Journal of Polymers and the Environment, Vol. 13, No. 2, April 2005 (© 2005)
DOI: 10.1007/s10924-005-3211-x

Novel Biocomposites Sheet Molding Compounds for Low Cost Housing Panel Applications

Geeta Mehta,[1] Amar K. Mohanty,[2] Kelby Thayer,[1] Manjusri Misra,[1] and Lawrence T. Drzal[1,3]

Biocomposites were made by a novel high volume processing technique named 'biocomposite sheet molding compound panel' (BCSMCP) manufacturing process. This process design was inspired by the commercial glass fiber–polyester resin composite fabrication method called sheet molding compounding (SMC). This process yields continuous production of biocomposites on a large scale, and thus can be easily adopted in industries. A unique fiber dispersion method, which enabled uniform distribution of natural fibers, was used in this process. Consistency of the process was tested by evaluating the repeatability of the resultant materials' mechanical properties. The low cost biocomposites produced as a result of the processing will be used for various panel applications such as housing and transportation. The molded samples were tested for various mechanical and thermal properties, in accordance with ASTM procedures. The biocomposites were made with various natural fibers including, big blue stem grass, jute, and industrial hemp. By combining different natural fibers in varying mass fractions, hybrid biocomposites were made using this process. Grass fiber reinforced polyester biocomposites processed by the SMC line showed very promising results.

KEY WORDS: Biocomposites sheet molding compound panel (BCSMCP); unsaturated polyester resin; natural fiber reinforced thermosets; thermoset resins; natural fiber.

INTRODUCTION

Natural and wood fiber plastic composites have continued their phenomenal growth in 2002. As per the recent market study by Principia Partners, the demand for these products in North America and Western Europe combined will reach nearly 1.3 billion pounds valued at roughly $900 million. This represents a growth of almost 20% from 2001 levels. There is a growing interest in the use of natural/biofibers as reinforcing components for thermoplastics and thermosets. Although thermoplastics have the added potential advantage of recyclability, thermosets are capable of superior mechanical properties compared to thermoplastics in the resulting biocomposites. Biocomposites derived from natural fibers and petroleum-based thermoplastics or thermosets are not fully environmentally friendly because matrix resins are non-biodegradable. However, these biocomposites do possess a balance between economics and environment allowing them to be considered for applications in the automotive, building, furniture and packaging industries. Natural fiber composites are mainly price-driven commodity composites that have useable

---

[1] Composite Materials and Structures Center, Department of Chemical Engineering and Material Science, Michigan State University, 2100 Engineering Building, East Lansing, MI 48824, USA.

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

[3] To whom all correspondence should be addressed: E-mail: drzal@egr.msu.edu

1566-2543/05/0400-0169/0 © 2005 Springer Science+Business Media, Inc.

structural properties at relatively low cost. Advantages of natural fibers over traditional reinforcing fibers such as glass and carbon are: low cost, low density, high toughness, acceptable specific strength properties, ease of separation, enhanced energy recovery, carbon dioxide sequesterization and biodegradability [1].

However, the hydrophilic nature of biofibers is a potential cause for incompatibility, adhesion and dispersion problems with hydrophobic polymer matrices. Since mechanical properties of the composites are related to the compatibility and interaction between the components, improvement of the interface and interphase interactions in natural fiber/polyester composites is essential [2]. Approaches to improve the interaction and thereby the stress transfer between lignocellulosics and polyester resin include, for instance, the use of chemical or physical modifications on the fiber. Surface modification of natural fiber with adequate modifiers has been shown to improve compatibility of the natural fibers with hydrophobic plastics.

Biocomposites can be designed and engineered from natural/biofibers and an unsaturated polyester resin/blend of polyester resin and derivative vegetable oil to replace existing glass fiber–polyester composites for use in housing structures. Natural fiber composites (biocomposites) would provide environmental gains, reduced energy consumption, lighter weight, insulation and sound absorption properties and would also eliminate health hazards unlike glass fiber composites thus providing many beneficial additions to the next generation of housing technologies.

Biocomposites have been traditionally manufactured with techniques like extrusion, injection molding, compression molding, resin transfer molding, and pultrusion, etc., but they have never been produced using an SMC line. SMC is a very useful processing technique usually used for fabricating glass–polyester resin composites. Today more and more parts and products from cars to skis, hockey sticks to microwaves are being molded from SMC. This is not too surprising since thermoset composites and plastics have been used for more than 60 years around the world. A recent US report states that the demand for thermoset composites in the automotive industry will rise by 68% to 467 million pounds by the middle of this decade. SMC combines characteristics of high strength and lightweight [3]. Other advantages of thermoset composites are durability and resistance to corrosion in a wide range of temperature environments. The application of a primer and topcoat will allow SMC components to meet automotive 'class A' surface finish requirements.

