Influence of Azodicarbonamide on Physical and Mechanical Behaviours of Thermoplastic Blend

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Abstract. Foaming technology is promising in producing foamed materials which can offer benefits such as lowering the cost and weight of materials as well as their utility in many applications. In this study, the thermoplastic blend of recycled high density polyethylene and recycled polyethylene terephthalate was foamed with azodicarbonamide chemical foaming agent (CFA) at 0.5-2.0 phr. The blending of thermoplastic was carried out via extrusion and the foaming process occurred during hot/cold pressing. The cell morphology, density and mechanical properties were examined. Field emission scanning electron microscope (FESEM) micrograph showed that the thermoplastic blend foamed with 1.5 phr CFA had the cellular morphology structure with non-collapsed cells and better cell distribution. As the CFA content increased, the density of blend foams decreased accordingly. The minimum density was achieved at 1.00 g/cm³ for 2.0 phr ADC. Tensile and bending properties generally decreased with the content of CFA added. The impact strength of blend foams increased when the CFA added up to 1.0 phr and then reduced with further CFA content. It can be concluded that the fabricated foamed blend can give value added to the recycled polymers.

1. Introduction
The public awareness on environmental pollution and the increasing cost of raw materials have attracted industries’ attention in exploring ways to fabricate lightweight products with desirable properties by using less material [1]. In this context, foamed materials have been investigated for replacing the solid counterparts as foaming technology can reduce the materials’ consumption, and hence lead to the reduction of weight and density as well as the production cost [1, 2]. Polymeric foams, so-called cellular polymers are defined as two-phase materials where a gas dispersed in a continuous macromolecular phase [3, 4]. Practically, polymer foams are applied in various fields, such as packaging materials, automobile industry, sports equipment and building materials, owing to the low cost and weight, good mechanical properties as well as energy insulation and high energy absorption [5].

Chemical blowing agent or chemical foaming agent (CFA) can be defined as a chemical compound that is used to promote foaming process in which the gas is generated via its thermal decomposition [4]. In literature, it was established that there are various CFA used for different types of thermoplastics. Chang et al. has investigated the mechanical and surface properties of polypropylene (PP) blown with 1-9 wt% endothermic CFA masterbatch, and they demonstrated that the best cell uniformity with the smallest average cell was at 5 wt% CFA [1]. Sodium bicarbonate is one of the endothermic CFAs which has been
added into natural rubber at 4-12 phr; the findings showed that the amount of decomposed carbon dioxide gas increased with the concentration of foaming agent and hence increasing the number of cells per unit volume with smaller average cell sizes [6]. On the other hand, for exothermic CFA, azodicarbonamide is one of the interesting chemical compounds because it decomposes at a relatively low temperature and produces closed-cell foam [7] for neat polymers and composites. Stange and Münstedt reported that with the use of azodicarbonamide, a remarkable enhancement of the foaming behavior where a higher expansion ratio and more homogeneous cell size distribution with a lower amount of connected cells are found with the increasing content of the long-chain branched-PP up to 50 wt% [8]. In the study of Bledzki et al., an exothermic CFA was used to foam PP/wood fibre composites and it was found that 2 wt% CFA produced a finer cellular structure and the density was reduced up to 30% [9].

In spite of numerous researches on polymer foaming, very few reports on the polymer foams based on polymer blending were found. In this research, the recycled polymer blend made from post-consumer high-density polyethylene and polyethylene terephthalate was used for foaming. The focus of this work was to investigate the effect of azodicarbonamide CFA content varying from 0 to 2 phr on the morphology, density and mechanical properties of recycled thermoplastic blend.

2. Experimental Section

2.1 Materials

Recycled high-density polyethylene (RHDPE) and recycled polyethylene terephthalate (RPET) used in this study were supplied by Bio Composites Extrusion Sdn. Bhd. The RHDPE has a melt flow index of 0.072 g/10 min (at 190˚C) and a density of 923 kg/m³. The Lotader AX8840 type of ethylene glycidyl methacrylate (EGMA) with a melt index of 5 g/10 min (190˚C, 2.16 kg) and a glycidyl methacrylate content of 8% was used as compatibilizer. Chemical foaming agent (CFA) used in this study is azodicarbonamide foaming agent, and zinc oxide (ZnO) foaming agent decomposition stabilizer were provided by Sigma Aldrich.

