PROCESSING METHODS AND PHYSICAL PROPERTIES OF NATIVE GRASS REINFORCED BIOCOMPOSITES

Wanjun Liu[1], Kelby Thayer[1], Manjusri Misra[1], Lawrence T. Drzal[1], Amar K. Mohanty[2]*

[1]Composite Materials & Structures Center, 2100 Engineering Building, Michigan State University, East Lansing, MI 48824 E mail: drzal@egr.msu.edu

[2]School of Packaging, 130 Packaging Building, Michigan State University, East Lansing, MI 48824

Abstract

Big blue stem grass fiber (BBSGF) reinforced thermoplastic biocomposites were fabricated with both extrusion followed by injection molding and sheet-molding compounding (SMC) followed by compression molding. The physical properties were evaluated with dynamic mechanical analysis (DMA), mechanical properties testing and microscopy observation. It was found that compression molding could achieve similar modulus values to injection molding for grass reinforced high density polyethylene (HDPE) composites. The stiffness of compression-molded specimens is related to the consolidation state of the samples, which depends on compression molding conditions such as temperature, pressure and mold type. Compression molded specimens exhibited a higher heat deflection temperature (HDT) and notched impact strength compared to injection-molded samples. Grass fiber reinforced cellulose acetate butyrate (CAB) biocomposites from SMC processing had similar physical properties with grass fiber reinforced HDPE composites, which indicates that natural fiber reinforced CAB biocomposites have the potential to replace polyolefin based composites for automotive applications.

Background

The use of biomass to develop plastic or composite materials for useful applications is becoming a fashion strategy because it is expected to reduce the dependence on non-renewable resources and to alleviate the crisis of energy derived from consumption of non-renewable resources. Agricultural products and byproducts are the main resources of biomass. For example, starch as a biomass has been used to develop bioplastic polylactic acid and polyhydroxylbutyrate; grass, rice straw, wheat straw, and corn stover have become new resources of biofiber because of their low lost and sustainable nature [1, 2]. Composites made from either using bioplastics or biofibers are called biocomposites.

Sheet molding compounding (SMC) is an effective process for fabricating fiberglass-reinforced composites in the manufacturing industry. The most common use of the SMC line is to make a continuous sheet of molding compound from both chopped fiberglass strands and a thermoset polymer resin such as polyester, epoxy and vinyl ester. The sheet material can then be compression molded into a final product. Recently, natural fibers were used in the SMC process in place of glass fibers for making a panel for automotive applications [3,4]. The most important advantages of using natural fibers in SMC are high strength and lightweight [4]. The advantages of the SMC process provide a viable method to fabricate biocomposites. Although processing techniques such as extrusion, injection molding and compression molding have been used to fabricate biocomposites, the SMC process has not been reported in literature for the fabrication of biocomposites from thermoplastic and biofiber.

In this research, big blue stem grass fiber along with thermoplastic high density polyethylene and cellulose acetate butyrate was used to fabricate biocomposites using a slightly modified SMC line. Big Blue Stem Grass Fiber (BBSGF) was used for two main reasons: BBSGF is a native grass of the USA and is easy to separate and feed in the SMC process. Extrusion followed by injection molding and SMC followed by compression molding both have been used to fabricate biocomposites to investigate the influence of processing on their performance.

Experiments

Materials

Big blue stem grass fiber (BBSGF) with 3-6 mm length was received from Smith, Adams & Associates LLC (Okemos, MI). High density polyethylene powder (MICROTHENE® FA709-00) was purchased from EQUISTAR Chemical Company (Houston, Texas). Polyethylene grafted maleic anhydride (PE-g-MA) (EPOLENE G2608) and cellulose acetate butyrate (CAB) (Tenite Butyrate 485E3720016) were supplied by Eastman Chemical Company (Kingsport, TN).