Ever since the 1960s there have been numerous patents on SMCs [4]. There have been many improvements in the SMC process since then, hence many more patents, but none of these cater to SMCs with natural fibers and resins. However, there is one patent and two papers which talk about SMC processing for biocomposites. These include, US 2003088007 [6] granted in 2003 to Wool et al., which is a patent for sheet molding compound resins from plant oils. van Voorn et al. [7] discussed a non-automated SMC process for making biocomposites in, "Natural fibre reinforced sheet molding compound", in 2001. Goswami et al. [8], made jute (cloth) reinforced sheets with the help of polyester resin in "Jute reinforced sheets based on shellac filled SMC", in 2003.

Authors' sheet molding process is different from all mentioned above because of its uniqueness of fiber dispersion, ability to run the process for any kind of natural fiber, automated fiber feeding to the production line, and flexibility with resin formulations.

EXPERIMENTAL

SMC Line Description

Figure 1 shows the schematic of the proposed processing. The process begins with an upper and lower supply roll of carrier film material. The upper and lower carrier film is fed beneath the upper and lower resin pots, respectively, which deposits a predetermined thickness of resin onto the carrier film by means of adjustable height wipe blades. The lower carrier sheet then passes beneath the biofiber vibratory feeder, which deposits a calibrated weight % of biofiber uniformly onto the surface of the resin. The upper and lower carrier sheets are then brought together creating a sandwiched SMC sheet material. The sheet material is then fed through compaction rollers to help provide a uniform sheet cross section and also to provide some mixing forces to ensure the even distribution of fiber in the resin. Grip wheels near the end of the line provide the necessary mechanical force to pull the sheet material through the system at a controlled rate. The sheet material is then cut to the desired length.

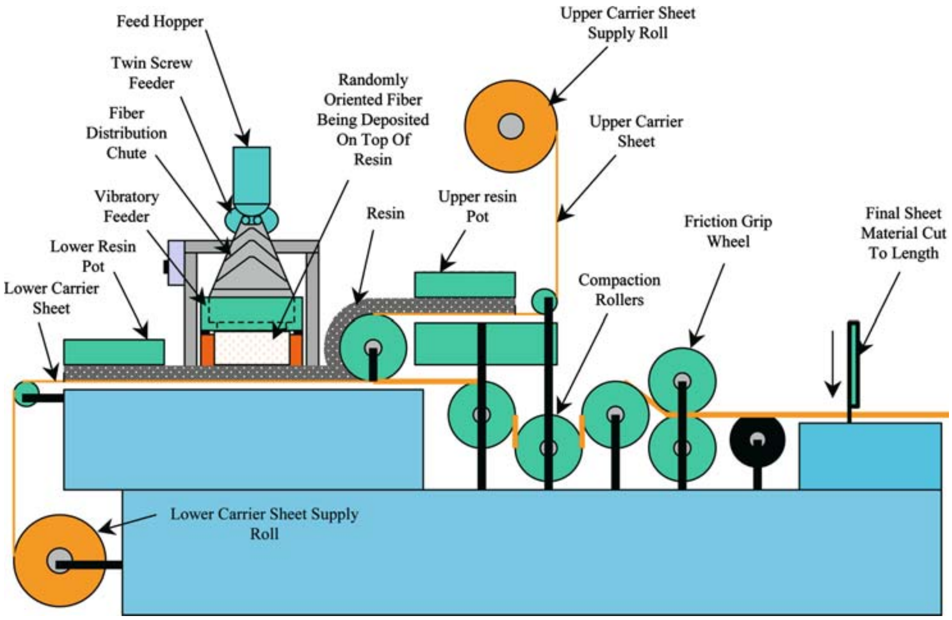

Fig. 1. Scheme of the BCSMCP process.

Feeder Description

The biofiber feeder system consists of four main components:

1. The material feed hopper
2. The twin-screw feeder
3. The fiber distribution feed chute
4. The vibratory feeder

Biofiber is put in the hopper for feeding. The fiber is then fed onto the fiber distribution feed chute by the twin-screw feeder, which is calibrated to output the required fiber weight/minute. The fiber distribution chute spreads the fiber out uniformly from the width of the twin-screw feeder to the width of the vibratory feeder. The vibratory feeder then conveys the fiber to the opening at the end of the feeder tray providing the uniform distribution of fiber onto the SMC sheet.

