

Influence of Wood Fiber Size on Extrusion Foaming of Wood Fiber/HDPE Composites

G. Guo,¹ Y. H. Lee,² G. M. Rizvi,³ C. B. Park¹

¹Microcellular Plastics Manufacturing Laboratory, Department of Mechanical and Industrial Engineering, University of Toronto, Toronto, Ontario, Canada M5S 3G8

²Center for Biocomposites and Biomaterials Processing, Faculty of Forestry, University of Toronto, Toronto, Ontario, Canada M5S 3B3

³Faculty of Engineering and Applied Sciences, University of Ontario Institute of Technology, Oshawa, Ontario, Canada L1H 7K4

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ABSTRACT: Foaming of wood fiber/plastic composites (WPC) with a fine-celled structure can offer benefits such as improved ductility and impact strength, lowered material cost, and lowered weight, which can enhance their utility in many applications. Although a great deal of attention is now being focused on these composites in the scientific literature, there are still numerous aspects of WPC processing that need elucidation. In this context, this article investigates the effects of wood fiber (WF) size on fine-celled extrusion foaming of WPC in terms of cell size,

cell size distribution, and foam density. The effects of WF size and coupling agent content on the viscosity of WPC are also investigated. The experimental results revealed that the small-sized WF provides a better cell morphology, a smaller cell size, and a better cell uniformity in WPC foams. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 107: 3505–3511, 2008

Key words: wood fiber; extrusion; composites; foaming; viscosity

INTRODUCTION

Wood fiber/plastic composites (WPC) have been commercialized and are replacing wood in many applications, including a variety of building products, automotive, infrastructure, and other consumer/industrial applications, because of their greater durability, low maintenance requirements, lower cost, and better recyclability.¹ However, their potential applications have been limited because of their low impact strength and high density compared to natural wood. Foaming WPC by either chemical blowing agents (CBA),^{2–5} physical blowing agents (PBA),^{6–11} or by stretching,^{12,13} can decrease the density, reduce material cost, and improve their mechanical properties such as impact strength and tensile strength. Producing a uniform fine-celled or microcellular structure in WPC has been demonstrated to be extremely effective at improving their mechanical properties.^{8,9}

During the melting (or plasticating) and subsequent processing stages in the production of WPC, the inherent moisture and volatiles released from

wood fiber (WF), often lead to gross deterioration in the cell structure and the surface quality of the extrudate.¹⁴ Dispersion of water vapor in the polymer matrix would not be good due to the low solubility and high surface tension of water in hydrophobic thermoplastics unless the amount of water vapor is extremely small.^{15,16} As a consequence, a nonuniform cell distribution and a large average cell size usually characterize the obtained foams, which causes the deterioration of properties. Many standard drying methods have been studied to remove moisture effects; namely, inline devolatilization,¹⁰ oven drying, hot air convective drying, drying in K-mixer,¹⁷ and the like. However, the TGA studies on the devolatilization behavior of WF show that even after WF is oven-dried, it still releases about 3% volatiles when the temperature is raised from 110 to 200°C.¹⁴ A strategy of lowering processing temperature to suppress the generation of volatiles has proved to be effective, and the corresponding critical processing temperature has been identified, which minimizes the deteriorating effects of volatiles.¹⁸ When CBA is used for foaming, it is difficult to obtain a fine-cell structure in extrusion of WPC, as a higher processing temperature is needed to decompose the CBA. Compared to CBA-based processing, PBA-based processing (with such as environmental-friendly CO₂ and N₂) does not require a decomposition temperature, apply less expensive blowing

Correspondence to: C. B. Park (park@mie.utoronto.ca).

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agents, and can produce a better cell morphology in general. Therefore, it is more logical to use PBA for WPC foaming; however, it is technologically challenging.