Extrusion

Big blue stem grass fiber and PE-g-MA were dried at 80 °C under vacuum for 16 hours before processing. HDPE was blended with the coupling agent (EPOLENE G2608) according to a ratio of 47/3 and was fed into a ZSK-30 Werner and Pfleiderer Twin-screw Extruder (L/D=30) with a barrel temperature of 190 °C and screw rotation speed of 100 RPM. The feeding rates of both matrix and grass fibers were 23g/min for each giving a ratio of 50 wt %/50 wt %. The extrudate was then pelletized for further processing.

Injection molding

An 85 ton Cincinnati Milacron injection molder with a screw L/D ratio of 17:1 was used to mold dog-bone specimens using the biocomposite pellets produced using the extruder. A barrel temperature of 190 ^{o}C and a mold temperature of 30 ^{o}C were used.

SMC Processing

The Continuous Biocomposite Sheet Molding Compound Panel Process consists of five main components (as shown in Figure 1).

1) The bio-fiber feeder assembly,
2) The high voltage alignment system,
3) The spray nozzle assembly or powder curtain feeder,
4) The infra-red heater bank,
5) The friction grip drive wheels/continuous polytetraflouro ethylene belt drive system.

A continuous glass reinforced Teflon belt is driven by a friction grip drive wheel assembly. The belt first passes underneath the vibratory feeder, which distributes biofiber evenly over the belt at a continuous set output rate creating a fiber mat. The vibratory feeder is fed fiber from a twin-screw feeder. The fibers then pass over the alignment electrodes, which cause the fibers to become aligned via. electrostatic forces. The fibers then pass through a PE powder/H_2O suspension spray or powder curtain feeder (for CAB) that coats them with the matrix material. Sintering is accomplished with an 18 kW infrared heater bank, which has variable temperature control. The semi-consolidated veil material is then removed at the end of the line.

