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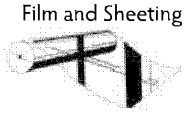

Article

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Improvements in processing characteristics and engineering properties of wood flour-filled high density polyethylene composite sheeting in the presence of hollow glass microspheres

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Abstract

Hollow glass microspheres were introduced into wood flour/high density polyethylene composites by melt compounding in a twin-screw extruder. The prepared composites were subsequently converted to extruded profiles in order to obtain composite sheeting. The presence of hollow glass microspheres highly reduced the density of the extruded sheets down to 0.91 g/cc, while improving its flexural modulus. The presence of hollow glass spheres further improved the visual appearance by eliminating warpage. Thermal conductivity of the sheets was reduced down to 0.25 W/mK without significantly changing the melt viscosity. The morphological analysis indicated a satisfactory state of dispersion of hollow glass microspheres in the sheeting. The presence of hollow glass

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microspheres resulted in sharper contours of the extruded profiles and improved nailability and screwability.

Keywords
sheet, hollow glass microspheres, extrusion, thermal conductivity, density

Introduction

Polymer composites reinforced with natural fibers have been technologically important materials in sheeting applications due to their relatively higher strength, moderate cost, and reduced environmental impact.[1–4] Improving the engineering properties of these composites, such as increasing the stiffness[5] and reducing the density,[6] has also been an active research field. A major challenge in design of wood/polymer composites is to obtain a balanced improvement in all major engineering properties without inducing significant reduction in one or more properties, such as in mechanical strength or processability.[7] Wood flour, in its as received form, is a low density filler (below 0.7–1 g/cc depending on type) that can be incorporated into low-melting polyolefins such as polyethylene and polypropylene. One issue has been the increase in the density of the wood flour from below 1–1.4 g/cc, when it is extruded or injection molded with polymers. This is due to compaction of the cellulosic walls by the compressive forces experienced during plastic processing. Increase in density of wood in processed plastics eliminates certain uses of these wood materials that require floatation in water. It also makes it very hard for the installer to nail and drill through the profiles. In order to attain real wood experience, decrease in the density of processed polymer wood composites has been exercised by foaming via chemical blowing agents. Foaming is known to decrease certain mechanical properties of wood such as its flexural strength. Water uptake in gas-foamed polymer wood products has also been a challenge, once water finds its path to the open pores. One alternative to foaming is the use of hollow glass microspheres (HGMs).

The HGMs are finely dispersed, free-flowing powders consisting of thin-walled (0.5–1.5 μm) spherical glass particles with an average diameter of 15–65 μm. These inorganic microadditives exhibit low densities (0.12–0.6 g/cc) and primarily used for weight reduction in plastics. They could improve a series of engineering properties of polymers and wood/polymer composites, such as density, tensile, and flexural modulus, thermal conductivity. Their presence usually exerts minimal impact on processing behavior due to their low aspect ratio (~1). The HGMs have been incorporated into liquid thermoset polymers as fillers for decades.[8] Until recently,

however, their use with thermoplastic polymers has been limited because of high rates of bubble breakage during thermoplastic processing operations as extrusion compounding and injection molding. However, the recently developed light weight, yet strong HGMs offer resistance to extremely high compressive and shear forces (Figures 1 and 2). In this work, we analyzed the improvements in engineering properties of wood/polymer composite sheets in the presence of HGMs.

Experimental

Materials

High density polyethylene (HDPE) homopolymer was obtained in the form of pellets from Channel Prime Alliance under the trade name Certene HGB-0760 with a density of 0.96 g/cc and with a melt flow rate of 0.7 g/10 min (at 190°C and under a load of 2.16 kg, according to ASTM D1238). Note that HDPE has been widely used in pristine and natural fiber-filled polymeric sheets.[9] Struktol® TPW 104, a blend of lubricants designed specifically for wood fiber/flour-filled polyolefins was used as the lubricant. Wood flour was Ponderosa pine wood flour (grade 4020) from American Wood Fibers. The 3 M™ HGMs, iM30K and K42HS, were used in extruded profiles.

