

# Effects of the Chemical Foaming Agents, Injection Parameters, and Melt-Flow Index on the Microstructure and Mechanical Properties of Microcellular Injection-Molded Wood-Fiber/Polypropylene Composites

Andrzej K. Bledzki, Omar Faruk

Institut für Werkstofftechnik, Kunststoff- und Recyclingtechnik, University of Kassel, Mönchebergstraße 3, D-34109 Kassel, Germany

Received 27 May 2004; accepted 14 September 2004

DOI 10.1002/app.21685

Published online in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** Wood-fiber-reinforced plastic profiles are growing rapidly in nonstructural wood-replacement applications. Most manufacturers are evaluating new alternative foamed composites, which are lighter and more like wood. Foamed wood composites accept screws and nails better than their nonfoamed counterparts, and they have other advantages as well. For example, internal pressures created by foaming give better surface definition and sharper contours and corners than nonfoamed profiles have. In this study, the microfoaming of polypropylene (PP) containing hardwood fiber was performed with an injection-molding process. The effects of different chemical foaming agents (endothermic, exothermic, and endothermic/exothermic), injection parameters (the mold temperature, front flow speed, and filling quantity), and different types of PP (different melt-flow indices) on the density, microvoid content, physicomechanical properties, surface roughness, and microcell classification of microfoamed PP/wood-fiber composites were studied. A maleic anhydride/polypropylene copolymer (MAH-PP) compatibilizer was used with the intention of improving the mechanical properties of microfoamed composites. The microcell classification (from light

microscopy) and scanning electron micrographs showed that an exothermic chemical foaming agent produced the best performance with respect to the cell size, diameter, and distance. The polymer melt-flow index and the variation of the injection parameters affected the properties and microstructure of the microfoamed composites. The density of the microfoamed hardwood-fiber/PP (with a high melt-flow index) composites was reduced by approximately 30% and decreased to 0.718 g/cm<sup>3</sup> with an exothermic chemical foaming agent. Tensile and flexural tests were performed on the foamed composites to determine the dependence of the mechanical properties on the density and microvoid content of the foamed specimens, and these properties were compared with those of nonfoamed composites. MAH-PP improved the physicomechanical properties up to 80%. With an increase in the mold temperature (80–110°C), the surface roughness was reduced by nearly 70% for the foamed composites. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 97: 1090–1096, 2005

**Key words:** composites; fibers; foams; injection molding; mechanical properties; poly(propylene) (PP)

## INTRODUCTION

Many new technologies are now being introduced and reintroduced for foam injection-molding processing. Often called *microcellular foaming*, these new technologies use a number of approaches to achieve fine cellular structures with double-digit weights and cycle time reductions. The keys to these innovative technologies are computerized process control and good tool design, including counter pressure, static melt mixing, and new chemical foaming agents (CFAs).<sup>1,2</sup>

Moreover, one of the fastest growing materials for plastics is also one of the oldest: wood fiber. The processing of microcellular foamed structures in poly-

mer/wood-fiber composites through a batch foaming process<sup>3–7</sup> and a continuous extrusion process<sup>8–12</sup> has been investigated. Park et al.<sup>13</sup> experimented with two system configurations (a tandem extrusion system vs a single extruder system) for wood-fiber/polymer composites to demonstrate the system effect on the cell morphology and foam properties. Microcellular foaming in an injection-molding process was also introduced with natural fiber (flax fiber) and wood fiber.<sup>14–17</sup>

A recent report<sup>18</sup> reviewed the different processing systems used for microcellular wood-fiber-reinforced polymer composites. It described the recent developments of microcellular natural-fiber-reinforced and wood-fiber-reinforced polymer composites.

There are several variables to consider when an injection-molding machine is operated. Some of these variables can affect the physical properties of foam composites. It is well established that the mold tem-

Correspondence to: A. K. Bledzki (kutech@uni-kassel.de).

perature and cooling time are important variables in this respect. Many other factors can be adjusted, including variables such as the front flow speed and filling quantity, that can affect one or more foam properties.

CFAs enable both commodity and engineering polymers to be processed more easily and make possible improved properties for injection-molding and extrusion processes. Compatibility and chemical reactions with foaming agent byproducts are becoming more important because of improvements in the weights of molded parts. Also, dramatic strides are being made with new CFAs in achieving reproducible cell distributions and sizes in microcellular foaming.