2.2 Method

Prior compounding, the RPET was oven-dried at 90˚C. Figure 1 shows the experimental section for fabrication of recycled thermoplastic blend foams. The RHDPE, RPET and EGMA were melt-compounded in a co-rotating twin screw extruder (Thermo Prisma TSE 16PC) with an extrusion temperature profile of 250-270-240-190˚C and a screw speed of 30 rpm. The composition of RHDPE/RPET/EGMA was fixed at 75/25/5 (wt/wt/phr). The extrudates were taken for second extrusion with CFA at 170-215-210-195˚C at the same screw rotation speed. The CFA content was varied at 0.0, 0.5, 1.5 and 2.0 phr. After extrusion, the extruded granules were compression molded at 200˚C under 1000 psi using a hot/cold pressing machine (LP50, LABTECH Engineering Company LTD). The periods of hot press and cold press were set to 10 min and 5 min, respectively.

For structural observation, field emission scanning electron microscopy (FESEM) was performed using LEO 1450 VP with an operation accelerating voltage of 14 kV. The density measurement of thermoplastic blend (either solid or foamed ones) was determined manually as: density = mass per unit volume. The tensile testing was done on 3 mm thickness dumbbell specimens (ASTM D638-03 (type I)) using a universal tensile machine (Model Testometric M500-50CT) at 5 mm/min crosshead speed. The Izod impact testing was performed using Ray-Ran Universal Pendulum Impact System (ASTM D256-05), with a velocity of 3.46 ms⁻¹. Bending measurement was determined using the same machine as tensile test, but the sample dimension was prepared follow the standard ASTM D790-03.
3 Results and Discussion

Figure 1. Schematic experimental section for fabrication of recycled thermoplastic blend foams.

Figure 2. FESEM micrograph of thermoplastic blend foams with (a) 0.0 phr, (b) 0.5 phr, (c) 1.5 phr and 2.0 phr of CFA.
Figure 2 shows morphological structure of thermoplastic blend foamed with different content of CFA at (a) 0, (b) 0.5, (c) 1.5 and (d) 2.0 phr. In Figure 1(a), without CFA, the solid thermoplastic blend displayed a compact structure. By adding CFA at low content (0.5 phr), the surface morphology of the specimen showed the effect of foaming as proven by the presence of the dents. In this case, the foaming effect seems not stable and sufficient as the foam cells formed are not perfect circular shaped. This indicates the content of CFA at 0.5 phr was insufficient in this blend. When the CFA was added at higher content, the foam cells are more stabilized. Amongst all the CFA contents, the blend with 1.5 phr CFA displayed the most satisfied structure with having perfect spherical foam cells with no coalescence and less variety of cell sizes, whereas, the blend with 2.0 phr CFA had an inhomogeneous distribution and collapsing of the foam cells. The coalescence between cells (bubbles) could be due to the high amount of gas produced from CFA [7]. The observations in FESEM micrograph are tally with the density measurement shown in Figure 3. In which the density results showed the downward trend of density with the increasing CFA content. The reduction of density is ascribed to the increasing number of cells generated (Figure 2). However, the reduction of density was insignificant (up to only 6%) and this is possibly due to the low uniformity of the foam cells in the blend. As shown in FESEM micrograph, the cells not fully distributed throughout the entire surface.

Figure 3. Density measurement of thermoplastic blend foams with different content of CFA.

Figure 4 presents the mechanical properties of thermoplastic blend as a function of CFA. The model of Gibson and Ashby stated that the tensile modulus of foams are determined by the factors of foam density, tensile modulus of the material used and foam structure [10]. In general, it is found that as the density of composite decreased upon the addition of CFA, the tensile and bending strength and modulus also decreased. For the solid blend, the mechanical properties were recorded at 19.1 MPa for stress at yield (tensile strength), 351.5 MPa for Youngs Modulus (tensile modulus), 29.9 MPa for bending strength, and 1166.6 MPa for bending modulus. A gradual linear reduction was shown for tensile and bending properties (except for Youngs Modulus) and the lowest properties (maximum reduction by about 8-18 %) were exhibited by 2.0 phr CFA. This phenomenon has been similarly reported in previous research regarding the neat polymer foams and polymer composite foams [9, 11]. This is expected as the voids resulting from foaming process could adversely affect the mechanical properties [11], especially in the case of non-uniform and non-homogenous cell sizes and distribution in the structure. The tensile and bending strength and modulus decreased with the rising of CFA content. In fact, the foamed blend with higher content of CFA generates larger voids which act as crack creation sites that may severely deteriorate mechanical properties (i.e. stiffness) [1]. For the modulus, especially tensile modulus (Figure 4 (b)), the negative effect by CFA content was inconsistent. Nevertheless, the lowering of modulus of the blend resulted in the higher impact strength and the softness of the final foam product [10].
Figure 4. Mechanical properties of thermoplastic blend foams with different content of ADC.

