The process might transform continuous fiber thermoplastic matrix composite sheets into high volume fraction discontinuous fiber sheet for thermoforming. In addition, the process might provide a method of recycling reground components into high-value sheets with a known fiber orientation.

Introduction

Automotive applications of high stiffness composite materials have been limited because the total cost of fabricating composite structures is high. Raw materials cost is high and the fabrication methods are labor intensive. Since most automotive body panels are created from stamped sheet metal, stamped composite sheets could provide a means of making structures at low cost. However, stamping composites that contain continuous fibers leads to wrinkling and buckling effects that reduce structural performance. Discontinuous fiber materials can reduce these buckling effects; however, it is difficult to create highly aligned fibers at the high volume fraction needed for best performance. ECAE might preprocess continuous fiber composites and change them into novel, highly functional, sheets for stamping.

Background

ECAE is an emerging process for creating novel microstructures in materials. The longest use of the process has dealt with metals; however, research has recently turned to polymer materials. V. Siegel invented the process while he was in the Soviet Union. After he immigrated to the United States, he worked with the faculty at Texas A&M University to develop processes for making advanced metals and metal matrix composites. Once the facility was in place for processing materials, the research expanded to include amorphous and semicrystalline polymer materials. Using ECAE to treat composites is the most recent application of the process.

Equal Channel Angular Extrusion

Equal channel angular extrusion (ECAE) is not a process for manufacturing parts. ECAE is a treatment that profoundly affects the properties of materials. Further machining and fabrication processes form the ECAE treated material into components. Metals processed by ECAE can be work hardened to previously unobtainable levels[1]. Polymers treated via ECAE can have

[1] Segal V.M., Hartwig K.T. and Goforth R.E., Materials Science and Engineering A, A224(1-2):107-115 (1997)

improved fracture toughness and molecular alignment[2]. This paper looks at the effects of ECAE on discontinuous fiber reinforced thermoplastic composites with the objectives of making a unique new composite material for sheet forming of automotive body panels.

ECAE forces a solid material through a die that has an entrance and an exit channel of identical cross section. The entrance and exit channels join at an abrupt change in angle. Figure 1 shows schematic views of two dies with different angles between the channels. The total shear strain applied to a material that passes through such a die is found from this equation:

$$\gamma = 2N \cot \varphi \qquad (1)$$

where γ is the total applied shear strain, φ is ½ of the angle between the channels, and N is the number of times the material passes through the die. A die with channels 90 degrees apart will subject the material to 100% shear strain for each time the material passes through the die. Part of this strain will be plastic deformation and the elastic part recovers as the material moves out of the shearing zone.

The treatment is controlled through five process parameters:

- Angle between the channels
- Temperature during treatment
- Pressure generated in compression
- Rate of extrusion
- Rotation of the material between treatments

Rotation of the material between extrusions is possible because the material retains its initial cross section. The ECAE community has given names to specific treatment strategies. If the cross section is rectangular, the material can enter the die with the same orientation, which is called Route A treatment, or after a rotation of 180 degrees, which is called Route C treatment. If the material is square or circular in cross section 90-degree rotations are possible. These strategies are Route B processing. For this paper the short fiber composites were treated with one or two passes of Route A.

ECAE and Composite Materials

The authors are studying the ECAE treatment's effects on composite materials with emphasis on achieving two objectives:

1) Converting highly concentrated, continuous fiber reinforced thermoplastic composites into high stiffness/strength materials for sheet forming.
2) Using the process to recycle used composites into a high value product in rod, bar, sheet or plate form.

[2] Xia Z.-Y., Sue H.J. and Rieker T. P., Macromolecules, 33(23):8746-8755 (2000)

High Volume Fraction Discontinuous Fiber Sheets

Thermoplastics loaded with high volume fractions of fibers have high stiffness and strength[1]. However, high fiber volume fractions are only available if the fibers enter the polymer as highly aligned tows of continuous fibers. Continuous fiber systems are difficult to form into complex shapes by high speed stamping operations. Long discontinuous fiber sheets produced complex stamped shapes with little waste, wrinkling, or buckling. However, the fibers were overly long and introduced nonlinear flow effects that limit the strain rate to flows that are too slow for commercial applications[3].