CFAs are substances that decompose at processing temperatures, and so gases such as CO<sub>2</sub> and nitrogen are liberated. Solid organic and inorganic substances are used as CFAs. The solid residues react as nucleation centers. This leads to a finer cell structure and better solubility of the gas in the polymer melt. There are several different types of CFAs, which differ mostly in the type of gas that is generated and the type of reaction that generates the gas. The reaction that produces the gas can either absorb energy (endothermic) or release energy (exothermic). Generally, CFAs are divided by their enthalpy of reaction into two groups: exothermic and endothermic foaming agents. Nowadays, a combination of exothermic and endothermic CFAs is also used for foaming.

We investigated the effects of different CFAs, injection parameters, and polymer melt-flow indices on the physicochemical properties of resultant hardwood-fiber/polypropylene (PP) microfoamed composites and the influence of the compatibilizer on the properties.

## EXPERIMENTAL

### Materials

PP [Stamylan P17M10 (melt-flow index = 10.5 g/10 min) and Stamylan P213MNK 40 (melt-flow index = 90 g/10 min)] was provided as granules by DSM (Gelsenkirchen, Germany).

Hardwood fiber (Lignocel HBS 150-500) with a particle size of 150–500 μm was supplied by J. Rettenmaier & Söhne GmbH (Holzmühle, Germany).

A commercially available maleic anhydride/polypropylene copolymer (MAH-PP; Licomont AR 504 FG; acid number = 37–43 mg of KOH/g) was used as a compatibilizer. It was obtained from Clariant Corp. (Frankfurt, Germany). It accounted for 5% of the weight percentage of the wood fiber.

To obtain foamed wood-fiber-reinforced composites, we used three types of CFAs (endothermic, exothermic, and endothermic/exothermic), which were obtained from Clariant Masterbatch GmbH (Ahrens-

burg, Germany). In this study, CFAs were used at a 4 wt % concentration in the composites.

### Processing and foaming

Hardwood fibers with PP were mixed in a Henschel HM40 KM120 high-speed mixer with and without a coupling agent. Hardwood fibers were dried at 80°C in an air-circulating oven for 24 h (moisture content < 1%) before mixing. The high-speed mixer was preheated to 180°C, the speed of the rotors was set to 2200 rpm, and the mixing time was 12–15 min. Cold agglomerated granules were then mixed with different CFAs. Before the foaming in the injection molding, the mixed granules were dried at 80°C for 24 h. The specimens (200 mm × 90 mm × 4 mm) of the hardwood-fiber foamed composites were prepared by an injection-molding process at a melting temperature of 150–180°C, a mold temperature of 80–110°C, and an injection pressure of 20 kN/mm<sup>2</sup>.

### Measurements

The tensile and flexural tests were conducted with a Zwick UPM 1446 at a test speed of 2 mm/min according to EN ISO 527 and EN ISO 178, respectively, for the hardwood-fiber/PP composites. All tests were performed at room temperature (23°C) at a relative humidity of 50%, and five to eight samples were tested for each treatment. The densities of the nonfoamed and microfoamed specimens were measured according to DIN 53479. Fifteen replicates were conducted for each treatment. The microvoid content was calculated according to ASTM Standard D 2734-70 for foamed composites. The surface roughness of the wood-fiber/PP composites was measured according to ISO 4287/1 with a laser-sensor autofocus measurement system.

### Scanning electron microscopy (SEM) and light microscopy

The morphology of the wood-fiber-reinforced microcellular PP composites and the cell size, shape, and distribution of the microcells in the microfoamed composites were investigated with SEM (Vega Tescan) and light microscopy. Cross sections of the sanded and polished surfaces were studied with light microscopy. Fractured surfaces of the flexural test samples were observed with SEM after they were coated with gold.

### Microcell classifications

This investigation was accomplished with a digital image analysis system with Digitrace image processing software.

The image processing and evaluation took place as follows:

- Picture introduction.
- Picture improvement.
- Object recognition.
- Binary image processing.
- Object measuring technique.

For the evaluation of the microcell classifications, the following measured variables from the geometrical relations of the binary picture were presented as results.

#### Microcell diameter

The cavity diameter was indicated with respect to the current scaling in micrometers. The diameter of the pores was calculated over the surface of a circle with same area.