Materials

The fibers used for making composites in the SMC line were: industrial raw hemp (retted hemp, average length 15–25 mm from HempLine, Ontario, Canada), big blue stem grass (BBSG) (leaf and stem, 4 mm, from Smith, Adams & Associates LLC, Okemos, MI), Hessian jute mats (IJIRA, Calcutta, India) and chopped E-glass (average length 10 mm, Number 985 from Owens Corning). The resin system used was ortho unsaturated polyester (UPE) (Kemlite Company Inc., Joliet, IL) with methyl ethyl ketone peroxide (MEKP, Sigma Aldrich) catalyst (0.5% by weight of resin), and cobalt naphthenate (CoNap, Sigma Aldrich) promoter (0.03% by weight of resin). The silane used for treating big blue stem fibers was methacryloxypropyltrimethoxy silane (γ-MPS) (SIM6487.4 obtained from Gelest Inc. Tullytown, PA). Ethanol was procured from J.T. Baker. Glacial acetic acid was purchased from Sigma Aldrich. All materials were used as such without further purification.

Processing

Experiments were conducted to produce biocomposites using the SMC line. The major goal of these runs was to produce sheet material using natural fibers as the reinforcement. The exact details of these experiments are given elsewhere [9]. Some composites were made with untreated fibers and some were made with chemically treated fibers. In particular, silane treatment was used on big blue stem grass. The silane treatment of these fibers was done in the following way: 1% methacryloxypropyltrimethoxy silane (γ-MPS) solution was made in 99% deionised water and ethanol (1:1), maintaining the pH of the solution at 4.0 by using 2% Glacial Acetic Acid. The silane solution was hydrolyzed for 2 hours by continuously stirring using a magnetic stir bar. The fibers were soaked in this solution for 1 hour. The solution was then drained from the fibers, and they were dried under the hood for at least one day. They were then cured in an air oven for 5 hours at 80°C. The fibers were then dried in a vacuum oven at 80°C and −30 in Hg.

When two different fibers were used for making hybrid biocomposites, the fibers were mixed well by continuous agitation, and the resulting uniform mixture was vacuum dried before using it to make biocomposites. The natural fibers also had to be introduced into the SMC line in a different way as compared to glass fibers. They were fed by a combination of twin-screw feeder and vibratory feeder. This way of fiber introduction was optimized after various attempts with vibratory feeding. All natural fibers (treated/untreated) were vacuum dried at 80°C and −30 in Hg for 5 hours, before fabrication of any composites. The resulting biocomposites contained 20 wt% fiber, 30 wt% $CaCO_3$, and 60 wt% UPE. This composition is different from that of glass composites, because, natural fibers have a tendency to soak up resin.

Procedure

A total of 6560 gm of resin was mixed shortly before the SMC run using the following formulation: 60% unsaturated polyester resin—5182.76 gm, 30% calcium carbonate filler—1350.00 gm, 0.03% cobalt naphthanate promoter—1.56 gm, 0.5% methyl ethyl ketone peroxide (MEKP)—26.06 gm.

The Ross mixer speed dial was set to 5.0. Cooling water to the mixing pot was connected and turned on. The polyester and calcium carbonate were combined and mixed for 10 minutes. The cobalt naphthanate was added and mixed for 5 minutes. Finally, the MEKP was added and mixed for 5 minutes. Resin was transferred into a 3.5 gal epoxy coated steel storage container.

The SMC line was turned on shortly before the upper and lower resin pots were filled and was run at a speed of 0.3 in/sec. Resin was then transferred to the resin pots. The K-tron twin-screw feeder and vibratory feeder were then turned on for feeding fibers. The process was allowed to run to see what the product looked like after the compaction rollers. The modified feeder setup distributed fiber evenly and consistently. Several feet of product were produced at these settings. The resultant sheet material looked very good. The finished product was cut into 1 ft long segments and was placed flat in a sealed polyethylene container. This container was labeled with the date and time and was immediately put into a freezer for storage. After cooling in the freezer for several hours, the sheet material was compression molded in the Carver press for making samples for testing and characterizations. The prepreg samples were placed between two aluminium plates pre-coated with teflon. The plates were pressed in a compression molding press (Carver Lab Press) at around 80 psi for 2 hours at 100°C followed by 2 hours at 150°C. The resulting composite is then cut into desired shapes for various tests. For making the control sample, a degassed UPE solution is poured into degassed silicone molds and cured in a conventional oven at the same curing conditions.