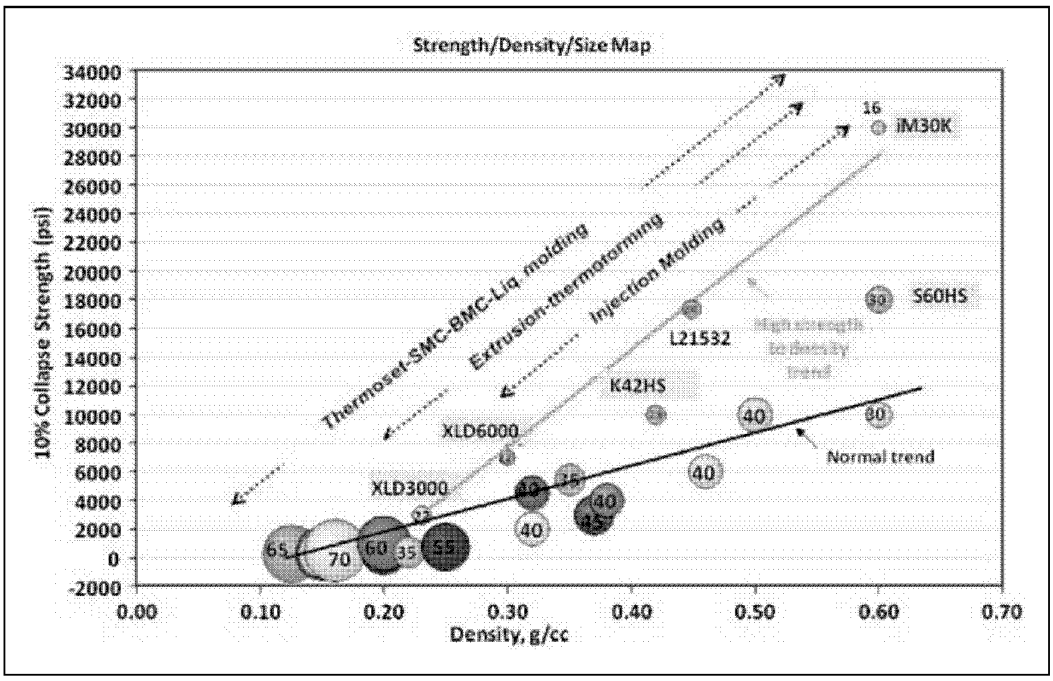

Figure 1. The strength vs density of commercially available 3 M™ hollow glass microspheres.

Sample preparation

The HGM-filled wood/HDPE composites were prepared in two steps (Figure 3). First, a precompound formulation without any HGMs was prepared in a twin-screw extruder. The wood flour and lubricant was dry blended and side stuffed, while the HDPE was added through the main feed throat. The strands were pelletized into precompound pellets. In the second stage, the precompound pellets were introduced in the main feed throat, while the HGMs were side stuffed into the extruder using the desired formulations. The screw speed of the extruder was set to 50 r/min. The temperatures of the zones ranged from 360 to 340 F. Profiles were extruded at 12 in/min (18 lbs/h) through a 2¼ × ¼ in die, water cooled, and dried at ambient temperature. The composition of the prepared samples is presented in Table 1.

Characterization

Density, glass bubble loading and survival rate measurements. In order to determine the amount of HGM compounded into the wood/HDPE composite and the volume loss due to HGM breakage, the compounded pellets were exposed to high temperatures in a Habertherm oven in order to volatilize the

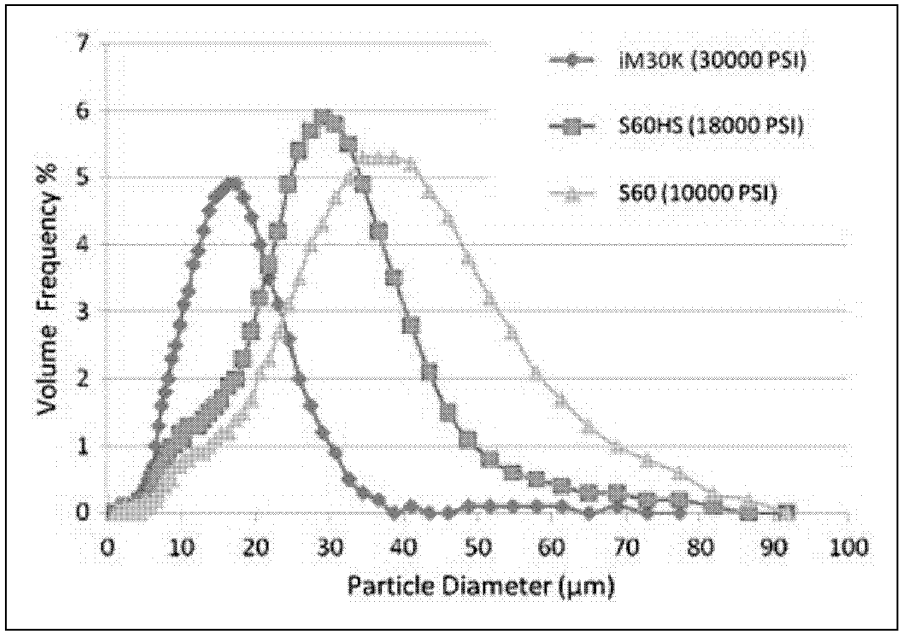

Figure 2. The size distribution of representative hollow glass microsphere (HGM) grades.

polymer resin and wood fibers. The temperature in the oven was set to a ramp profile to run from 200 to 550°C in 5 h. After the temperature reached 550°C, it was kept constant for 12 h. The amount of inorganics and HGMs were calculated from the known weights of wood/HDPE/HGM composite before and after burn process. In order to determine the amount of volume loss due to bubble breakage, the density of the residual material after burn off, which is mainly HGMs, was determined using a helium gas pycnometer, AccuPcy 1330 from Micromeritics. The density values of wood composite samples (dry and wet) were also determined by the gas pycnometer.