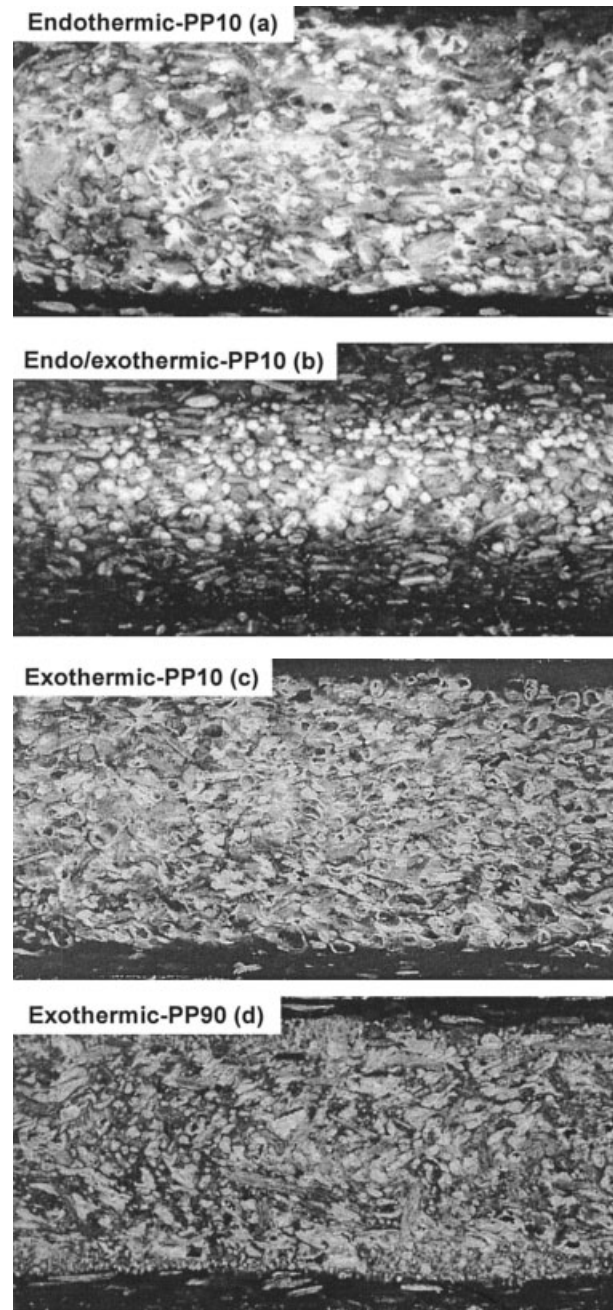
#### Cell distance

This value described the distribution of the microcells. The distances of the neighboring cells were measured. With the help of an interactive interference, it was possible to correct wrongly reconstructed line distance measurements.

### RESULTS AND DISCUSSION

The microstructure of the foamed composites was influenced by the CFAs in the injection-molding process. The foaming area and distribution of the cells are shown in light micrographs of the hardwood-fiber/PP microfoamed composites in Figure 1(a–d). The micrographs illustrate that the foamed structure, near the injecting point, had a three-layer sandwich structure. It contained a middle layer with distributed cells, and a compact outer hull was identified. Between the foaming area and surface layers, there was a transition zone, in which the microcells rode from the injecting point to the boundary area. Moreover, the foaming area (middle layer) of the composites with an endothermic foaming agent was thinner than that with other CFAs. The effect of the melt-flow index on the microfoamed structure can be observed in Figure 1(c,d). For composites with high-melt-flow-index PP (PP90), the microcells were bigger in the middle area, whereas in the boundary area, many smaller microcells existed. This occurred because PP with a high melt-flow index improved the expansion of the blisters in comparison with PP with a lower melt-flow index (PP10).