Interestingly, the strain at break and impact strength showed some improvements with the addition of less than 1.5 phr CFA. The maximum strain at break (16.2 %) and impact strength (1.4 kJ/m²) were shown for samples with 1.0 CFA. According to Chang et al., the impact strength is governed by several parameters like cell size, skin thickness and void fraction [1]. The trend of earlier increase in strains at break and impact strength has been published in the previous work on polypropylene foam with 5 phr CFA [1].
unique properties of foamed material are owing to the outstanding energy-absorbing characteristics induced by the presence of the gaseous phase [6]. The latter reduction implies the weakened ability of the foam cells to absorb impact energy as a result of increasing number of cells per unit volume which led to poor crosslink density. It is stated that the properties of the foam product will be deteriorated when cells with large size are present. This is because these cells will have lower cell wall thickness and the cell wall is more likely to collapse and rupture when a force is applied [6]. This can be obviously observed in foamed blend with 2.0 phr CFA.

4. Conclusions

The solid and foamed RHDPE/RPET/EGMA (75/25/5) thermoplastic blends were prepared via melt-blending technique and followed by compression molding. The effect of exothermic azodicarbonamide content varied from 0.5 to 2.0 phr on the microstructure, density and mechanical behaviours of blend foams was investigated. By increasing azodicarbonamide content, the amount of cells formed in the blend structure increased, thereby led to the reduction of blend density. With the presence of 2.0 phr azodicarbonamide, the cells structure collapsed as shown in FESEM micrograph. From tensile and bending results, the strength reduced by 17-19% and modulus by 8-14% for foamed thermoplastic blends with respect to azodicarbonamide content. Whilst, the strain at break and impact strength of the blend foams showed an optimum increment at 1.0 phr before decreasing trend with higher azodicarbonamide content. The findings of this study preserve a good preliminary research in polymer foaming for further improvement in weight reduction while maintaining or even improving the material performance.

5. Acknowledgements

The authors are grateful for the supports provided by Electron Microscopy Unit of Universiti Kebangsaan Malaysia (UKM) and UKM Research Grant Scheme (GGPM-018-061).

Reference

[1] Chang E, Mahmud M B, Li X, Mohebbi A, and Park C B. Optimizing chemical blowing agent content in foam injection molding process of polypropylene. in SPE-ANTEC meeting, Society of Plastics Engineers, Anaheim, CA, USA. 2017.
[2] Hassan N A A, Ahmad S, Chen R S, Zailan F D, and Shahdan D 2019 Materials Today: Proceedings, vol 601-606
[3] Saiz-Arroyo C, de Saja J A, Velasco J I, and Rodriguez-Pérez M Á 2012 J Mater Sci, vol p 5680-5692
[4] Lopez-Gonzalez E, Salmazo L O, Lopez-Gil A, and Rodriguez-Perez M A 2019 Polymer Engineering & Science, vol p 791-798
[5] Heidari A and Fasihi M 2019 EXPRESS Polymer Letters, vol p 429-442
[6] Najib N, Arife T, Manan N, Bakar A, and Sipaut C 2009 Journal of Physical Science, vol p 13-25
[7] Charoeythornkhajhornchai P, Samthong C, Boonkerd K, and Somwngthanaraj A 2017 Journal of Cellular Plastics, vol p 287-303
[8] Stange J and Münstedt H 2006 Journal of cellular plastics, vol p 445-467
[9] Bledzki A K and Faruk O 2006 Journal of Cellular Plastics, vol p 63-76
[10] Reichelt N, Stadlbauer M, Folland R, Park C B, and Wang J 2003 Cell Polym, vol p 315-327
[11] Jeoung S K, Hwang Y J, Lee H W, Kwak S B, Han I-S, and Ha J U 2016 AIP Conference Proceedings, vol p 100002