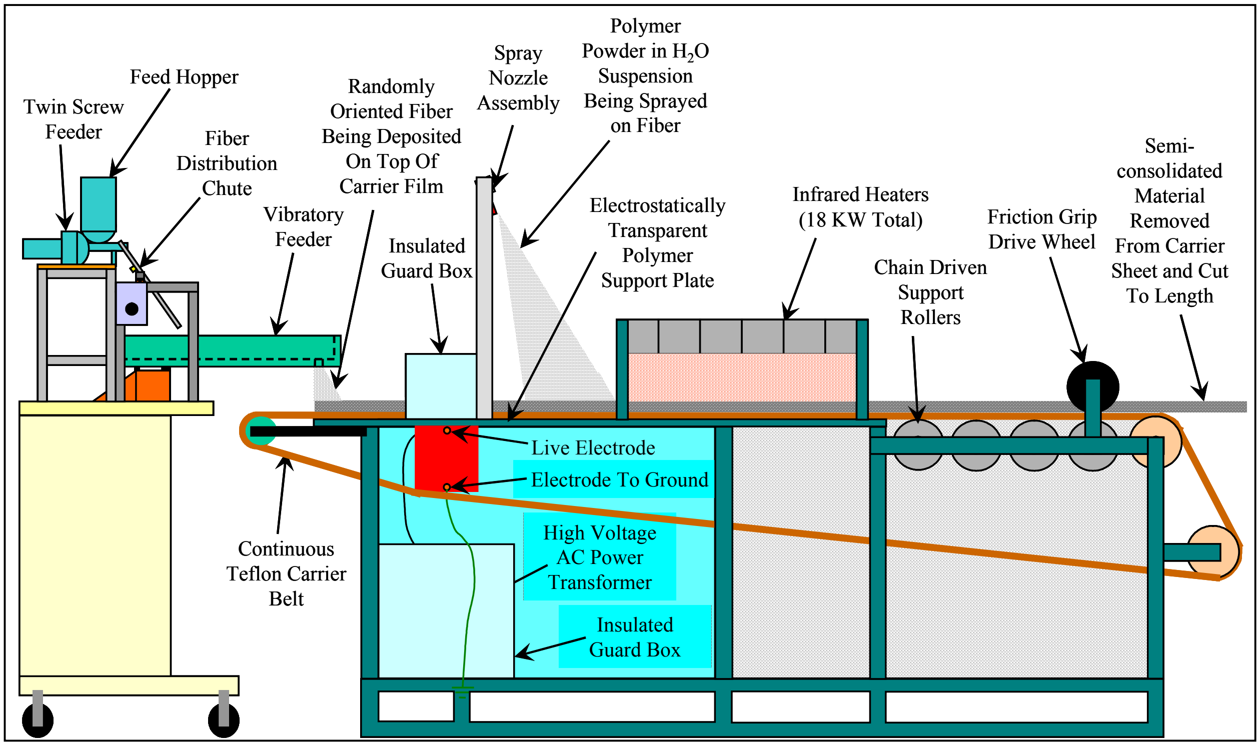


Figure 1. Schematic of continuous biocomposite sheet molding compounding panel (BCSMCP) manufacture process with spray nozzle assembly

For HDPE/BBSGF biocomposite SMC processing, the belt speed was set to 300 mm/min. The speed of the K-tron twin-screw feeder was 500 RPM. Air pressure for the rotary ball vibrator on the fiber distribution chute was set to 517 KPa. Biofiber output was measured to be 11.9 g/min over a 0.3 m wide area. The alignment system was not used for random fiber composites. A suspension of 50 wt % powder to 50 wt % DI water was sprayed on the fiber surface with a peristaltic pump setting of 10 ml/min (approx. 11.85 g of PE powder/min) giving a ratio of fiber/resin of approximately 50 wt %/50 wt %. The infrared heater controller was set to 338 °C (sample surface temperature of 188 °C as measured with an optical pyrometer). The biocomposite semi-consolidated sheet produced by the SMC process is shown in Figure 2.

Figure 2. Semi-consolidated biocomposite sheet produced from SMC processing

Compression molding of big blue stem grass fiber biocomposites

Twelve 50×150 mm^2 rectangles were cut from the semi-consolidated sheet biocomposite. The 12 layers were then put into a closed mold as shown in Figure 3. A temperature of 190 °C was used at a pressure of 2.76 MPa for the first 15 min followed by an increase in pressure as indicated in the following figures for 10min. The sample was kept under the designated pressure until it cooled to 25 °C. The compression-molded panels were then cut into rectangular specimens for thermal and mechanical properties measurement.

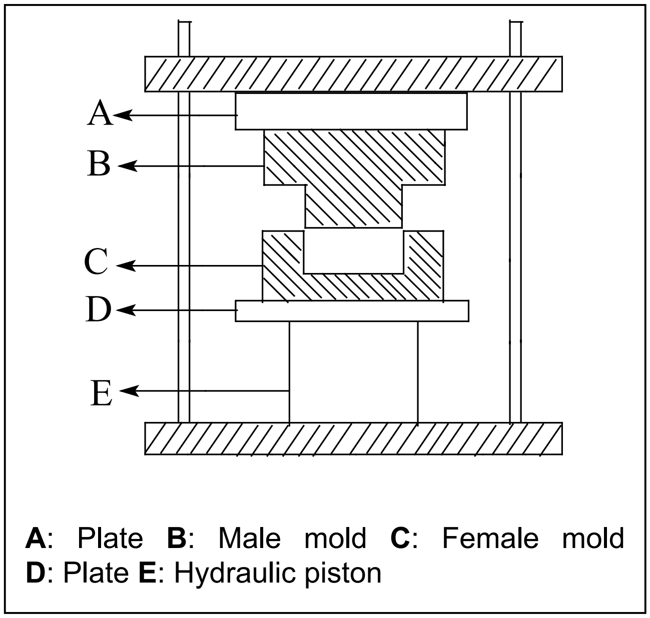


Figure 3. Schematic of compression molder

Dynamic mechanical properties

The dynamic mechanical properties were studied with a dynamic mechanical analyzer (2980 DMA, TA instruments, USA). DMA multi-frequency and three point bending modes were used. A frequency of 1 Hz. and a temperature ramp rate of 4 °C/min from 20 to 100 °C were used.

Density measurement

The density of specimens was measured using Archimedes method.