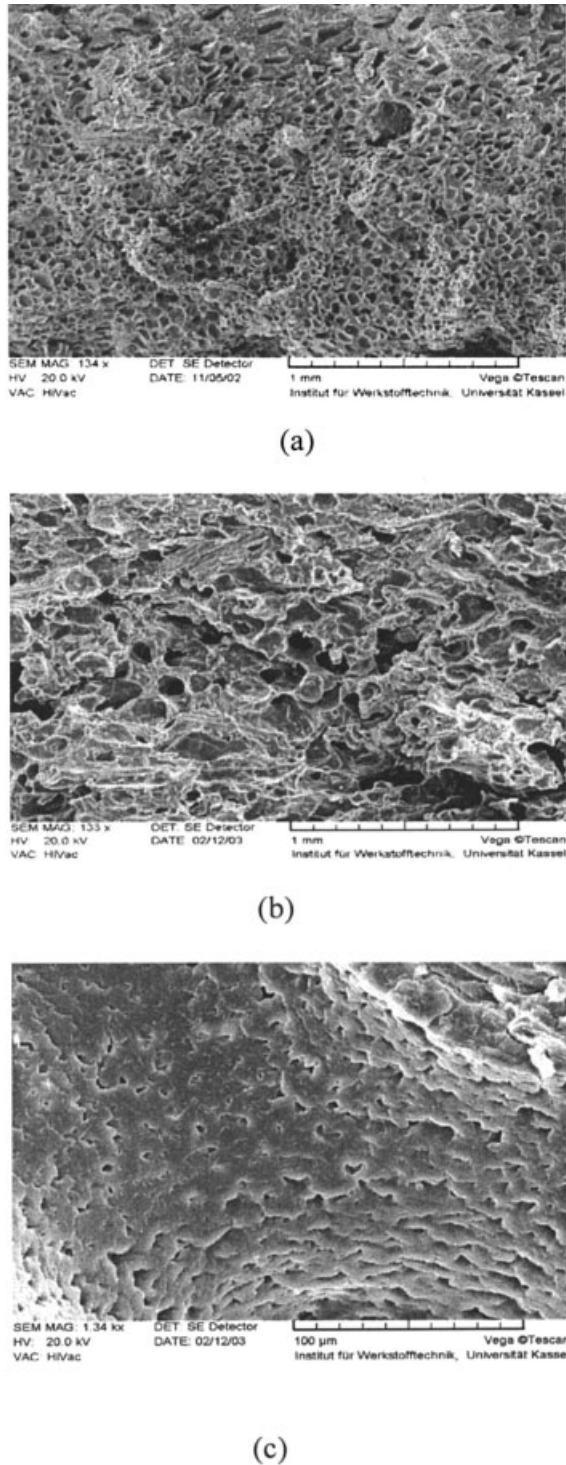
Our previous experiment<sup>19</sup> demonstrated that CFAs play an important role in the cell dimensions and distributions. The biggest cell sizes were observed in



**Figure 1** Light micrographs of hardwood-fiber/PP microfoamed composites: (a) endothermic PP10, (b) endothermic/exothermic PP10, (c) exothermic PP10 (melt-flow index = 10 g/10 min), and (d) exothermic PP90 (melt-flow index = 90 g/10 min). The wood-fiber concentration was 30%, and the magnification was 12.5:1.

composites with endothermic foaming agents. Composites with exothermic foaming agents, however, presented the finest cells. Figure 2 shows SEM micrographs of hardwood-fiber/PP (both types) microfoamed composites with an exothermic foaming agent. Bigger cells were observed for the high-melt-flow-index PP composites rather than the regular PP composites. It is also remarkable that, for the high-



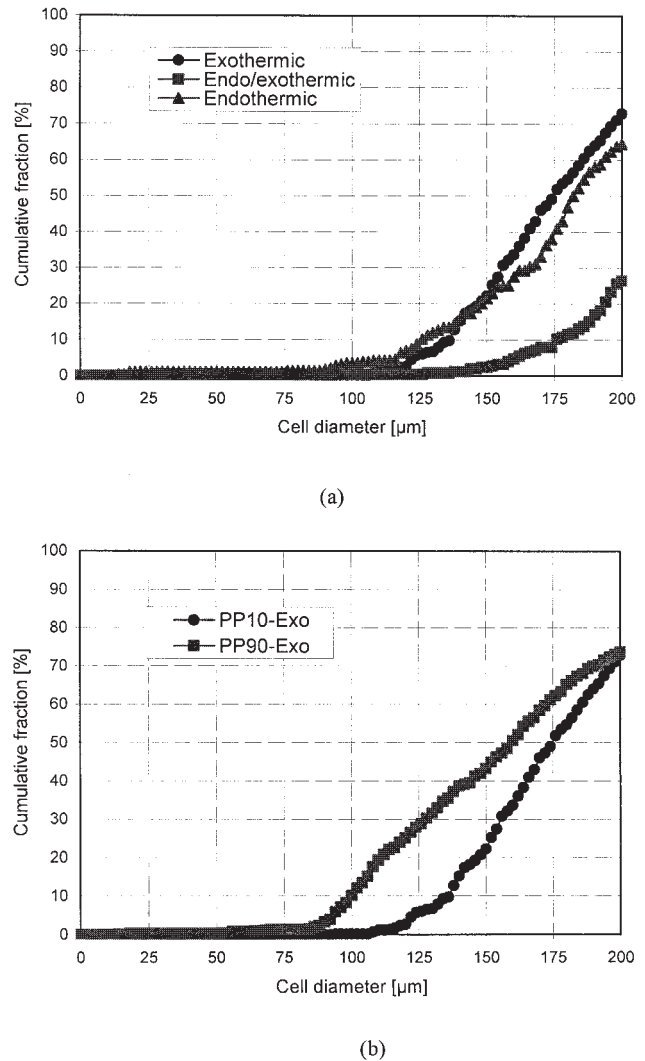


**Figure 2** SEM micrographs of hardwood-fiber/PP microfoamed composites: (a) exothermic PP10, (b) exothermic PP90, and (c) exothermic PP90. The wood-fiber concentration was 30%.

