Manufacture of Integral Skin PP Foam Composites in Rotational Molding*

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ABSTRACT: The feasibility of applying the single-charge rotational foam molding processing principle to the fabrication of integral skin polypropylene (PP) foams comprising a PP solid skin and a PP foamed core is investigated in this study. A systematic process interruption and sample evaluation approach was used to quantify the experimental results and explore possibilities for improving the process control strategies to ultimately achieve a desired homogeneity and thickness uniformity of the solid PP skin layer that would be fully encapsulating the PP foamed core of a desired cell population density and average cell size. The experimental results revealed that this is quite a challenging task, not only because of the well-known intrinsically poor foaming nature of PP due to its low melt strength at elevated temperatures, but also because, in single-charge rotational molding, the processing parameters are often conflicting with each other and therefore, have to be optimized within a very narrow processing window. However, simultaneous, single-charge, quality PP integral skin and foamed PP core formation in rotational foam molding is feasible. Optimizing the heating profile, heating rate, heating time, and the mold

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rotational speed as well as careful selection of PP resins (or resin blends), chemical blowing agents (CBA), and their composition formulations is strongly recommended.

KEY WORDS: rotational molding, plastic foams, polyethylene, polypropylene, composites, chemical blowing agents, integral skin, core.

INTRODUCTION

Foamed rotational moldings comprising of a solid skin that encapsulates the cellular structure offer multiple advantages over conventionally manufactured rotational moldings (Figure 1). Unlike respective polyurethane filled conventional rotational moldings that usually act as two separate bodies and show disruptive borders around the adjacent incompatible polymer textures, these integral skin foamed moldings are, and act as, single bodies thereby being characterized with a substantially added stiffness and durability as well as significantly more uniform thermal properties. Therefore, integral skin foam composites are very well suited to satisfy quite demanding requirements even when being exposed to most severe service environments (e.g., mooring marine buoys).

The enhanced properties of such integral skin cellular composites are normally attributed to the continuous and seamless interface between the distinct external solid skin layer and a smooth transition into the gradually developing foamed core structure (Figure 2). Such uninterrupted interlayer interfaces can be best achieved by creating both the skin and the foamed core layers in a concurrent fashion, i.e., simultaneously within the same rotomolding cycle (single-charge processing principle), and while using same or different but compatible



Figure 1. Cross section schematic of an integral solid skin and foamed core rotationally molded composite.



Figure 2. SEM of a PE solid skin-PE foamed core interface.



Figure 3. SEM of a PE solid skin-PP foamed core interface.

basic polymeric resins (or resin blends) to formulate both the nonfoamable and foamable materials for the layers, respectively. Since, as a direct consequence of this concept, different resins can be used for forming the foam layer and the skin, the production of integral articles with substantially different properties between the core and the skin is feasible. Figure 3 presents a scanning electron microscope (SEM) R. POP-ILIEV ET AL.



Figure 4. Schematic illustration of the principle of operation of the single-charge rotational foam molding process for manufacturing integral skin cellular composites.

micrograph of the typical interface morphology between a single-charge made polyethylene (PE) solid skin and a PP foamed core.

The main feature of the single-charge processing principle is the simultaneous introduction of both the foamable and non-foamable resins into the cavity of the mold at the outset of the rotomolding cycle (Figure 4). Although this advanced concept eliminates the need for process interruptions and the use of drop boxes or plastic bags inside the mold, it requires an appropriate process control strategy that would assure that the execution of the adhesion of the non-foamable thermoplastic resin to the internal surface of the mold always takes place prior to the activation of the foamable resin. The commonly used approach towards solving this problem includes charging the mold with pulverized non-foamable resins intended to form the skin and pelletized foamable resins intended to subsequently form the foamed core or layer.

BACKGROUND

Mori et al. [1] disclosed a method for producing foamed shaped articles in single-step rotational foam molding. The key to this invention is the use of resins having different heat capacities.

Needham [2,3] disclosed a method that uses two resins of significantly different particle sizes for charging the mold. The principle used guarantees that the resin having the smaller particle size will form the outer skin while the powder having the greater particle size will form the inside layer or the core.

Strebel [4–6] disclosed a compounding-based single-step method that uses two distinct thermoplastic resin components. The first component is charged into the mold in a pellet form. It is obtained by extrusion melt compounding of the ethylene polymer with a CBA. The second component is charged into the mold in a powder form and is in fact a blend of ethylene polymer powders with different particle sizes and melt indexes.

Lee [7] disclosed molding compositions essentially consisting of a mixture of a skin-forming component and a foam core-foaming component physically dry-blended together. The skin-forming component comprises two distinct phases blended together, a high zero-shear viscosity material and a low zero-shear viscosity material. The foam core-forming component comprises a foamable polymeric material including a suitable blowing agent. The three materials are selected from suitable homopolymers and copolymers of ethylene.