Heat deflection temperature

A dynamic mechanical analyzer (2980 DMA, TA instruments, USA) was used to measure the heat deflection temperature (HDT) of the biocomposite with a load of 455KPa as specified by ASTM D 648. DMA controlled force mode and three point bending with a span of 50mm were used in the experiments. Heating rate was 2 °C/min.

Impact strength measurement

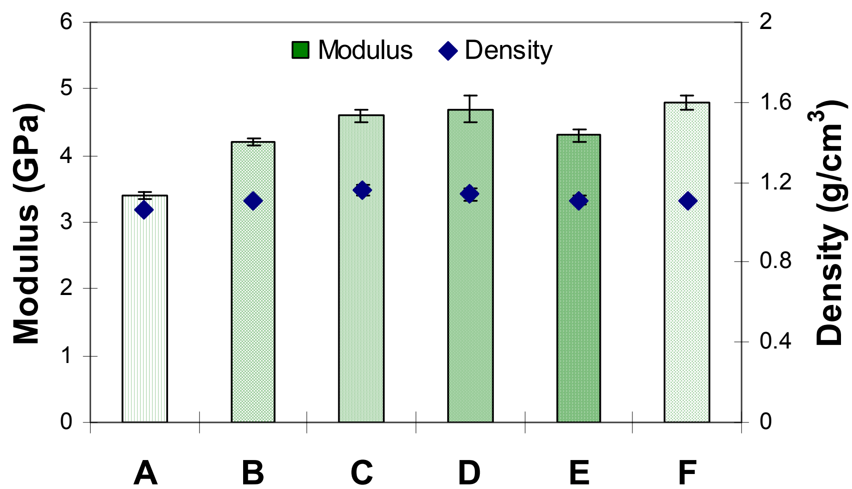
The notched izod impact strength of the composites was measured with a Testing Machines Inc. 43-02-01 Monitor/Impact machine according to ASTM D256.

Environmental SEM

The morphology of the specimens was observed with a Phillips Electroscan 2020 Environmental Scanning Electron Microscopy (ESEM) with an accelerating voltage of 20 kV to check the consolidation of the composites. The surface observed was prepared by cutting with a clean razor blade.

Results and discussion

Figure 4 shows the storage modulus at 25 °C and the density of HDPE/grass composites compression molded from pelletized extrudate. It was found that the composites fabricated with the frame mold (sample A) gave the lowest stiffness value compared to those fabricated with injection molding. It was found that the use of a picture frame mold is not sufficient for composite fabrication because the total applied pressure on the sample is limited by the presence of the picture frame. This results in a drop in the applied hydrostatic pressure to almost zero when the press platens come in contact with the picture frame leading to inadequate consolidation of the composite. Alternatively, a closed matched mold is a good choice for use in compression molding of HDPE/grass extrusion samples because of the higher pressures this type of mold can produce.



A= Compression molded at 180 °C/1.03 MPa with frame

B= Compression molded at 180 °C/6.90 MPa with closed mold

C= Compression molded at 190 °C/6.90 MPa with closed mold

D= Compression molded at 190 °C/5.17 MPa with closed mold

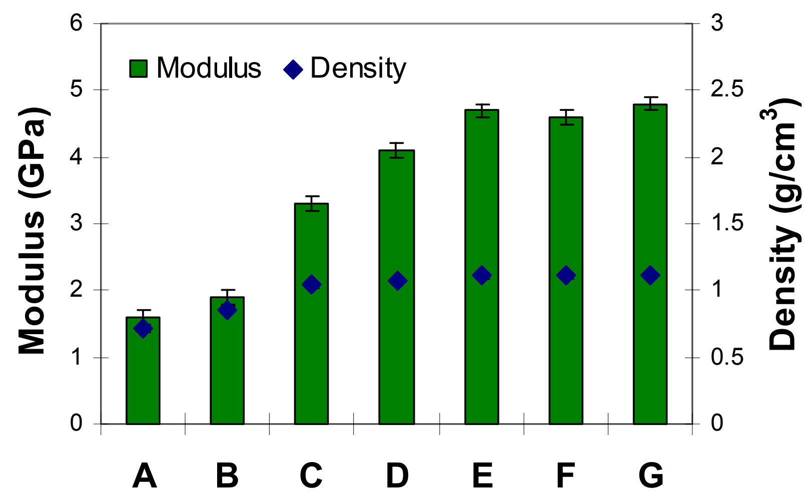
E= Compression molded at 190 °C/3.45 MPa with closed mold