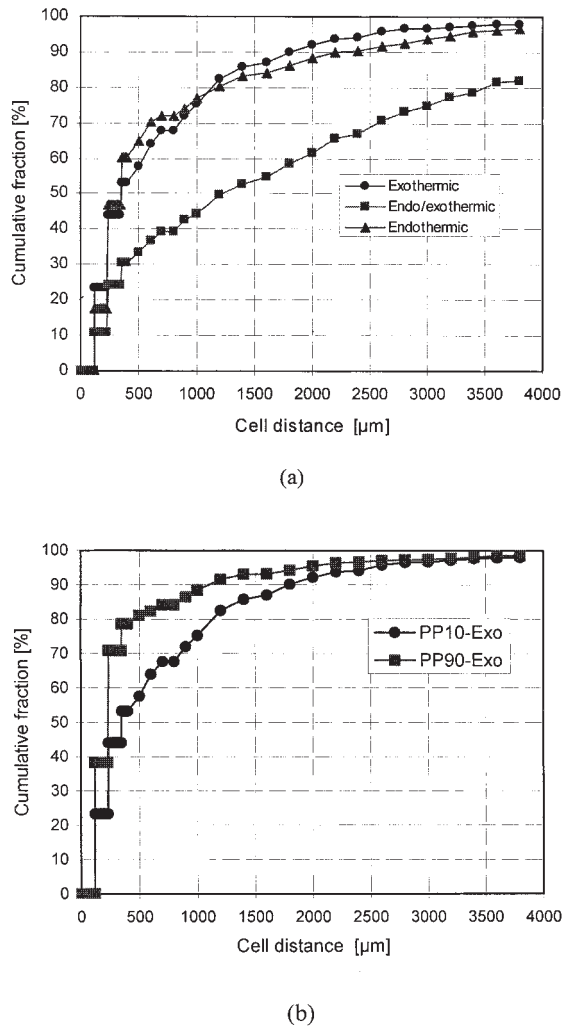
melt-flow-index PP composites, the cells contained very small microcells in their walls, as shown in Figure 2(c) in a higher magnification micrograph. This was due to the lower viscosity of the high-melt-flow-index PP, which favored the expansion of gases more

than PP with a lower melt-flow index because the viscosity was lower in the boundary area than in the middle area.

From the light micrographs, the cumulative fractions of the cell diameters and the distances of the hardwood-fiber/PP microfoamed composites were measured with Digitrace computer software. Figure 3 shows the cumulative fractions of the cell diameters with all CFAs for the hardwood-fiber/PP microfoamed composites with 30% wood fiber [Fig. 3(a)] and also the effect of the melt-flow index [Fig. 3(b)]. The cumulative fractions of the cell distances of the microfoamed composites are presented in Figure 4. The exothermic foaming agent showed better cell distribution and form than the other CFAs. PP with a high melt-flow index showed better performance with respect to the cell distribution and form.



**Figure 3** Cumulative fraction of the cell diameter of hardwood-fiber/PP microfoamed composites: (a) PP10 with different CFAs and (b) regular PP (PP10) and high-melt-flow-index PP (PP90). The wood-fiber concentration was 30%.



**Figure 4** Cumulative fraction of the cell distance of hardwood-fiber/PP microfoamed composites: (a) PP10 with different CFAs and (b) regular PP (PP10) and high-melt-flow-index PP (PP90). The wood-fiber concentration was 30%.

The densities of the microfoamed composites (with all the CFAs) and the nonfoamed composites are summarized in Table I. The exothermic foaming agent reduced the maximum density for the hardwood-fiber composites. With the addition of MAH-PP, the density showed a more reducing tendency, which was around 30%, and decreased to  $0.741 \text{ g/cm}^3$ . For high-melt-

flow-index PP, the composites' density decreased at most to  $0.718 \text{ g/cm}^3$ .