Analysis

The compression molded biocomposites and control samples are used for tensile, flexural and notched Izod impact tests complying with ASTM D638, ASTM D790 and ASTM D256 standards, respectively. A United Calibration Corp SFM-20 testing machine was used for tensile and flexural testing. The impact testing was carried out using a model #43-OA-01 impact tester from Testing Machines Inc. A Dynamical Mechanical Analyzer (TA DMA 2890) was used to measure the storage modulus, loss modulus and tan delta. For DMA testing, rectangular bars, 50 mm × 12 mm × 3 mm are placed on the 3 point bending fixture in the furnace and heated at 4°C per minute from room temperature to 150°C.

RESULTS AND DISCUSSION

The tensile properties of biocomposites were higher than that of neat resin (figure not shown). The tensile strength of silane treated BBSG biocomposite is 29% more than the strength of untreated BBSG biocomposite. The strength of untreated jute−hemp (20 wt%) hybrid biocomposite is 35% higher than that of untreated BBSG biocomposite. The strength of chopped E-glass composite is 170% higher than that of untreated BBSG biocomposite.

The chopped E-glass composite had the highest tensile strength and modulus. The untreated jute−hemp (20 wt%) hybrid biocomposite had the second highest tensile strength and modulus.

In the case of tensile modulus, silane treated BBSG biocomposite has a modulus value 12%

greater than that of untreated BBSG biocomposite. The tensile modulus of untreated jute–hemp (20 wt%) hybrid biocomposite is 20% higher than that of untreated BBSG biocomposite. The modulus of chopped E-glass composite is 100% higher than that of untreated BBSG biocomposite.

The comparison of specific tensile strength (tensile strength divided by density of composite) and specific tensile modulus (tensile modulus divided by density of composite) of SMC produced biocomposites containing calcium carbonate is shown in Fig. 2. The bars represent tensile strength and the points denote tensile modulus.

The specific tensile strength of silane treated BBSG based biocomposite was 30% higher than that of untreated BBSG based biocomposite. The specific strength of E-glass based composite was 36% higher than that of untreated BBSG based biocomposite, 5% higher than silane treated BBSG based biocomposites, and almost same as jute–hemp hybrid fiber based biocomposite.

In terms of specific tensile modulus, the silane treated BBSG based biocomposite was 12% higher than that of untreated BBSG based biocomposite. The specific tensile modulus of E-glass based composite was 3% higher than that of untreated BBSG based biocomposite, while it was 8% lower than silane treated BBSG based biocomposites, and 13% lower than that of jute–hemp hybrid fibers based biocomposite. Thus it was seen that the specific tensile strength of biocomposites and hybrid biocomposites is in the same range as the E-glass based composite. However, in terms of specific tensile modulus biocomposites and hybrid biocomposites produced by SMC are better than the glass fiber based composites.

Table I shows the impact strength of the SMC produced biocomposites containing calcium carbonate. The impact strength of silane treated BBSG-UPE-$CaCO_3$ is 65% less than the strength of untreated BBSG-UPE-$CaCO_3$. The impact strength of untreated jute–hemp (20 wt%) hybrid biocomposite is 86% higher than that of untreated BBSG biocomposite. Also, the impact strength of chopped E-glass composite is 13.3 times higher than that of untreated BBSG biocomposite.

The impact strengths of the composites followed a pattern completely opposite to that of bending and tensile strengths. This is a common behavior for fiber-reinforced plastics. The highest impact strength was of the samples containing chopped E-glass composite, followed by that of untreated jute–hemp (20 wt%) hybrid biocomposite. The impact strength of composites containing BBSG was very small because these fibers were very short in length. In particular, the length of BBSG fibers was about 4 mm, while the length of chopped E-glass was about 10 mm.

The storage modulus of SMC produced biocomposites containing calcium carbonate are shown in Fig. 3. The storage modulus of silane treated BBSG-UPE-$CaCO_3$, at 40°C, is 18% more than that of untreated BBSG-UPE-$CaCO_3$. The storage modulus of untreated jute–hemp (20 wt%) hybrid biocomposite is 45% higher than that of untreated BBSG biocomposite. The modulus of chopped E-glass composite is 105% higher than that of untreated BBSG biocomposite.