Morphological analysis. The scanning electron microscope (SEM) images were generated using a FEI–SEM system (http://www.fei.com/company/). The HGM diameters were measured using measureIT program.

Thermal properties. Thermal conductivity (k-value; W/mK) was determined with a LaserComp Fox 50 thermal conductivity measurement system. For thermal conductivity (k-value) tests, samples were cut out from extruded profiles in the form of a disk with 50 mm diameter and 6 mm thickness. In order to assess the effect of HGMs on the cooling rate from the melt, prefabricated profiles were heated to a partially molten state in a compression molder followed by the rapid cooling to ambient room temperature.

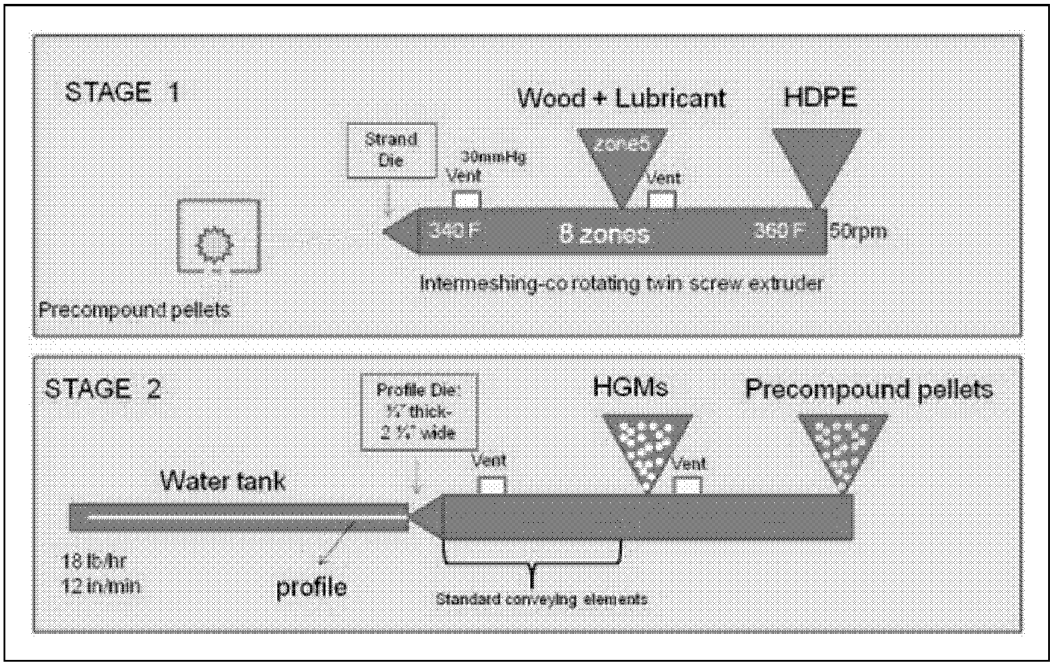

Figure 3. Processing scheme of hollow glass microsphere (HGM)-filled wood/high density polyethylene (HDPE) composite sheeting. The compounding and extrusion of HGMs required no significant changes in typical polymer processing equipment.

Simultaneous temperature measurements were performed by using a noncontact infra-red (IR) camera. In order to elucidate the potential effects of the presence of HGMs on crystallization of HDPE, differential scanning calorimetry tests were performed on the extruded sheets with a DSC device (TA instruments Q-2000) in nitrogen atmosphere. During the first thermal scan, the sample was heated at a scanning rate of 20°C/min to 210°C and kept at this temperature isothermally for 5 min, in order to erase the thermal history. The results (data not shown) revealed no significant impact of HGMs on crystallization of HDPE.

Mechanical properties. Flexural properties were determined with a Sintech 1/G system by following the guidelines presented in ASTM D790 three-point bending test. Izod impact strength was determined using a Tinius Olson Izod impact measurement system according to the ASTM D 256 standard. It is important to note that the determined experimental properties of the prepared composites—flexural modulus, flexural strength, and Izod impact strength—using these instruments are not absolute values and should be considered relatively, in conjunction with the properties of the unfilled specimens. For each test, 10 specimens were tested and their arithmetic average was reported. The standard deviation in the measurements was observed to be around 5%.

Table 1. Composition of the prepared samples. HDPE stands for high density polyethylene and HGM stands for hollow glass microspheres. Note that the compositions of the precompound and the control sample (HGM-0) are the same, although their processing histories were different. Precompound was passed through the twin-screw extruder only once, although the control sample (HGM-0) was passed twice in order to achieve the same processing history with the HGM-containing samples. The samples which contained iM30K grade HGMs were labeled with the corresponding commercial trade names. The number following HGM stands for the HGM content (wt%) introduced to the composition