F= Injection molded from pelletized extrudate

Figure 4. Modulus and density of HDPE/grass composites from extrusion by different processing conditions

The composites fabricated with a closed matched mold achieved higher modulus values than those made with the picture frame mold as expected. Values are very close to those achieved with injection molding (sample F). In addition, the stiffness of sample C was higher than those fabricated with lower temperature (sample B) or lower pressure (sample E). These results indicate that the stiffness of the composites is dependent on the mold type (i.e. frame mold and closed mold) and processing conditions (such as temperature and pressure).

In addition, the stiffness was closely related to the density of the composite. This is because the density is directly related to the consolidation state of the final product. The density of the composites plateaued around 190 °C and 5.17 MPa, indicating that closed compression molding produces sufficient pressure for full consolidation. As a result, closed compression molding at the optimum molding conditions of 5.17 MPa and 190 °C produced biocomposite samples with a comparable modulus to injection molded samples.



A= Compression molded at 190 °C/1.03 MPa with frame

B= Compression molded at 190 °C/20.69 MPa with frame

C= Compression molded at 190 °C/3.45 MPa with closed mold

D= Compression molded at190 °C/ 6.90 MPa with closed mold

E= Compression molded at 190 °C/10.34 MPa with closed mold

F= Compression molded at 190 °C/13.79 MPa with closed mold

G= Injection molded from pelletized extrudate

Figure 5. Physical properties of HDPE/grass composites from SMC line

Figure 5 shows the storage modulus at 25 °C and the density of HDPE/grass composites compression molded from biocomposite sheet material made on the SMC line. It was found that the composites fabricated using the picture frame mold had low stiffness even at high pressures (sample B) compared to the injection molded specimen (sample G). In contrast, the composites fabricated with the closed matched mold had a high stiffness similar in value to the injection molded specimen. Among the samples fabricated with the closed mold, the order of stiffness was sample C<D<E=F, indicating that the stiffness of the composites increased with increasing mold pressure. Stiffness plateued at 10.34 MPa and 190 °C (sample E). By optimizing processing conditions and mold type, composites processed with compression molding from SMC sheet material achieved similar stiffness values to injection molding from pelletized extrudate.