The surface area of WF particles varies for different particle sizes, which can have effects on the viscosity, the cell nucleation, and cell expansion behavior during extrusion foaming of WPC. On the other hand, the surface area may also have an effect on the amount of volatiles generated during processing, which in turn affects all the earlier mentioned parameters, not necessarily synergistically. However, very few studies can be found about the effects of WF particle size on foaming of WPC in literature. Rodrigue et al. studied the effects of wood particles on the cell size and cell density of WPC foams in a twin-screw extruder with CBA.¹⁹ Three different WFs with particle sizes of 45–61, 104–125, and 180–210 μm were used, and the WF content was varied within 0–5 phr. It turned out that wood particles, as a nucleating agent, substantially reduced the cell size and increased the cell density. However, the effect of particle size was not clear in the range of WF content (0–5 phr) studied.¹⁹ In this context, the main purpose of this article is to investigate the effects of WF size on extrusion foaming of WPC with a PBA (i.e., CO₂). Specifically, the effects of WF size on the cell size, the cell size distribution, and the foam density are presented. Since the wood loading levels of 40–70 wt % are typically used in the WPC industry, a fixed WF content (50 wt %) was chosen in this study for providing some qualitative information closer to the practice of the WPC industry. The effects of WF particle size on the rheological property of the molten WPC were also studied.

EXPERIMENTAL

Materials

The matrix material used in this study was high density polyethylene (HDPE) (2710, MFI = 17 g/10 min, Nova Chemicals, Calgary, Canada), and the WF (50 wt %) used was standard softwood (pine) with three grades 2020, 6020, and 12020, supplied by American Wood Fibers (Columbia, MD). Maleic anhydride-*g*-HDPE (PE-*g*-MAn, Fusabond MB-100D, MFI = 2.0 g/10 min, DuPont Canada, Mississauga, Canada, 3 wt %) was used for improving the adhesion between the hydrophobic HDPE and the hydrophilic WF as a coupling agent (CA). CO₂, which is an environmental-friendly PBA, was used in this study to investigate the effects of WF size on the foam expansion and on the cell morphology in PBA-based foaming. All the materials were used as received. The WF size distribution for the three grades is shown in Table I.

TABLE I
Woodfiber Size Distribution

Wood fiber grade	12020	6020	2020
Mesh	120–200 mesh	60–100 mesh	20–60 mesh
Particle size (μm)	75–125	150–250	250–850

Data is provided by American Wood Fibers Inc.

Preparation of WPC

The vacuum-oven-dried WF (at 105°C for 12 h) was dry-blended with the HDPE and the CA. The mixture was melt-blended by using a high shear K-mixer (Werner and Pfleiderer Corp, Stuttgart, Germany) at 180°C. The melt-blended bundles were made into granules by a granulator (C.W. Brabender, Hackensack, NJ). Subsequently, the granules were fed into the counter-rotating intermeshing twin-screw extruder (C.W. Brabender: Model D6/2), which was driven by a 5 hp motor (Reliance Electric: Power Matched/RPM Rectified Power Motor, Greenville, SC). The mixture was uniformly mixed by the intensive counter-rotation of the twin screws. The temperature was maintained at 175°C throughout the extruder in order to devolatilize the extrudate. The extrudate, which came out of the filament die, was cooled by blowing air over it, and was fed into a pelletizer to obtain WPC pellets of uniform size. During the pelletizing, most of the vaporized water and other volatiles, which were released during processing, were purged out to atmosphere.

Uniform dispersion of WF in the polyethylene matrix is rather difficult due to surface incompatibility between the hydrophobic matrix and the hydrophilic WF. The low bulk density and stickiness of WF also causes feeding problem in many processing systems, as WF easily heaps and clogs the passage at the bottleneck of hopper. Hence, uniform feeding of the mixture of polymer pellets and WF requires a carefully controlled feeding rate in order to prevent any cramming in the feeding section of the processing system. Therefore, the use of pelletized WPC improves the WF dispersion in the polymer matrix, and ensures the stable feeding during the extrusion foaming process.