	Content (wt%)				
Sample	Wood flour	Lubricant	HDPE	HGM	HGM grade
Precompound	50.0	6.0	44.0	0.0	NA
HGM-0	50.0	6.0	44.0	0.0	NA
HGM-iM30K-5	47.5	5.7	41.8	5.0	iM30K (density = 0.60 g/cc)
HGM-iM30K-10	45.0	5.4	39.6	10.0	iM30K (density = 0.60 g/cc)
HGM-iM30K-15	42.5	5.1	37.4	15.0	iM30K (density = 0.60 g/cc)
HGM-iM30K-18.5	40.7	4.9	35.9	18.5	iM30K (density = 0.60 g/cc)
HGM-K42HS-10	45.0	5.4	39.6	10.0	K42HS (density = 0.42 g/cc)

Table 2. The density of the samples in wet and dry states and the extent of HGM breakage in the samples. The density in dry state was determined immediately after profile extrusion. The density in wet state was determined after immersing the samples in distilled water for 48 h at room temperature

Sample	Theoretical HGM content (wt%)	Actual inorganic residue content (HGM + ash; wt%)	Density of the inorganic residue (g/cc)	Volume loss due to HGM breakage (%)	Dry density (g/cc)	Wet density (g/cc)	Note
HGM-0	0.00	0.63	NA	NA	1.112	1.140	The inorganic content originates from the ash.
HGM-iM30K-5	5.00	5.51	0.7249	17.23	1.065	1.110	The inorganic content originates from the ash and HGM
HGM-iM30K-10	10.00	9.94	0.6846	12.36	1.030	1.070	
HGM-iM30K-15	15.00	15.26	0.6749	11.10	0.975	1.030	
HGM-iM30K-18.5	18.00	18.46	0.6806	11.84	0.930	0.970	
HGM-K42HS-10	10.00	10.38	0.6057	30.66	0.910	0.950	

HGM: hollow glass microspheres.

Results and discussion

The extent of glass bubble breakage, the density reduction in the presence of HGMs, and the processing characteristics

A small amount of HGMs were observed to be broken during compounding operation (Table 2). The extent of HGM survival iM30K bubbles was observed to be around 90%, whereby the extent of survival was determined to be 70% for relatively weaker K42HS grade bubbles. The morphology of the iM30K HGMs-filled wood/HDPE composites is presented in Figures 4 and 5. The SEM images also revealed that the majority of the iM30K HGMs were intact and a satisfactory state of dispersion for HGMs was observed even at high loadings. As anticipated, the presence of intact HGMs significantly reduced the density of the wood/polymer composites (Table 2). It was further determined that the presence of HGMs did not alter the melt viscosity as shown in Figure 6.

Engineering properties

Mechanical properties and water vapor absorption properties. The presence of HGMs increased the flexural moduli of the composites at the relatively slight expense of impact resistance (Table 3). Note that the presence of 18.5 vol% iM30K HGM increased the specific flexural modulus by about 35%.

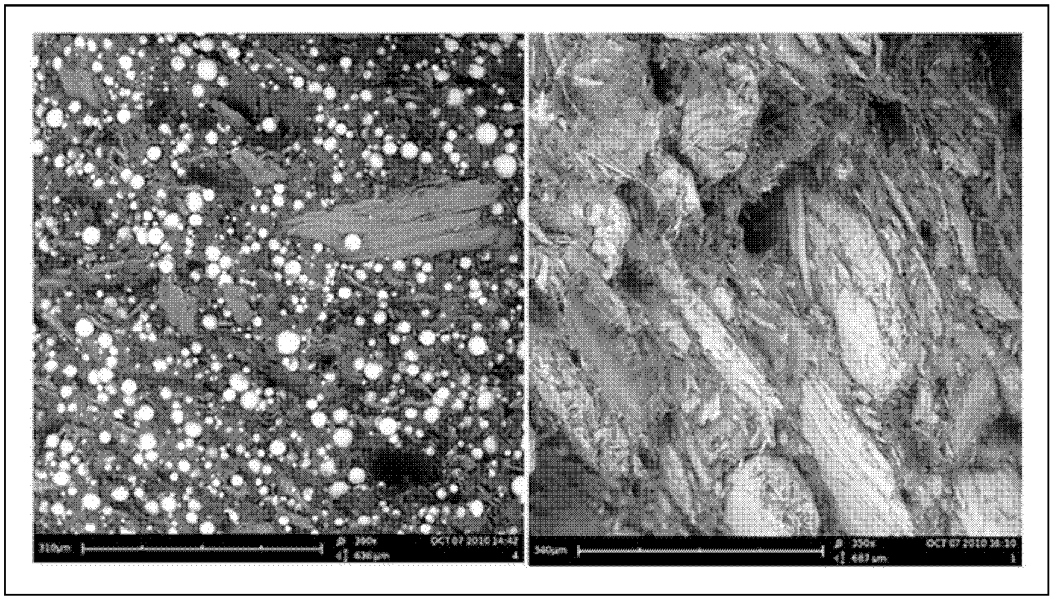

Figure 4. Morphology of 15 wt% hollow glass microsphere (HGM)-filled wood/high density polyethylene (HDPE) composite (right) and the control without any HGM (left). The HGMs were observed to be highly dispersed in the matrix.