The data for storage modulus followed the same trend as tensile modulus and modulus of elasticity. The highest storage modulus at 40°C was recorded for samples containing chopped E-glass. The biocomposite with untreated jute–hemp (20 wt%) hybrid had second highest storage modulus at 40°C.

As can be seen from the mechanical and thermal properties, chemical treatment of the natural fibers produced better composites. Biocomposites had almost the same properties as E-glass composites in terms of specific strength and stiffness. The impact strength of BBSG based composites was low due to short length of the fibers compared to that of glass fibers. Better mechanical properties of biocomposites could be achieved by increasing the length of grass fibers. The hybrid biocomposite containing hemp and jute fibers is promising in tensile, impact as well as thermal properties. The relevant mechanical properties of this biocomposite can be further improved by surface treatment of hemp and jute fibers.

However, it must be emphasized that the physical properties of natural fibers are strongly influenced by their chemical structure such as cellulose content, degree of polymerization, orientation and crystallinity, which are affected by the plant genetic makeup, conditions during growth of plants as well as extraction methods used. As a result, there is an enormous variability in fiber properties [10]. Bast fibers generally have a lower microfibrillar angle, and higher cellulose content leading to higher values of tensile modulus compared to bast fibers. Higher cellulose content in grasses leads to higher mechanical properties. Leaf fibers generally have a higher microfibrillar angle, leading to lower values of

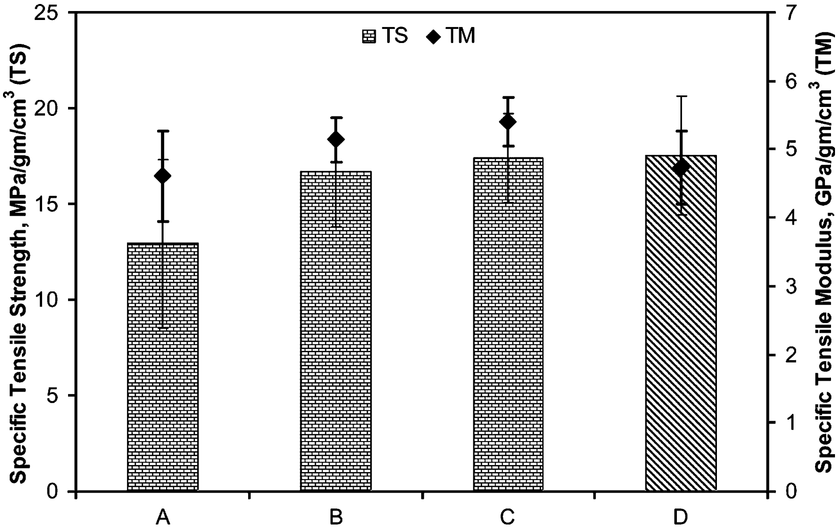

Fig. 2. Tensile properties of composites. Legend: A = Untreated BBSG (20%)-$CaCO_3$-UPE (SMC); B = Silane treated BBSG (20%)-$CaCO_3$-UPE (SMC); C = Jute–Hemp (20 wt%)-$CaCO_3$-UPE (SMC); D = Chopped glass (20 wt%)-$CaCO_3$-UPE (SMC).

tensile modulus compared to bast fibers. Thus, one surface treatment which improves the mechanical properties of a leaf fiber might not have the same effect on a bast fiber.

Table I. Impact Properties of Composities

Sample	I.S. (J/m)	SD (J/m)
A	9.3	2.7
B	13.5	1.9
C	8.1	1.9
D	20.2	3.9
E	192.3	21.2

I.S., impact strength (J/m); SD, standard deviation (J/m).
Legend: A = UPE control; B = Untreated BBSG (20%)-$CaCO_3$-UPE (SMC); C = Silane treated BBSG (20%)-$CaCO_3$-UPE (SMC); D = Jute–Hemp (20 wt%)-$CaCO_3$-UPE (SMC); E = Chopped glass (20 wt%)-$CaCO_3$-UPE (SMC).