Rheological characterization

The samples were subjected to oscillatory shear in a dynamic stress rheometer (Rheometric Scientific SR-200) (Piscataway, NJ) with parallel plates (diameter = 25 mm, gap = 1 mm). Dynamic frequency sweep experiments were conducted to measure the storage modulus (G'), loss modulus (G''), and complex viscosity (η^*) over a frequency range of 0.1–100 rad/s at 170°C.

Extrusion foaming and characterization of WPC

The WPC pellets were processed in a single-screw, three-zone extruder with a gas injection system

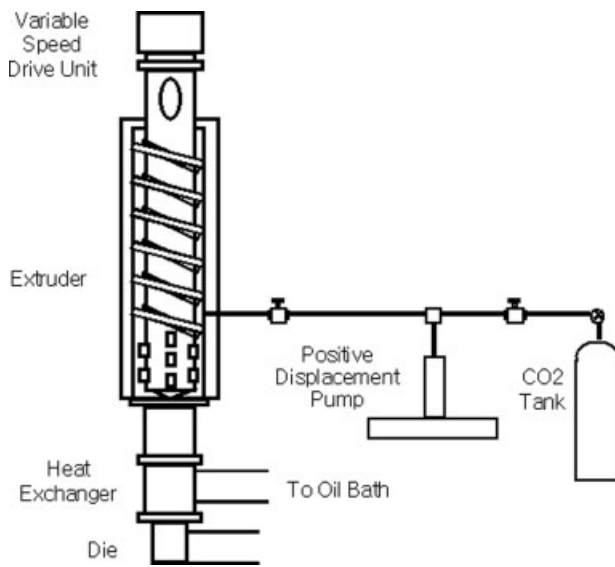


Figure 1 PBA-based single-screw extrusion system.

(Fig. 1). The temperature in this extruder was maintained at 135, 145, and 150°C in Zones 1, 2, and 3, respectively. A static mixer, a heat exchanger and a

filament die (L/D = 0.7"/0.040") were attached downstream of the barrel. Their temperatures were set at 150, 150, and 150°C, respectively. The heat exchanger and die temperature were reduced at an interval of 5°C from 150°C to the lower temperatures, in order to reduce the composite density by preventing gas loss from the extrudate. The foam samples were collected at each 5°C interval from 150°C when the stable processing conditions were obtained, and characterized for foam density and cell morphology.

For cellular morphology, each sample was first dipped in liquid nitrogen and then fractured. The fractured surface was then gold-coated, using a sputter coater (E 50000C PS3), and the microstructure was examined using a scanning electron microscope (SEM, Hitachi 510) (Schaumburg, IL).

RESULTS AND DISCUSSION

Effects of WF size on viscosity

The variation of complex viscosity (η^*) as a function of angular frequency for WPC with different WF

