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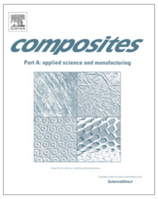

Experimental observation and modeling of fiber rotation and translation during foam injection molding of polymer composites

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ABSTRACT

We investigated the interactions between dispersed carbon fibers and growing cells in high-pressure foam injection molding experiments using a polystyrene/carbon-fiber/carbon-dioxide system with an in-situ visualization technique. We found that the fibers exhibited both translational and rotational displacements in close proximity to the growing cells. Their rotational and translational displacements were measured quantitatively using visualization snapshots. These were found to be a strong function of the cell size, the initial cell-fiber distance, and the initial fiber angle. We developed an analytical model to describe the instantaneous fiber orientation and location as a function of the corresponding cell size and the fiber's initial orientation and location. The theoretical predictions were in a good agreement with the experimental results. This showed the model's accuracy in predicting the fiber displacement. Our research provides a deeper understanding of the mechanisms through which foaming influences the percolation threshold of conductive polymer composites.

1. Introduction

Compared with their metallic counterparts, conductive polymer composites (CPCs) offer numerous advantages. These include chemical resistivity, barrier properties, low density, and fast, inexpensive manufacturing methods. The applications for this class of conductive materials have consequently increased. They are used in electromagnetic interface shielding [1,2], actuators [3,4], and fuel cell bipolar plates [5,6]. To establish the (electrical) connectivity, microsize [7–10] or nanosize [11–15] fillers can be added to a polymer matrix in a powder shape or in a fiber form. Of all the fillers, the carbon fiber (CF) application is more popular due to its low percolation threshold, its relatively high aspect ratio, and its lower processing cost compared with nanosize fillers [10,16].

The content of the fibers and the percolation level's threshold critically affect the CPCs' functionality, processability, and manufacturing costs. While a higher fiber content increases the CPCs' conductivity, it limits their processing window and increases their cost [17]. Therefore, a lower fiber loading will prove more favorable, provided that the composites' conductivity is maintained at a high degree.

The fibers' alignment and orientation plays a crucial role in determining such functional properties as CPC's electrical conductivity [18], electromagnetic interference shielding effectiveness [19], and dielectric behavior [20]. It has been also shown that a severe alignment of the fibers increases the percolation threshold and decreases the electrical conductivity [21,22]. For instance, the through-plane conductivity of injection-molded parts has been demonstrated to be several orders of magnitude lower than it is in the machine direction. This is due to the machine-direction or the fibers' in-plane orientation [17,23]. Further, in some applications, such as bipolar plates, a high level of conductivity is required in both the in-plane and the through-plane directions. Therefore, anything that could reduce the anisotropy of the CPC's functional properties by controlling the fiber orientation would be of great value.

The introduction of a cellular structure to a solid CPC has been reported to reduce the percolation threshold and to improve the composite's conductivity [17,24,25]. The dissolved blowing agent contributed to lowering the electrical percolation threshold by enhancing the fiber dispersion and distribution, and by reducing the fiber agglomeration [26–28] and breakage [25,29]. Ameli et al. discussed how foaming reduced the fiber breakage in polypropylene (PP)/stainless-steel fiber composites and resulted in a significant percolation threshold reduction [25]. Zhang et al.

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reported that a lower percolation threshold was achieved by foaming the polymethylmethacrylate/graphene composites [30]. The electrical percolation threshold of PP/multiwalled carbon nanotube (MWCNT) foams was reduced significantly by an increased volume expansion, which was attributed to the localization of fillers within the cell walls after foaming [31]. Tran et al. also showed that the electrical conductivity of a polymethylmethacrylate/MWCNT composite was increased by foaming [32]. Conductive networks were established with a lower fiber content in PP/carbon-nanofiber composite foams compared with their solid counterparts [33].