In our previous work, we showed that the characteristic mechanical values of hardwood-fiber/PP microfoamed composites did not differ significantly with all types of CFAs. Table II presents the specific tensile and flexural strengths of both types of PP/hardwood-fiber composites. The specific tensile strength decreased proportionately in the foamed composites. The specific tensile strength and flexural strength for both types of PP/wood-fiber composites were calculated from the ratio of the tensile or flexural strength to the density. With the addition of the MAH-PP (5%) compatibilizer to the foamed composites, the specific tensile strength and flexural strength increased for the regular PP/wood-fiber composites (at most 80% for the specific tensile strength). However, for the high-melt-flow-index PP/wood-fiber composites, the specific tensile and flexural strengths did not increase more than those of the nonfoamed composites. This was due to not only the higher microvoid concentration but perhaps also the larger size of the microcells.

Figure 5 presents the influence of the variation of the filling quantity on the specific flexural strength. With the filling quantity increasing, the specific flexural strength decreased. This indicated that a suitable injected mass should be selected for microfoaming the materials because a large injected mass increased the density and at the same time reduced the specific flexural strength. This confirmed that with this production process, raw material savings and improvements in the specific mechanical properties could be achieved at the same time.

Figure 6 shows the influence of the front flow speed on the density and microvoid concentration of the composites. As the front flow speed increased in the tool, the density decreased and the microvoid concentration decreased. This proves that the viscosity of the injected mass decreased under the influence of the resulting shear stress from the front velocity of the flow.

For microfoamed products, many researchers and manufacturers of foaming agents have reported that microfoaming produces a smooth surface in comparison with the surfaces of nonfoamed products.<sup>20</sup> Our

**TABLE I**  
Density ( $\text{g/cm}^3$ ) of Nonmicrofoamed and Microfoamed Hardwood-fiber/PP composites (30% Wood Fiber)

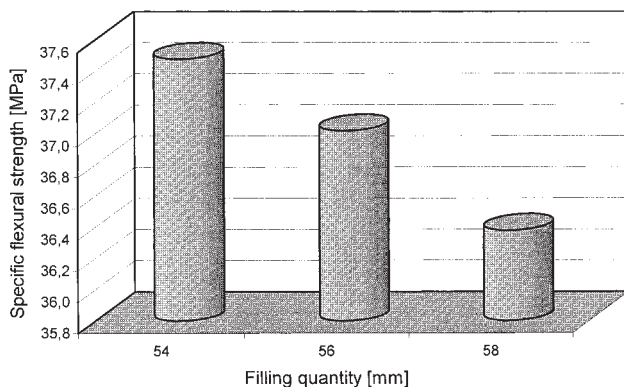
	Endothermic regular PP	Endothermic/exothermic regular PP	Exothermic	
			Regular PP	PP with a high melt-flow index
Nonfoamed	1.01	1.01	1.01	0.94
Foamed	0.85	0.78	0.76	0.73
Foamed with MAH-PP	0.84	0.77	0.74	0.71

**TABLE II**  
**Specific Tensile and Flexural Strength of Nonmicrofoamed and Microfoamed Hardwood-fiber composites (30% Wood Fiber and Exothermic Foaming Agent)**

	Specific tensile strength [MPa/(g/cm <sup>3</sup> )]		Specific flexural strength [MPa/(g/cm <sup>3</sup> )]	
	Regular PP	PP with a high melt-flow index	regular PP	PP with a high melt-flow index
Nonfoamed	25.58	24.85	46.9	40.4
Foamed	16.71	15.43	43.5	36.09
Foamed with MAH-PP	29.81	18.37	47.5	39.52

previous results<sup>19</sup> showed that the surface roughness of composites could be significantly improved by microfoaming. The surface roughness of microfoamed composites with an endothermic foaming agent was reduced by at most 70% with respect to that of nonfoamed composites.

The mold temperature had a great influence on the surface roughness of the microfoamed composites. Figure 7 shows laser surface photographs of hardwood-fiber/PP microfoamed composites containing 30% wood fiber. The upper and lower peaks (the number of peaks and the area of the peaks) indicate the surface roughness of the composites. With an increase in the mold temperature from 80 to 110°C, the surface roughness decreased for the microfoamed composites. The roughness maximum and arithmetical roughness mean deviation of the foamed composites are illustrated in Figure 8. The surface roughness of the microfoamed composites was significantly reduced (at most 70%) as the mold temperature increased. With an increasing mold temperature, the temperature difference was reduced between the foamed core and surface of the composites. Thus, the gas expanded with the mass against the smooth mold wall. Moreover, PP exhibited a better surface as the mold temperature increased.