The increase of flexural modulus and strength, while decreasing the density, is an advantage when compared to air-foamed counterparts. The presence of HGMs increased the water vapor absorption slightly as presented in Figure 7. One may anticipate that the formation of vacuoles on the interfaces between the HGMs and the matrix polymer could be responsible for increased moisture absorption in the present system.[10] In spite of this, the water absorption stabilized at small percentages. This is contrary to that seen in gas-foamed composites where the water absorption continuously increases once the water ingress into the open pores happen.

Thermal properties. The presence of HGMs significantly reduced the thermal conductivity of the composites (Figure 8). In addition, the presence of

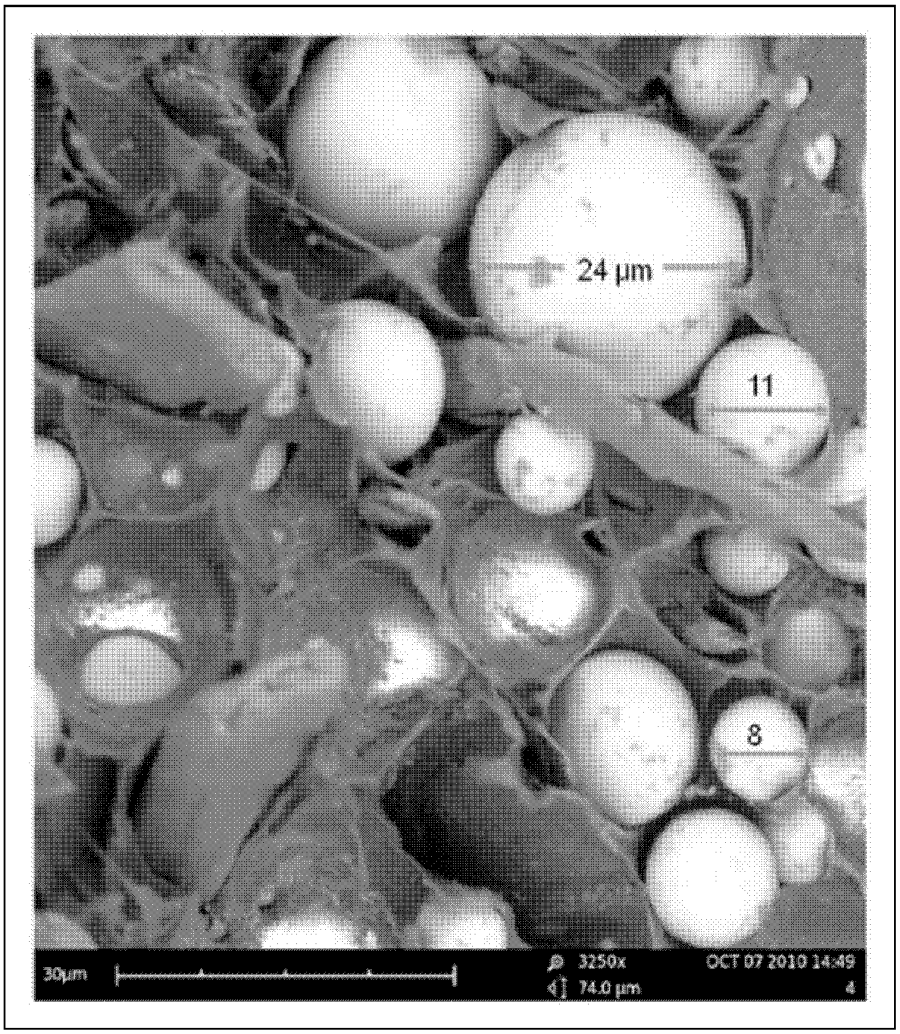

Figure 5. High resolution scanning electron microscope (SEM) micrograph of 15 wt% hollow glass microsphere (HGM)-filled wood/high density polyethylene (HDPE) composite. Note the size distribution of HGM particles.