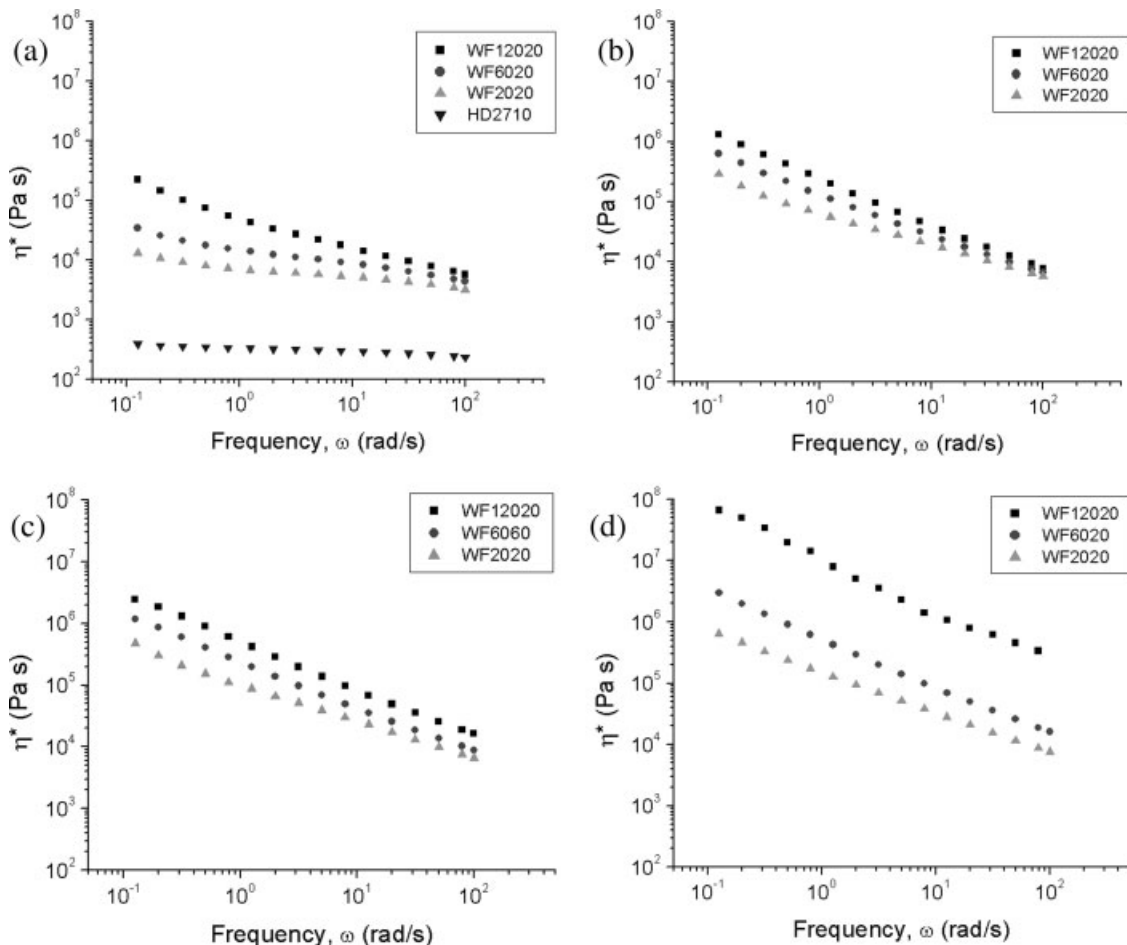


Figure 2 Effects of WF grade on the complex viscosity of HDPE/WF composites with different coupling agent contents: (a) 0%; (b) 3%; (c) 6%; (d) 10%.