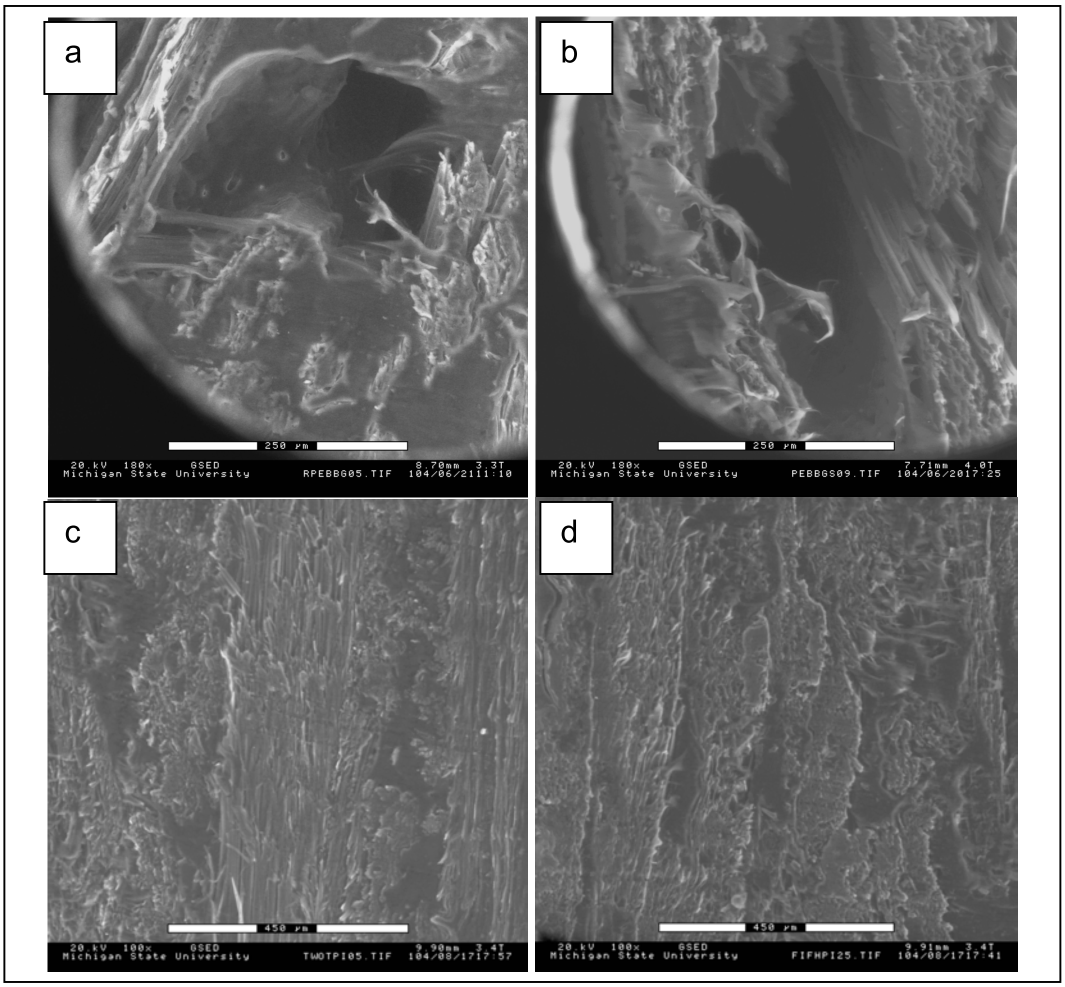


Figure 6. ESEM images of HDPE/grass biocomposites for (a), 1.03 MPa molded sample with picture frame, (b), 20.69 MPa molded sample with picture frame (c), 10.34 MPa/190 °C molded sample with closed mold, and (d), 13.79 MPa/190 °C molded sample with closed mold

As stated above, the stiffness of the composite is closely related to the density, which reflects the consolidation state of the composite. Sample A and B in Figure 5 had lower stiffness and lower density because of poor consolidation which is obvious as voids in E.S.E.M. micrographs a and b as shown in Figure 6. On the other hand, sample E and F in Figure 5 had almost the same stiffness as sample G because of good consolidation which is evidenced by a lack of voids in E.S.E.M. micrographs c and d shown in Figure 6. It is clear from these micrographs that higher pressures effectively pack the fiber and matrix and produce a composite with a higher density and significantly improved mechanical properties.

A comparison of the physical properties of compression molded SMC samples to samples injection molded from pelletized extrudate is shown in Figure 7. Modulus values of samples from SMC are essentially equal to the injection molded samples from extrusion. The impact strength of compression molded samples from SMC is higher than that of injection molded samples. Also, at elevated temperatures, the modulus of compression molded samples is higher than that of injection molded samples. This suggests that the modulus decrease rate of compression-molded samples is lower than that of injection molded samples, which is evidenced by the fact that compression molded samples had higher HDT values than the injection molded samples. The main reason for the higher impact strength is that the fiber bridging effect dominates the impact strength of fiber reinforced composites. Injection molding reduces fiber length through shear degradation but compression molding preserves fiber length. The absence of long fibers in the injection molded samples is the primary reason for the lower impact strength. The compression molding process creates less fiber damage during processing and improves physical properties such as impact strength and HDT.