CONCLUSIONS

Biocomposites have been successfully made using natural fibers and unsaturated polyester resin by sheet molding compound panel processing. These biocomposites were made in the same SMC equipment used to fabricate glass–polyester

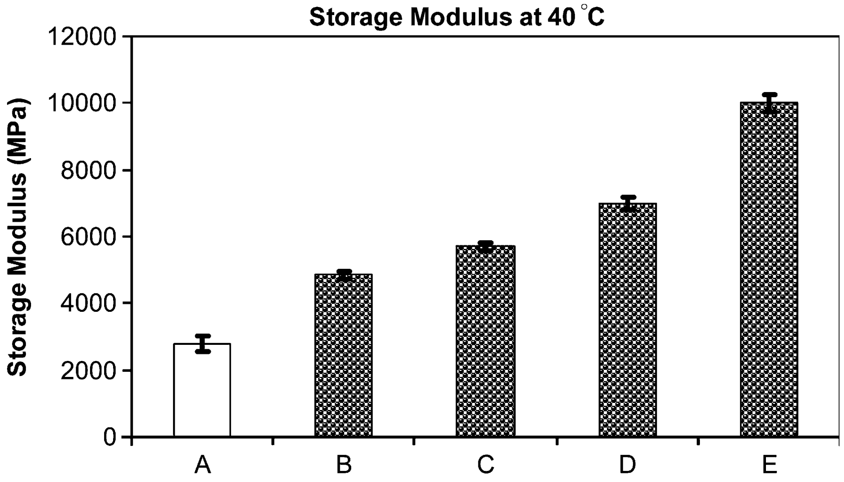

Fig. 3. Storage modulus of composites at 40°C. Legend: A = UPE control; B = Untreated BBSG (20%)-$CaCO_3$-UPE (SMC); C = Silane treated BBSG (20%)-$CaCO_3$-UPE (SMC); D = Jute–Hemp (20 wt%)-$CaCO_3$-UPE (SMC); E = Chopped glass (20 wt%)-$CaCO_3$-UPE (SMC).

composites. As a comparison, we also fabricated glass–polyester composites on this equipment. The biocomposites were made on this SMC line after a few minor adjustments. Instead of using the traditional fiber feeding system, we used a screw feeder and a vibratory feeder to supply natural fibers to the setup. Repeatable results were obtained showing that this process is consistent and can be used for fabrication of biocomposites. In the future, bioresin will be used instead of polyester resin as a matrix for these systems. We have also found that glass–UPE composites have almost the same specific strength and modulus as those of natural fiber–UPE composites. But, with optimization of the entire BCSMC process, use of engineered natural fibers, and inclusion of desirable additives, we seek to achieve best mechanical, thermal and physical properties as comparable as to glass based SMC, and thus replace/substitute glass–UPE composites with natural fiber biocomposites.

ACKNOWLEDGMENTS

Authors are grateful to National Science Foundation–Partnership for Advancing Technologies in Housing (NSF-PATH) 2001 Award No. 0122108 for financial support. The collaborating efforts and many fruitful discussions with, as well as fiber and resin samples from, our industrial partners Flaxcraft Inc., Cresskill, NJ and Kemlite Inc., Joliet, IL are gratefully acknowledged. Authors are also thankful to Smith, Adams & Associates LLC, Okemos, MI and Hempline, Ontario, Canada for grass fiber and hemp fiber samples.

REFERENCES

1. A. K. Mohanty, M. Misra, and G. Hinrichsen (2000) *Macromol. Mater. Eng.* **276/277**, 1–24.
2. A. K. Mohanty, M. Misra, and L. T. Drzal (2002) *J. Polym. Env.* **10**, 19–26.
3. A. K. Mohanty, M. Misra, and L. T. Drzal (2001) *In* Proceedings of the 24th Annual Meeting of the Adhesion Society—Adhesion Science for the 21st Century, February 25–28, pp. 418–420.
4. Composite Industry News Letter (2000), **10**(2), March 7.
5. G. Mehta (2004) *Biocomposites from Engineered Natural Fibers and Unsaturated Polyester Resin for Housing Panel Applications*, Michigan State University, Masters Thesis.
6. *US 2003088007*, (2003) Wool, Richard P., Lu, Jue, Khot, Shrikant N., Sheet molding compound resins from plant oils.
7. H. H. G. Voorn van, R. J. Smit, B. Sinke, and de Klerk (2001) *Composites Part A*. **32**, 1271–1279.
8. D. N. Goswami, P. C. Jha, K. Mahato, and K. K. Kumar (2003) *Popular Plastics & Packaging* **48**(3), 68–71.
9. Biocomposites Sheet Molding Compounds and methods of making those, L. T. Drzal, G. Mehta, M. Misra, A. K. Mohanty, and K. Thayer (US Patent Pending, 2003).
10. McGovern JN (1987) in *Encyclopedia of Polymer Science and Engineering*, Vol. 7 (H. F. Mark, N. M. Bikales, C. G. Overberger, G. Menges, and J. I. Kroschwitz, Eds.), John Wiley & Sons, New York, 16 pp.