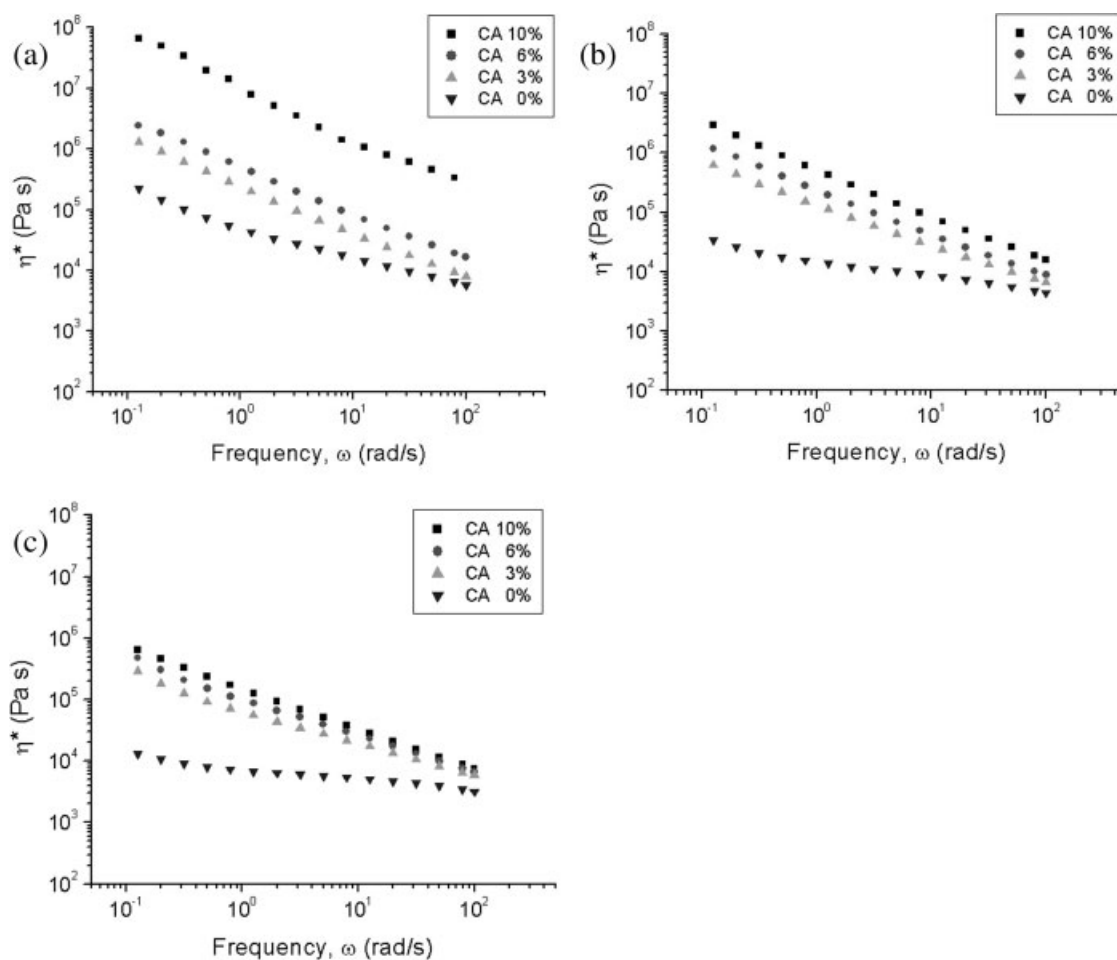


Figure 3 Effects of coupling agent content on the complex viscosity of HDPE/WF composites with different WF grades: (a) 12020; (b) 6020; (c) 2020.

sizes and different CA contents is shown in Figure 2. The change in shape of the flow curves shows strong shear-thinning behavior for WPC with all WF grades. This shear-thinning behavior is due to alignments of WF particles and disentanglements of the long polymer chains during shearing. Compared with the neat HDPE, the viscosity of WPC significantly increased, as shown in Figure 2(a). This was due to the interactions amongst the WF particles and the polymer matrix. Figures 2(a–d) clearly show that the small size WF (Grade 12020) composites with the same CA content consistently display the higher viscosity in the entire frequency range due to the largest surface area offered by the small size WF particles. Since the WF content was fixed (50 wt %), the decrease of WF particle size would result in an increase of the WF particle number and the total surface area. This would increase the degree of particle-particle and particle-matrix interactions. These two types of interactions may have some mixed effects on the viscosity. On the one hand, the increased degree of particle-matrix interactions would increase

the melt viscosity. On the other hand, the increased degree of particle-particle interactions may affect the viscosity two different ways. First, the rigid particle-particle contact will cause a higher resistance and thereby increase the viscosity. However, the particle-particle interactions may increase the tendency for WF particles to form some large agglomerates through strong intermolecular hydrogen bonding. This will lead to a poor dispersion of WF in the polymer matrix and thereby a poor encapsulation of the WF particles with the polymer melt, which may decrease the melt viscosity of the polymer/WF mixture. In Figure 2(a), at the CA content of 0%, the smallest size WF composites show the highest viscosity, which indicated that most of the WF particles were encapsulated by the polymer melt and that the particle-matrix interactions took a dominant role in influencing the melt viscosity. It is well-known that CA plays a role of strengthening the interfacial bonding between the WF and the polymer matrix, as well as helping the dispersion of WF particles in the polymer matrix.²⁰ As a result, the increase of the CA