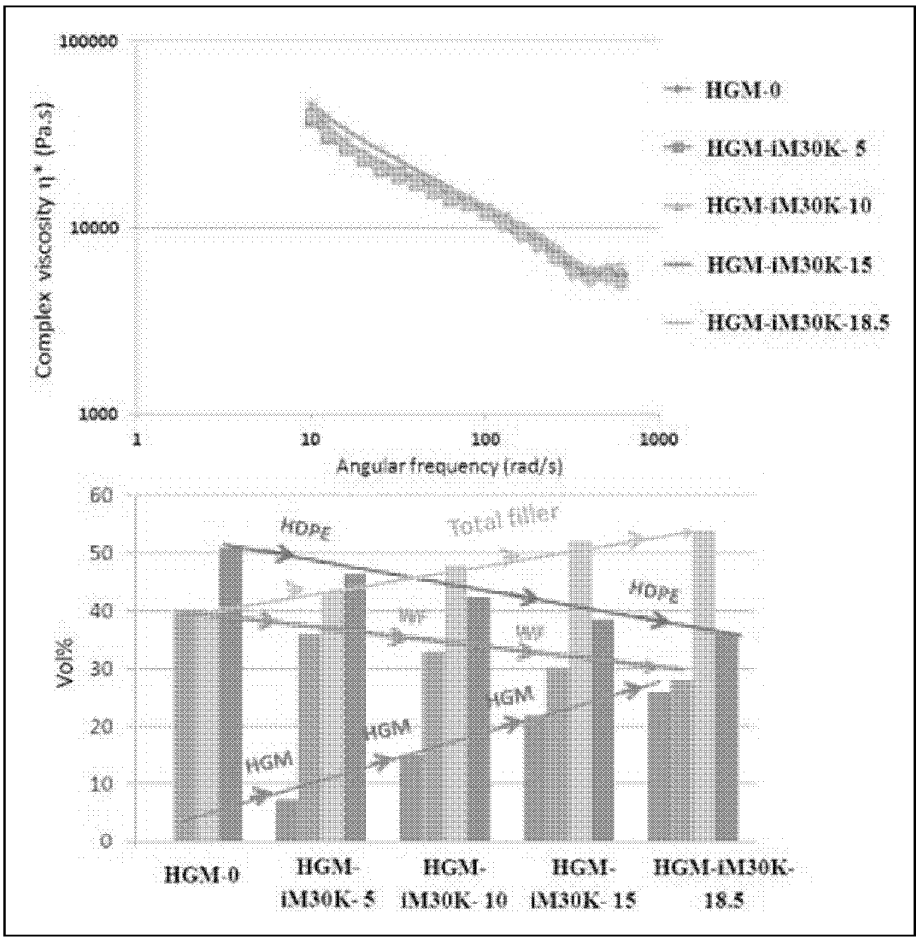

Figure 6. Melt shear viscosity of different formulations. The term "WF" stands for the wood flour content. The presence of hollow glass microspheres (HGMs) did not significantly alter the melt viscosity even at relatively high loadings.

Table 3. Flexural properties and impact strength of the samples. The specific flexural modulus was determined as the ratio of flexural modulus to the density of the sample. Note the significant increase in specific flexural modulus, while reducing density in the presence of iM30K HGMs

Sample	Flexural strength (MPa)	Flexural modulus (GPa)	Izod impact strength (J/m^2)	Specific flexural modulus (GPa/[g/cc])	Dry density (g/cc)
HGM-0	27.6	3.1	3080	2.8	1.112
HGM-iM30K-5	31.0	3.3	2680	3.1	1.065
HGM-iM30K-10	28.9	3.5	2380	3.4	1.030
HGM-iM30K-15	26.2	3.6	2180	3.7	0.975
HGM-iM30K-18.5	22.8	3.5	2080	3.8	0.930

HGM: hollow glass microspheres.

HGMs remarkably increased the rate of cooling of the extruded profiles (Figure 9) by reducing the total thermal mass required to be cooled down. One may anticipate that the improved rate of cooling would be very useful in profile extrusion. Recall that the relatively low rate of cooling of the extruded

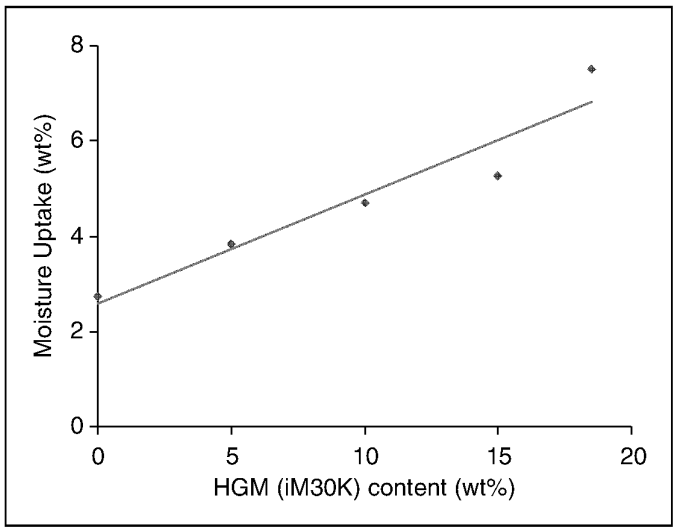

Figure 7. The moisture uptake of the samples as a function of hollow glass microsphere (HGM) content. The HGM grade in the samples was iM30K. The moisture uptake was determined based on dry weight of the samples. The straight line is presented to guide the eye.

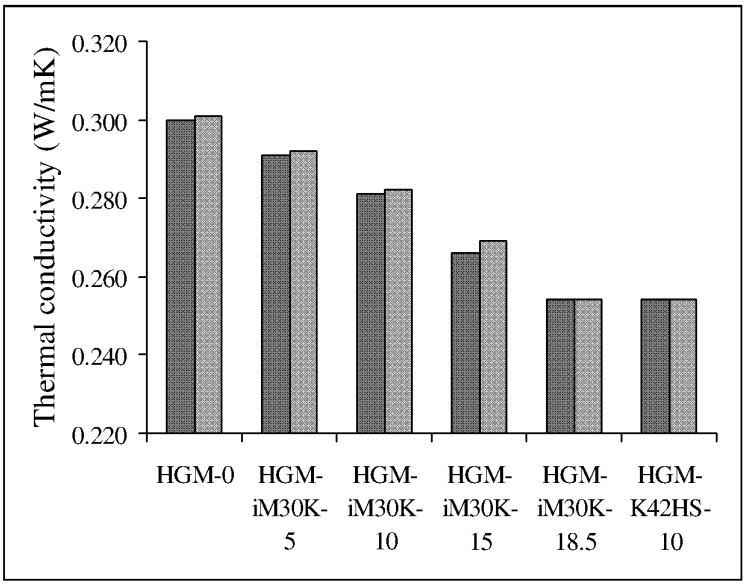

Figure 8. Thermal conductivity of the samples. The blue data points were determined at 40°C and the red data points were determined at 60°C.