Recent research works related to single-charge integral skin polyolefin foaming in rotational foam molding [8–14] have demonstrated that by implementing the single-charge rotational foam molding method [8–10] along with a two-step oven temperature profile [10-12], and a highspeed mold rotation processing strategy [13,14], polyethylene (PE) based PE/PE integral skin foams (PE solid skin/PE foamed core) can be successfully manufactured.

In addition, isolated attempts to manufacture PE/PP (PE solid skin/ PP foamed core) integral skin rotational foam moldings have also been reported [12,13].

Conversely, a strong motivation exists for exploring the manufacturing prospects of even more robust and stiff integral skin rotational moldings that would be suited to withstanding service temperatures above 125°C. Taking into consideration the advantages of PP over PE as a basic resin and the current state-of-the-art technology in the manufacture of PP foams in rotational foam molding [13–19], it would be desirable to develop integral skin PP based foam composites in rotational molding, i.e., PP/PP (PP solid skin/PP foamed core). However, it is important to be aware that, based on previous research on skinless PP foams, it has been found that in general: (i) PP foams in rotational foam molding achieve significantly lower cell densities and larger average cell sizes in comparison with respective PE foams, (ii) unlike in PE foams, the dry blending based technology is almost equally or even better suited than the melt compounding based technology to the production of PP foams in rotational foam molding,

(iii) successful foaming of PP in rotational foam molding can only be performed over a very narrow range of melt temperatures that are close to the melting point of the polymer and by using PP grades belonging to a limited range of melt flow rate (MFR), (iv) desirable (finer celled) PP foam structures in compounding based rotational foam molding could be obtained only if pellet sintering takes place prior to the decomposition of the CBA and if the processing temperature during the foaming process is kept lower than the temperature of cell coalescence, and (v) higher melt strength PP grades having a branched molecular structure and high viscosity, i.e., low MFR (e.g., 5.5 g/10 min) demonstrated the most desired rotofoamability among the investigated PP grades [20].

EXPERIMENTAL

Materials

Foamed Core-forming Basic PP Resins

Dedicated PP grades for foaming in rotational molding are not available. Thus, taking in consideration previous knowledge about the rotofoamablity of PP grades as a criterion [20], four high melt strength (HMS) propylene-based resins were selected to participate in the framework of the present experimental study. These include: Daploy WB130 HMS (Borealis), a structurally isomeric polymer designed for foam extrusion, Daploy WB120 HMS (Borealis) a specially designed structurally isomeric extrusion grade polymer particularly suitable for the conventional blown film process with improved bubble stability, Pro-faxTM PF814 HMS (Basell) PP for low density foam extrusion, and Pro-faxTM SD632 HMS (Basell), a PP copolymer resin designed for the production of low density foam extruded products having stable cell growth during expansion. Table 1 presents the basic properties of the selected PP resins.

Commercial name	Company	Density (g/cc)	MFR (g/10 min)
WB130	Borealis	0.900-0.910	2.5
WB120	Borealis	0.900-0.910	2.5
PF814	Basell	0.902	2.5
SD632	Basell	0.902	3

 Table 1. Properties of the PP materials used for compounding foamable pellets for the core-forming component.

Commercial name	Gas yield (cc/g)	Decomposition temperature (°C)
Celogen-AZ	210	210–220

Table 2. Properties of the CBA used for forming foamable resins.

Table 3. Properties of the PP material used for the formation of non foamable skin-forming component in powder form.

Commercial name	Company	Density (g/cc)	MFR (g/10 min)
MT4390	Basell	0.900	20.0

Each of the selected four PP-basic polymer materials was used for preparing melt compounded foamable compositions for six-fold expansion with 1.83 wt% Celogen AZ. Table 2 presents the basic properties of the selected CBA.

Skin-forming PP Resin

MT4390 HU (Equistar Chemicals) is a PP copolymer resin of a rotomolding grade and was thus supplied in a powder form. Table 3 presents the basic properties of MT4390.

Rotational Foam Molding Experimental Setup

A uniaxial, lab-scale, custom-made rotational molding machine served for conducting rotational foam molding experimentation. The rotating speed of the arm could be continuously, electronically controlled from 0 to 30 rpm.

Rotating High-temperature Wall Setup

Figure 5 presents the schematic of the experimental setup developed for investigating the adhering behaviors of polymer powders and pellets to a high temperature wall. It is, in fact, a custom made aluminum hot plate uniformly heated within $\pm 1^{\circ}$ C accuracy which was conveniently attached on the existing rotational foam molding setup so that it could rotate simulating accurately the rotation acceleration of the actual mold.



Figure 5. Experimental setup for investigating the sticking behavior of PP powders to a hot rotating mold wall.