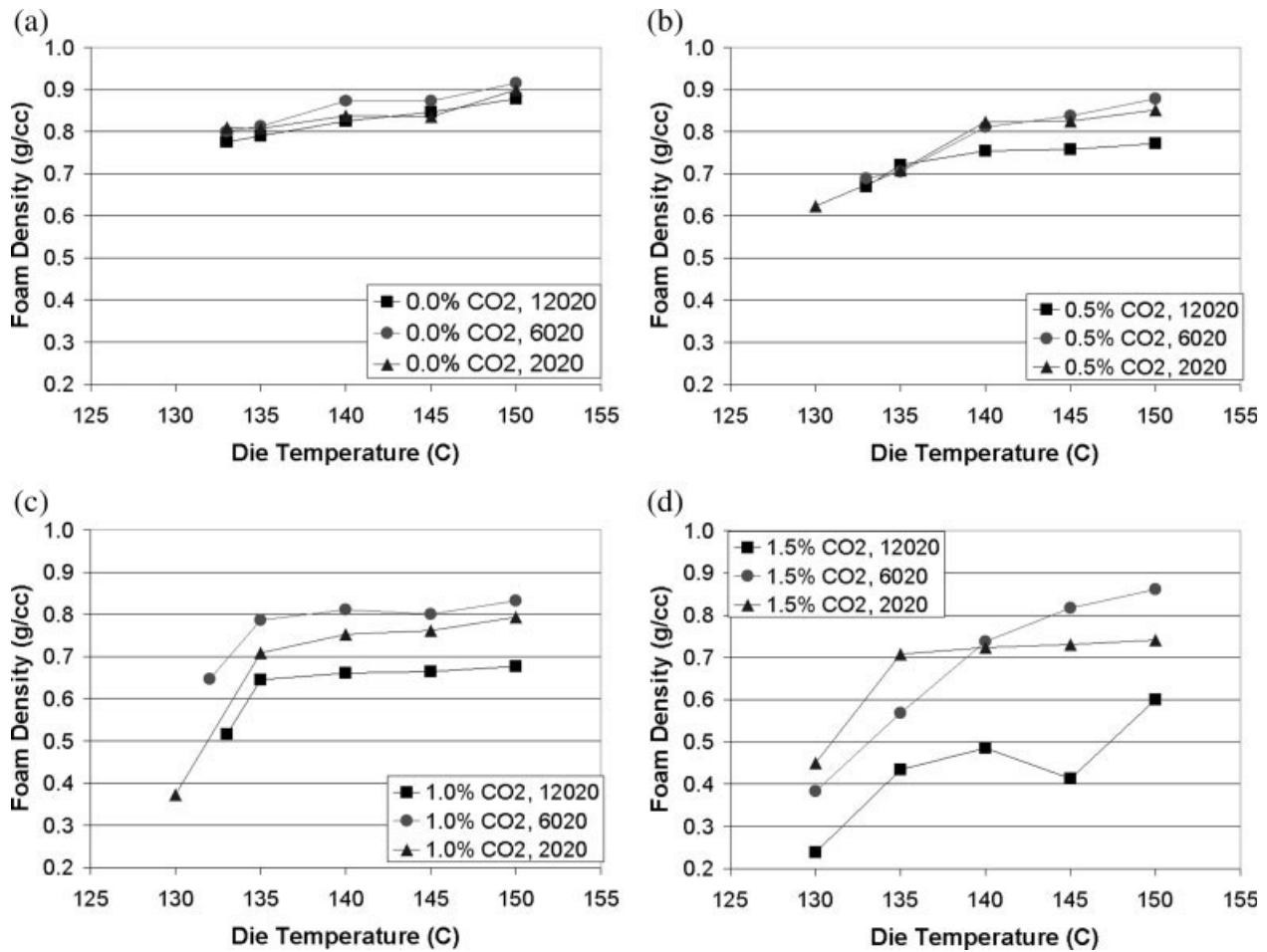


Figure 4 Foam density as a function of die temperature for WPC with different WF grades at various contents of gas injection: (a) 0.0% CO₂, (b) 0.5% CO₂, (c) 1.0% CO₂, and (d) 1.5% CO₂.