The ECAE treatment may provide a means of converting continuous fiber reinforced sheets into discontinuous fiber sheets with fibers at a controlled length. An optimum length will provide the stiffness and strength needed for an application while allowing forming at high stamping rates. To provide these materials, we must control the fiber length distribution in the material and control keep the fibers aligned.

Recycling of Thermoplastic Matrix Composites

Recycling of automotive components is a growing concern. If high performance thermoplastic composites are recycled, they are reground into pellets that can be reprocessed with virgin material in an extruder. If the composite started as a high performance product, it ends up in a low performance form following extrusion processing. The regrind and plastification processes reduce the fiber length and the fiber orientation varies from random in the middle of the extrudate to aligned at the die/polymer or mold/polymer interface. These effects occur because of the characteristics of melt flow processing of fiber suspensions.

ECAE occurs in the solid state and it affects the entire cross section of the material with the same shear history. This should produce a uniform state in the material that aligns the fibers to the same orientation throughout the rod or sheet. This would produce a material with consistent known properties with higher performance from the highly aligned fibers.

Experiments with Short Fiber Materials

The flow processes in ECAE are complex. The high amount of strain and the interaction of the fibers and the matrix require a careful experimental approach. The best approach is to start with short fiber composites that have a low concentration of fibers. This limits the fiber/fiber interactions during extrusion and allows the fibers to interact freely with the matrix by tumbling, bending and stretching. As the interaction is understood for a given fiber loading, the length and volume fraction of the fibers can be increased to approach the goals of high performing recycled and stretchable virgin materials.

The initial composite used was a commercially prepared glass fiber/Delrin-570 composite. The rods were 12.7-mm diameter by 121 mm long. Aluminum cans held the rods and carried them through the square cross section ECAE die. The die channels were 25 mm square with an included angle of 90° between the entrance and exit channel. We followed the effect of the ECAE treatment on the fiber length and fiber alignment of the material when processed with one or two passes using the Route A method.

[3] Creasy T.S., Advani S.G., and Okine R.K., Rheologica Acta, 35:4 347-355 (1996)

Effect on Fiber Length

The fiber length of the composite was determined at each process state from fibers extracted by a polymer burn off. Treated composite was placed in a high temperature furnace in air until only bare glass fibers remained. A sample of the fibers was spread on a glass slide and at least 60 fiber lengths were measured using a calibrated digital imaging system.

Table I lists the fiber lengths found for each condition of the composite material. ECAE extrusion at 73 °C reduced the initial mean fiber length of 106 μm to 95 μm; however, this drop is not statistically significant. This result shows that softening the polymer can keep the fibers from experiencing further reduction in their lengths.

Room temperature (RT) processing of the materials did reduce the fiber length. At room temperature the polymer is stiffer than it is at 73 °C. The mean fiber length is only 81 μm and this mean value is significantly different from the initial fiber length. A second ECAE treatment did not produce a further drop in mean fiber length. The mean of 78.4 μm is not significantly different from the fiber length following one treatment.

The results indicate that fiber length can be controlled by using the process temperature to adjust the stiffness of the matrix and the strength of the fiber/matrix interface. In addition, repeated processing at a given temperature will not continue to reduce the length of the fibers.

Effect on Fiber Orientation

In this work we define the fiber angle as the angle between the long axis of the composite cylinder and the long axis of the glass fiber. The composite was cut perpendicular to the long axis of the cylinder and the exposed surface was polished. Fibers crossing the polished surface at an angle appear as an ellipse. Digital analysis of micrographs of the polished cross section of the composite provided a measure of the major and minor axes of the fibers that intersect with the polished surface of the composite. These numbers provide the angle through this equation:

$$\cos\theta = \text{minor } axis / \text{major } axis \qquad (2)$$

where θ is the angle of the fiber axis with respect to the plane of the image.