RESULTS AND DISCUSSION

Multiple series of parametric search single-charge integral skin rotational foam molding experiments were conducted to determine the optimal duration of heating time for each PP material skin-core combination. The preprocessing experimental procedure included rotating uniaxially the charged mold for 5 min in each direction at the maximum rotational speed (30 rpm) to allow the migration of nonfoamable polymer powder particles towards the internal surface of the mold. During actual processing, the oven temperature was kept constant ($T_{\rm oven} = 300^{\circ}$ C), and the rotational speed of the mold was kept at 10 rpm. After the heating time elapsed, the mold was externally cooled with forced air for 20 min.

Although best results were obtained from the WB130/MT4390 combination, the experimental results generally revealed an unsatisfactory morphology of the PP solid skin layer which was typically consisting of a distinct thin layer of solid skin having a rough non-uniform thickness. Interestingly, unlike the processing behavior of PE/PE based integral skin foams, no foam invasion spots into the solid skin were observed. It was hypothesized here, however, that since PP based materials (having relatively significantly higher melting temperatures than PE based materials) were used for both the solid skin-forming and the core foam-forming material, the foamable PP pellets continued to tumble on top of the already formed PP skin layer, failing to remain attached to the skin, thereby



Figure 6. Sticking behavior of PP powders (MT4390) to a hot rotating mold wall. The critical sticking temperature of MT4390 is 144° C.

permanently avoiding the hottest region within the mold, which would otherwise result in their CBA content activation. An additional reason why their CBA contents were not prematurely activated was the relatively high onset decomposition temperature of Celogen AZ (210 $^{\circ}$ C), and thus, there was no evidence that the non-foamable skin-forming PP melt was pushed inwards by the expanding core-forming PP cellular structure. This important hypothesis was experimentally proved by conducting rotating hot mold wall sticking experiments using the PP resin selected for skin formation (MT4390) in powder form. The resulting plots presented in Figure 6 indicate that the critical sticking temperature on a hot rotating mold wall for MT4390 powders is 144°C, which is the mold's wall temperature at which the PP skin is completely formed, but still well below the activation temperature of the CBA. In summary, there is no need for applying a two-step oven temperature profile when processing PP/PP integral skin foams.

In order to characterize and quantify the thickness uniformity of the skin, a common method of specifying the surface finish or roughness, root mean square (RMS or R_q) [21] was used. The skin thickness of each sample was measured at 35 points (Figure 7),



Figure 7. Illustration of the root mean square (RMS) method parameters used for determining skin thickness uniformity.

and as the projected average skin thickness was 3 mm, the RMS value to denote skin thickness uniformity was calculated using Equation (1),

$$R_{\rm q} = \sqrt{\frac{1}{N} \sum \left(a - 3\right)^2} \tag{1}$$

where R_{q} , N, and a refer to the RMS value, number of points, and sample thickness at each point, respectively.

Similarly to PE/PE integral skin processing, it has been found that an increased mold rotational speed can be utilized to improve the uniformity of the skin layer. Basically, the higher the mold's RPM the lower the RMS coefficient and the higher the PP skin thickness uniformity. Figures 8 and 9 illustrate this relationship for WB130/MT4390 foamed composites. This processing strategy even eliminates the need for pre-tumbling of the mold outside the oven before in-oven processing. The rationale behind the positive effects obtained from increasing the rotational speed of the mold (up to 30 rpm) is based on the fact that the heat capacity difference and the increased mechanical tumbling make smaller powder particles tend to be heated and softened faster while the heating of the bigger foamable pellets is being suppressed. This indicates that high speed rotation of the mold is favorable for the powders sticking on the wall, i.e., skin formation prior to the sticking of foamable



Figure 8. Effect of the rotational molding speed on both the skin and foamed core quality of WB130/MT4390 integral skin foams.



Figure 9. Relationship between skin thickness uniformity (RMS) and the mold RPMs in rotational foam molding of WB130/MT4390 integral skin foamed composites. The higher the RPM the lower the RMS coefficient.

pellets on the wall. Figure 10 presents the effect of the processing time on both the skin and foamed core quality of WB130/MT4390 integral skin foamed composites at a high rotational molding speed of 30 rpm.



molding speed of 30 rpm.

CONCLUSIONS

Single-charge fabrication of integral skin PP solid skin/PP foamed core cellular composites in rotational foam molding is feasible. Unlike the processing of PE/PE integral skin analogs, there is no need for applying a two-step oven temperature profile when processing PP/PP integral skin foams. Similarly to the processing of PE/PE integral skin analogs, boosted mechanical agitation from the increase of mold rotational speed causes a deeper contrast between the degree of heating for skin and that of heating for foam material. Thus, the PP skin formation process becomes completely separated from the PP foam formation process which occurs with a desired delay. Further processing strategy refinement is necessary.

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