content leads to the increase of viscosity, shown in Figure 2(a–d). It is also worth to note that the viscosity difference for the different size WF composites becomes smaller at the high frequency (e.g., 100 rad/s), this could be attributed to the improved alignment of the polymer chains and WF at the high shear rate. Figure 3 shows the effects of CA content on the complex viscosity of WPC with different WF grades. For the large size WF composites (WF grade 2020) in Figure 3(a), 3% CA results in a significant increase of the viscosity. However, with the further increase of CA content to 10%, there is no significant viscosity increase. This implies that 3% CA is enough to coat the 2020 WF particles. As the WF size decreases for the WF (6020) composites shown in Figure 3(b), the surface area increases, and therefore, it requires more CA. This is indirectly indicated by the viscosity difference (the gap between the lines) increases. Therefore, the small size WF composites need more amount of CA for the WF particles, as they have a larger surface area. The viscosity graph could provide the indirect information about the proper CA content.

Effects of WF size on foam density

The density variations of the foamed WPC with varying die temperatures are shown in Figure 4. With no gas injection, the density reduction, which results from volatiles generated from WF during processing, seems to be insufficient and the obtained density range is 0.8–0.9 g/cc. The composites with all WF grades show the similar density reduction behavior. As the gas content increases, the density reduction becomes larger. Moreover, the composites with the 12020 WF display a bigger density reduction, which could be due to the fact that they release more volatiles because of the larger surface area of smaller but numerous WF particles. Another reason could be that the small WF particles provide more nucleation sites than the larger WF particles, thereby enhance the heterogeneous nucleation.^{21,22} As the die temperature approaches the melting point of HDPE (about 133°C), the rate of density reduction increases sharply, indicating the formation of a solid skin of the extrudate which prevents gas escape due to the decreased gas diffusivity.^{14,23}

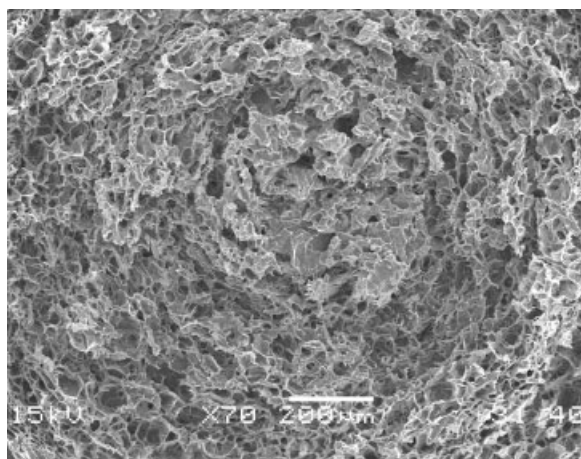


Figure 5 Typical cell morphology of WPC for fine WF grade 12020 at die temperature 130°C and the gas content of 1.5%.