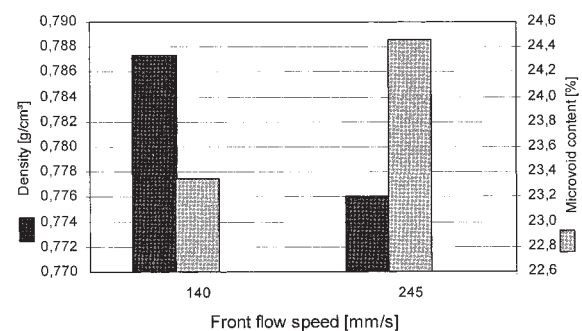
**Figure 5** Influence of the filling quantity on the specific flexural strength. The hardwood-fiber concentration was 30%, and the exothermic foaming agent was regular PP.

## CONCLUSIONS

Microcellular hardwood-fiber-reinforced PP composites were prepared with an injection-molding process. This study examined the effects of the different types of CFAs, the polymer melt-flow index, the variation of the injection parameters, and the compatibilizer on the microstructure and mechanical properties of the resultant foamed composites. The following conclusions were made:

1. The microfoam structure of the composites with an exothermic foaming agent showed the best performance with respect to the cell size, diameter, and distance for the microfoamed composites.
2. With the addition of a compatibilizer, the physico-mechanical properties of the wood-fiber/PP microfoamed composites improved up to 80% (30% hardwood fiber and an exothermic foaming agent).
3. The melt-flow index and the variation of the injection parameters had a great influence on the properties and structure of the microfoamed composites:

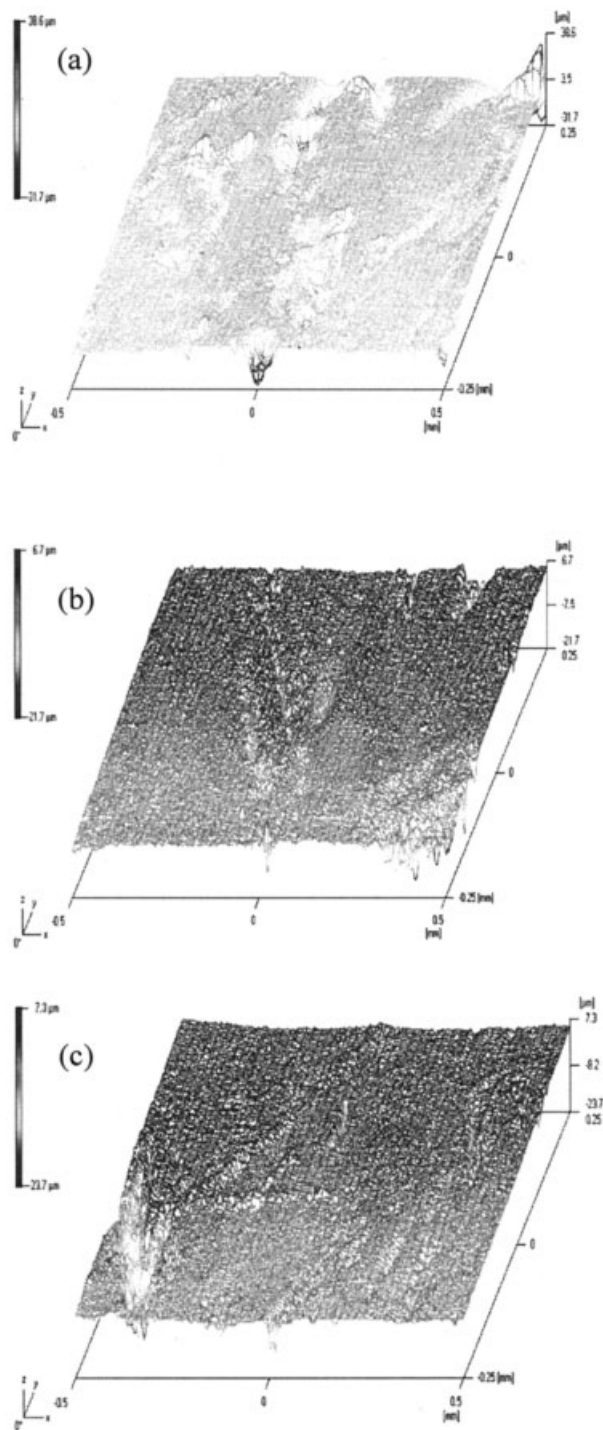
The density was reduced by approximately 30% and decreased to 0.71 g/cm<sup>3</sup> for the hardwood-