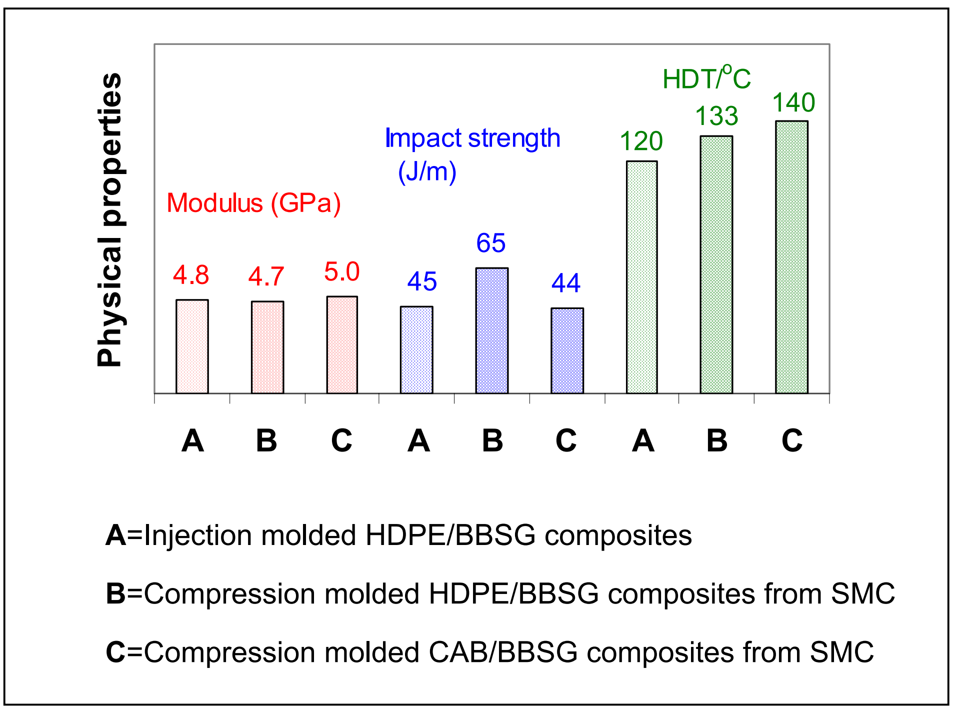


Figure 7. Comparison in physical properties of 50%BBSG/HDPE and 50%BBSG/CAB biocomposites

The dynamic modulus of the CAB/grass composite was found to be higher than that of HDPE/grass composites from SMC as shown in Figure 7. However, the CAB/grass composites had lower impact strength and similar HDT compared to the HDPE/grass composites. Impact strength of the CAB/grass was, however, essentially equivalent to that of the injection molded sample. These results indicate that SMC and subsequent compression molding processing is a viable processing method for manufacturing fully biobased biocomposites with comparable performance to common plastic composites from cellulose acetate butyrate bioplastic and grass biofiber.

Summary

SMC sheet material made from native grass reinforced biocomposite has been successfully made using a slightly modified SMC line. Subsequent compression molding of the SMC sheet material has produced biocomposites with improved physical properties such as modulus at higher temperatures, greater impact strength, and a higher heat deflection temperature compared to biocomposites injection molded from pelletized extrudate. The use of a closed mold in compression molding significantly improved the properties of the biocomposites because of the excellent consolidation achieved with the higher pressures produced by this type of mold. Compression molding SMC sheet material made from cellulose acetate butyrate/natural fiber biocomposites was also shown to be effective and produced panels that had similar impact properties and better modulus and HDT values than petroleum based plastic/natural fiber composites.

Acknowledgement

The financial support from USDA-NRI (Grant No. 2001-35504-10734) and NSF-EPA (Grant No. 2001-DMI-0124789) is gratefully acknowledged for this research. The Authors would also like to thank Eastman Chemical Company (Kingsport, TN), EQUISTAR Chemical Company (Houston, TN), and Smith Adams & Associates LLC (Okemos, MI) for their supply of cellulose ester, high density polyethylene, and grass fiber, respectively.

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Key words: Grass fiber, Biocomposites, SMC, Impact strength, Modulus, and HDT**.**