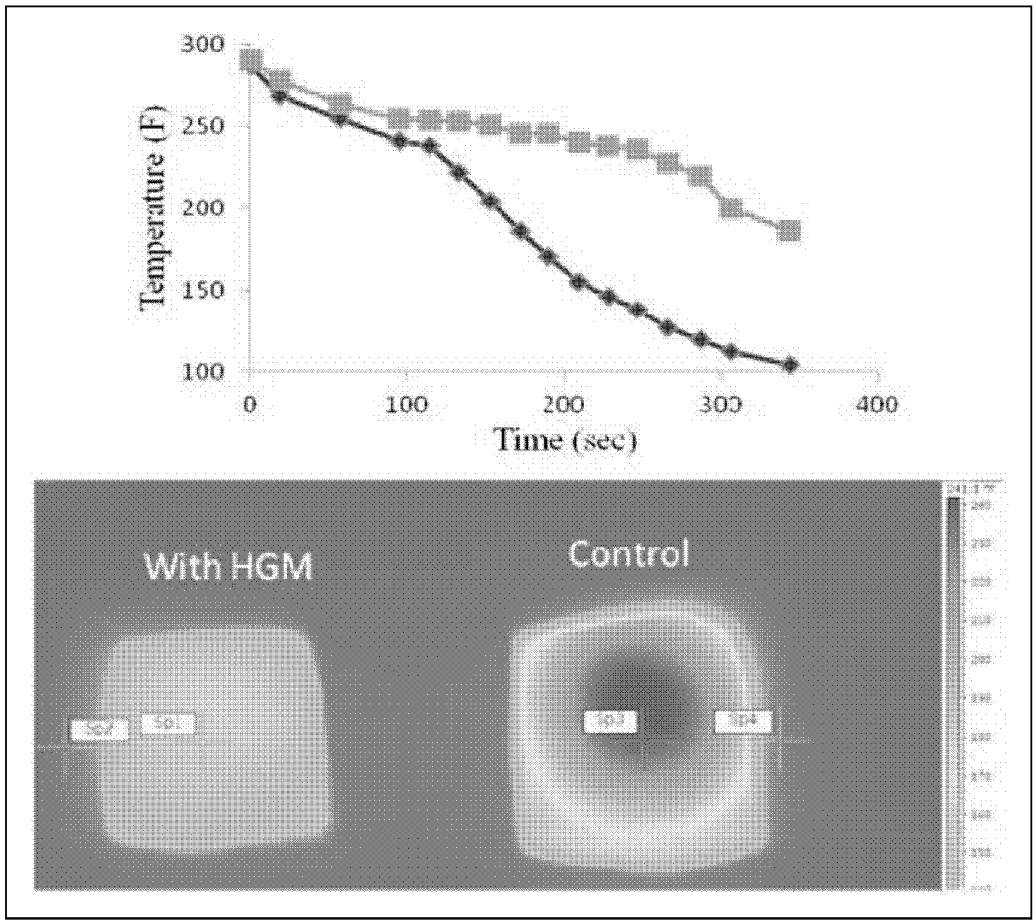

Figure 9. Top: The measured temperatures of the unfilled (red) and 10 wt% hollow glass microsphere (HGM)-filled (blue) extruded profile sheets upon cooling from melt. Bottom: Representative infra-red (IR) camera images. The temperature readings were color coded—dark red indicated the highest observed temperature (~120°C), whereby light green/blue indicated the lowest temperature (~60°C). The presence of HGMs facilitated rapid cooling from melt by reducing the thermal mass required to be cooled down.

profiles has been one of the major bottlenecks in profile extrusion[11] due to the significant thickness of the profiles. Our observations suggested that introducing HGMs might significantly improve the rate of profile extrusion, without any need for additional cooling operation or without the need of reducing the sheet thickness. Our results also suggest that the presence of HGMs may facilitate melt processing of significantly thick sheets as well.