Effects of WF size on cell morphology

Figure 5 presents the typical scanning electron micrograph for WPC with the WF grade 12020 at the die temperature of 130°C and the gas content of 1.5%. An extremely fine-celled structure with a very uniform cell size distribution having an average cell size of less than 50 μm , was successfully obtained. Figure 6 shows the effects of gas content on cell size and cell size distribution at the die temperature of 150°C. As the gas content increases from 0.0 to 1.5%, the cell size becomes smaller, and the cell uniformity becomes better, indicated by the steeper curves. This could be so because the increased gas content reduces the viscosity, thereby enhancing the mixing effects and making the gas concentration in polymer more uniform. Figure 7 shows the effects of die tem-

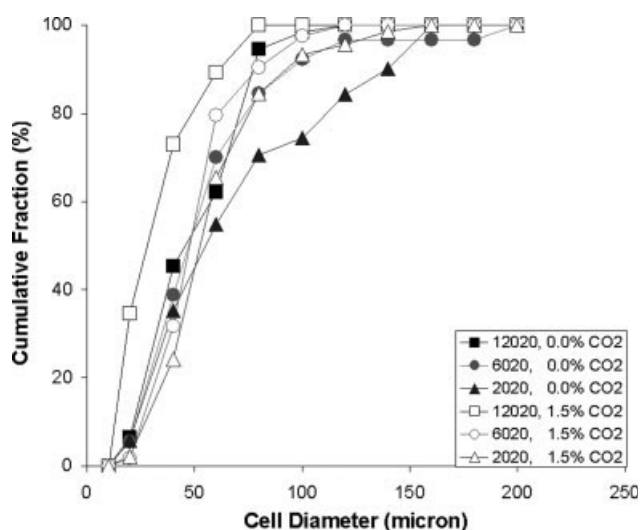


Figure 6 Effects of gas content on cell size and cell size distribution at die temperature of 150°C.

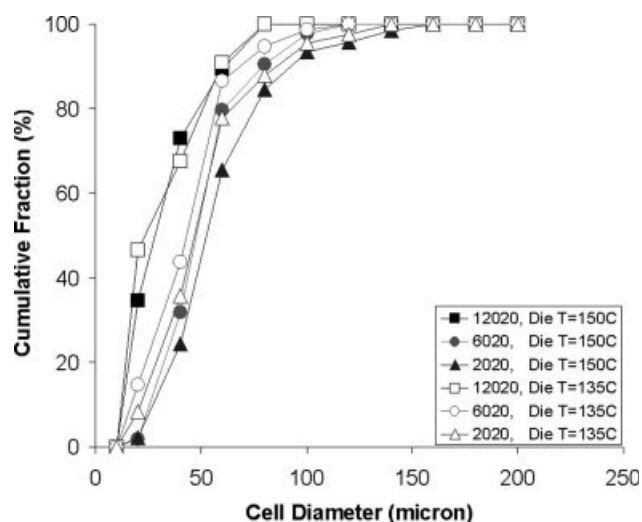


Figure 7 Effects of die temperature on cell size and cell size distribution at the gas content of 1.5%.

perature on cell size and cell size distribution at the gas content of 1.5%. As the die temperature decreases from 150 to 135°C, the cell size becomes slightly smaller and the cell uniformity becomes better. This could be because the low die temperature enhances the melt strength, significantly reducing cell coalescence.¹⁴ In both Figures 6 and 7, the small-sized WF composites (12020) show a smaller cell size, and the majority of cells have the cell size below 50 μm . The small-sized WF composites also demonstrate more uniform cell size distribution indicated by the steeper curve and the narrower cell size distribution range. This could be due to the heterogeneous nucleating effects of fine WF particles.

CONCLUSIONS

The small-sized WF (12020) composites demonstrated a better cell morphology, with a smaller cell size and a better cell uniformity. This could be because the finest WF enhanced the cell nucleation. A very fine-celled structure, with the majority of cells smaller than 50 μm , and a desired density range of 0.4–0.8 g/cm^3 was successfully obtained. The small-sized WF composites also have a higher complex viscosity, as the small WF particles offer a larger surface area for particle-particle and particle-matrix interactions. Introducing CA reduces the particle-particle interaction, but enhances the particle-matrix interfacial bonding, which increases the viscosity. In addition, the viscosity data vs. the CA content could give information about the proper content of CA to be used in WPC.

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