The ECAE process strongly affects the fiber orientation. Table II lists the mean fiber angle for each treatment condition. The initial fiber angles were uniformly distributed at angles from 10 to 55 degrees.

The single treatment fiber angles for the 73 °C and room temperature conditions are 23.5 ± 18.5 and 20.8 ± 8.7 degrees respectively. Instead of a uniform distribution, the treated composites fit a loglogistic distribution; therefore, the mean fiber angles represent an expected value for the fiber angle. The softer matrix at 73 °C had a weak orienting effect—the standard deviation of the distribution is broad at 79% of the mean fiber angle. The treatment at RT was stronger with a standard deviation that is 42% of the expected value.

When the composite is treated twice with ECAE, the mean fiber angle is 23.6 ± 9.0. This is not significantly different from the fiber angle obtained with the first pass at either temperature. In addition, the standard deviation (38% of the mean) is about the same as that found from using a single treatment to align the fibers. This suggests that most of the fiber orientation occurs in a single treatment and that the expected fiber angle is fixed for a given processing temperature and die channel angle.

Conclusions and Discussion

We state these conclusions:

1) Fiber length might be fixed by the treatment conditions and multiple treatments with the same parameters will not reduce the fiber length further.
2) Fiber angle is a strong function of the ECAE process and might be tied to the shear angle of the die.

Our first conclusion suggests that ECAE can obtain a specific distribution of fiber lengths. After one or two treatments, the fibers will be no longer and no shorter than permitted by the treatment conditions.

The second conclusion suggests that composites processed with a 90° die channel angle might need a secondary stretching treatment to orient the fibers to the extrusion axis. An alternative might be to use a die with a larger angle between the entrance and exit channels.

Tables

Table I. Fiber length of the short fiber composite for each process condition.

Process Condition	**Mean Length (μm)**
As Received	105.6 ± 35.6
One ECAE Extrusion at 73 °C	95.0 ± 27.6 μm
One ECAE Extrusion at Room Temperature	81.0 ± 32.2 μm
Two ECAE Extrusions at RT	78.4 ± 26.9 μm

Table II. Fiber orientation of the short fiber composite for each process condition.

Process Condition	**Distribution Function**	**Fiber Angle (degrees)**
As Received	Uniform	10 to 55
One ECAE Extrusion at 73 °C	Loglogistic	23.5 ± 18.5
One ECAE Extrusion at Room Temperature	Loglogistic	20.8 ± 8.7
Two ECAE Extrusions at RT	Loglogistic	23.6 ± 9.0

Illustration

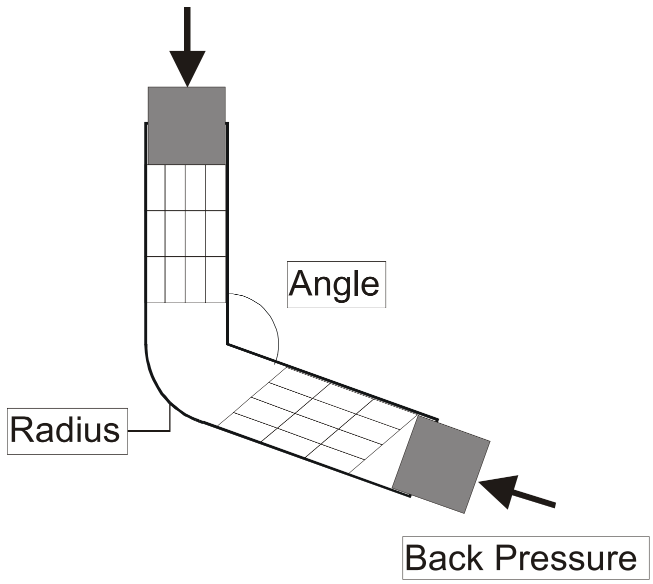

Figure 1. The geometry of an ECAE die. The entrance and exit channels have the same cross section.