Warpage, appearance, and application-related properties. The presence of HGMs was observed to reduce the warpage and sink marks in the extruded sheet (Figure 10). The presence of HGMs also rendered sharper contours and

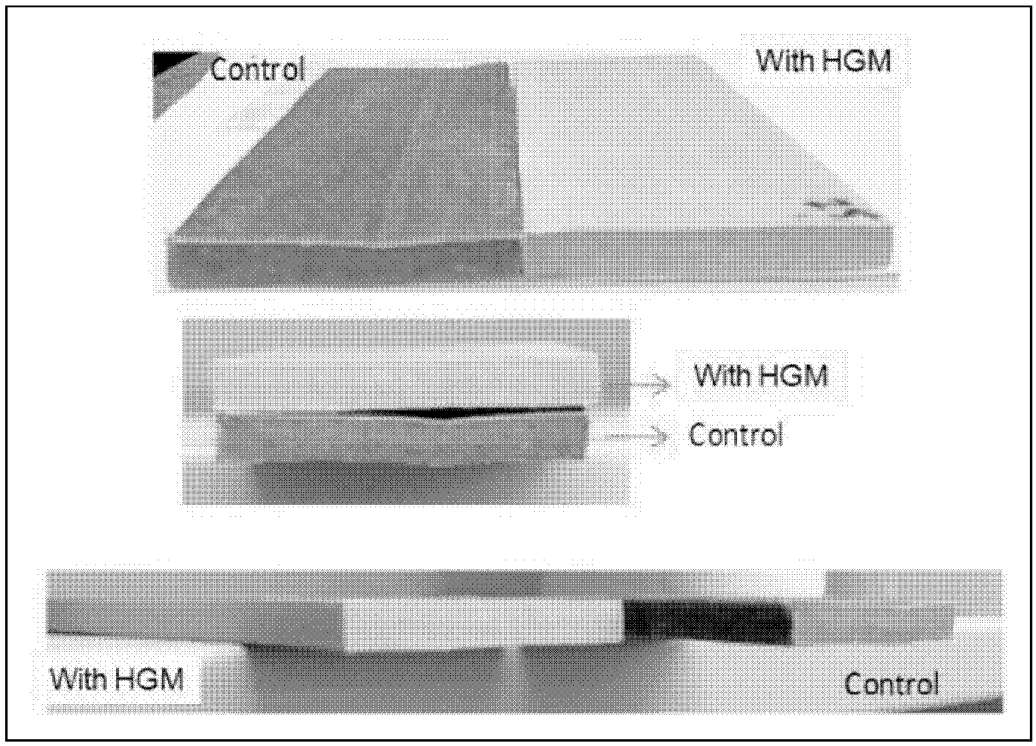

Figure 10. Visual appearance of the extruded sheets. The hollow glass microsphere (HGM) content was 10 wt%. Note the significantly improved and uniform visual appearance of the HGM-filled sheets.

corners in the extruded profiles, as well as better surface definition than profiles without any HGMs.

In addition, it was observed that the presence of HGMs induced a distinct visual appearance with a lighter brown–yellowish color (Figure 10). One may infer that the increased extent of light scattering due to the HGMs was responsible for the distinct visual appearance.[12] It was also observed that the presence of HGMs rendered drilling the extruded profiles much easier and faster. Recall that one of the challenges of common wood/polymer composite sheeting is that it is harder to nail and screw into the composite material as compared to regular wood. This has been mainly originating from the presence of the stiffer polymer phase in the composite. Upon the incorporation of HGMs into the wood/HDPE composite, it accepted screws and nails more like real wood than do their counterparts without HGMs. In order to demonstrate this effect, a simple experiment was performed. Two profiles, one filled with 15 wt% HGMs (HGM-iM30K-15) and the other without any HGMs, were drilled using a heavy-duty drill with a ¼ in drill bit. The electric drill was held on the wood composite sheets without applying any extra pushing force other than the weight of the drill. One could approximate the pressure at the wood-drill bit contact point by dividing the weight of the drill by

the cross-sectional area of the ¼ in drill bit. Hence, one might anticipate that 1800 *g* drill weight applies a pressure of ~80 ibf/in^2 at the contact point. With this pressure, it takes about 75 s for the drill to go through the ¼ in thick sheet without any HGMs, while it took only 15–20 s for the sheet with the HGMs. The HGMs, when broken due to contact with the drill bit, provided void volume for the drill bit to penetrate through. The same observation was validated for the nails as well. It was observed that the nails get inserted with much less effort into the composites filled with HGMs.

Conclusions

The following conclusions could be drawn from the presented study:

1. The presence of HGMs significantly reduced the density of the composites, while increasing flexural modulus and strength.
2. The presence of HGMs improved thermal and mechanical properties without negatively impacting the processing and flow characteristics.
3. The presence of HGMs improved the dimensional characteristics of the composites and imparted better nailability and screwability.

Acknowledgments

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Biographies

Baris Yalcin is an application development specialist in 3M Energy and Advanced Materials Division. He has a Ph.D. in Polymer Engineering from The University of Akron, Akron, OH, USA. His expertise is on the manufacture and investigation of advanced functional polymer based materials with innovative process design concepts. He has over 25 technical publications/patents. He serves as member of the editorial board for *Journal of Plastic Film & Sheeting,* as technical program chair for Society of Plastics Engineers Polymer Modifiers and Analysis Division (SPE PMAD) (2011–2012), and is an active reviewer for several refereed plastics journals.

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