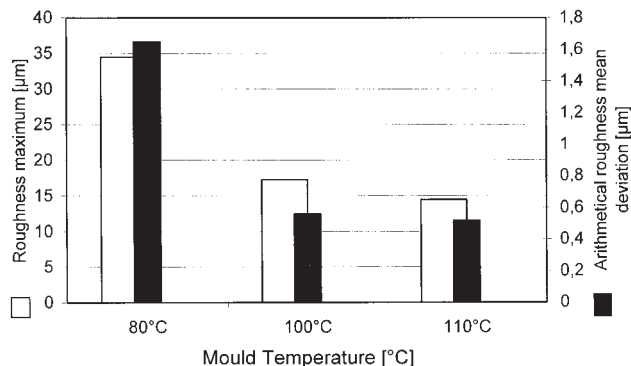
**Figure 6** Influence of the front flow speed on the density and microvoid concentration. The hardwood-fiber concentration was 30%, and the exothermic foaming agent was regular PP.



fiber microfoamed composites (high-melt-flow-index PP) with an exothermic foaming agent. With an increase in the mold temperature from 80 to 110°C, the surface roughness of the foamed composites decreased by nearly 70%.



**Figure 7** Influence of the mold temperature on the surface roughness (laser-sensor photographs) of microfoamed composites: (a) 80, (b) 100, and (c) 110°C. The hardwood-fiber concentration was 30%, and the exothermic foaming agent was regular PP.



**Figure 8** Influence of the mold temperature on the surface roughness (roughness maximum and arithmetical roughness mean deviation) of microfoamed composites. The hardwood-fiber concentration was 30%, and the exothermic foaming agent was regular PP.

An optimum filling quantity could offer raw material savings with better specific mechanical properties at the same time.

## References

1. Reedy, M. E. Proceedings of Blowing Agents and Foaming Processes, Heidelberg, Germany, May 2002.
2. Schut, J. H. *Plast Technol* 2001.
3. Matuana, L. M.; Park, C. B.; Balatinecz, J. J. *Polym Eng Sci* 1998, 38, 1862.
4. Matuana, L. M.; Park, C. B.; Balatinecz, J. J. *Polym Eng Sci* 1997, 37, 1137.
5. Matuana, L. M.; Park, C. B.; Balatinecz, J. J. *J Cellul Plast* 1996, 32, 449.
6. Matuana, L. M.; Mengelglu, F. *J Vinyl Addit Technol* 2001, 7, 67.
7. Matuana, L. M.; Rachtanapun, P.; Selke, S. E. M. *J Appl Polym Sci* 2003, 88, 2842.
8. Matuana, L. A.; Mengelglu, F. *Soc Plast Eng Annu Tech Conf Tech Pap* 2001, 3, 2997.
9. Park, C. B.; Rizvi, G. M.; Zhang, H. Presented at the 5th International Conference on Wood Fiber-Plastic Composites, Madison, WI, 1999; 105.
10. Matuana, L. M.; Balatinecz, J. J.; Park, C. B. Presented at the 5th International Conference on Wood Fiber-Plastic Composites, Madison, WI, 1999; 318.
11. Matuana, L. M.; Mengelglu, F. *J Vinyl Addit Technol* 2001, 7, 142.
12. Matuana, L. M.; Li, Q. *J Appl Polym Sci* 2003, 88, 3139.
13. Zhang, H.; Rizvi, G. M.; Lin, W. S.; Guo, G.; Park, C. B. *Soc Plast Eng Annu Tech Conf Tech Pap* 2001, 2, 1746.
14. Bledzki, A. K.; Faruk, O. *Soc Plast Eng Annu Tech Conf Tech Pap* 2002, 2, 1897.
15. Bledzki, A. K.; Faruk, O.; Zhang, W. Presented at the 5th International AVK-TV Conference for Reinforced Plastics and Thermoset Moulding Compounds, Baden-Baden, Germany, 2002; D5-1.
16. Bledzki, A. K.; Faruk, O. *Cellul Polym* 2002, 21, 417.
17. Bledzki, A. K.; Faruk, O. Presented at the 7th International Conference on Wood Fiber-Plastic Composites, Madison, WI, 2003.
18. Bledzki, A. K.; Sperber, V. E.; Faruk, O. *Rapra Rev Rep* 2002, 13(8), 1.
19. Bledzki, A. K.; Faruk, O. *Compos Part A*, submitted.
20. Holzberg, T.; Luebke, G. Presented at the 7th Technical Conference on New Developments in Extrusion, Wuerzburg, Germany, 2002.