Many researchers have shown that the rearrangement and alignment modifications of fibers, platelets, and high aspect-ratio additives, occurring during cell growth in CPC foams is one of the major mechanisms responsible for enhancing the CPC's functionalities. In microcellular foamed polymer/nanoclay composites, Okamoto et al. [34], Nam et al. [35], and Yuan and Turng [36] observed that nanoclays were aligned along the curvature of cell walls as a result of the squeezed/stretched matrix around the growing cells, whereas they were randomly oriented in areas far from the cells. Motlagh et al. found that foaming disrupted the flow-induced fiber orientations and enhanced the through-plane conductivity in foam injection-molded parts [10]. Ameli et al. observed that the electrical conductivity of PP/MWCNT composites was increased by foaming, and that this was due to the rearrangement and reorientation of the MWCNTs up to the optimum void fraction, after which further cell growth deteriorated the conductivity [31]. Thompson et al. reported that while foaming enhanced the through-plane conductivity in composites containing carbon black, or carbon black mixed with CF, it had the opposite impact on samples filled only with CF [37]. Ameli et al. demonstrated that the through-plane electrical conductivity of the PP/CF composite foams was increased, and its uniformity was improved along the injection-molded sample, due to the increased fiber-to-fiber contacts [17,19]. They argued that foaming altered and increased the orientation of the fibers in the thickness direction by the biaxial stretching the matrix around the growing cells in the core regions of CPC foams. In the skin layer, the plasticizing effect of the dissolved blowing agent reduced the thickness of the solid layer and the degree of the longitudinal fiber orientation by reducing the melt viscosity and the resultant applied shear [17,19]. In foam injection molding (FIM) experiments conducted with PP/MWCNT nanocomposites, Ameli et al. showed that the cell growth caused a unique arrangement in the MWCNTs. The translational and rotational displacement of the MWCNTs was confined between the adjacent cells, and this imparted superior dielectric properties to the CPC foams compared with their solid counterparts [20].

Therefore, the orientation and re-alignment of the fiber fillers, and the disturbance which occurred during the foaming process, exerted a significant influence on the CPC parts' functional properties. Fiber orientation has been extensively studied in injection molding [38–45]. Folgar and Tucker introduced a model to predict fiber orientation distribution function, and they suggested a phenomenological relationship, including a fiber–fiber interaction coefficient [38]. Advani and Tucker later introduced a second-order tensor of orientation in order to describe fiber orientation [39]. Bay and Tucker simulated the filling and fiber orientation in the injection molding of a center-gated disk and a film-gated strip, and compared them with the experimentally measured fiber orientation [40,41]. Wang et al. then modified the equation of the orientation tensor from Ref. [39] and proposed the reduced-strain closure (RSC) model. The latter accounts for the short fibers' slower rate of orientation, compared with previous models based on Jeffery's equation [42]. Others have proposed methods to improve the computational work required for orientation predictions [43–45].

Despite the rich literature about fiber orientation prediction, less attention has been devoted to fiber displacement, that is, the rotation and/or translation, during *foam* injection molding. Thus, the effect of foaming on fiber re-orientation and its effect on interconnectivity and/or distribution require more investigation. Most analyses have been carried out on stabilized specimens, and hypotheses have been suggested based on the obtained properties. Specifically, there has been no reported observation of the real-time behavior of dispersed fibers around growing cells, although visualization has been used to study the mold-filling phenomena during the injection molding of plastics and composites [46,47]. To continue our previous work [17,19,20,25,31] an in-situ visualization technique [48] was adopted to investigate and quantitatively analyze the effect of foaming (that is, the biaxial stretching of the matrix during cell growth) on the translation and orientation of the fibers. We also proposed a geometrical model to further investigate and to predict the fiber displacements in close proximity to the growing cells. Using the experimental measurements and the model analyses, we identified the most influential factors governing the final orientation and location of the fibers in the foamed CPCs.

2. Experimental

2.1. Materials

Polystyrene (PS) MC3650 with an MFI of 13.0 g/10 min and a density of 1.04 g/cm^3 from Americas Styrenics was used as the polymer matrix. Carbon fiber with a density of 1.8 g/cm^3 in PP carrier was used as the fiber additive (Proprietary PP–CF composite, grade Carbo-Rite F261 with 10 vol% CF was provided by Lubrizol Corp., Wickliffe, Ohio, USA). The average CF diameter was about 8 μm, and their average length was about 90 μm [17,19]. Inasmuch as the addition of CF darkens the composite and prevents light transmission, the final CF content in the injection-molded PS was maintained at less than 0.1 wt%. Carbon dioxide (CO_2) from Linde Gas Canada was used as the physical blowing agent. The in-situ visualization and the SEM observations in [17,19] showed that the fibers remained rigid throughout the cell/fiber interactions.

2.2. Experimental setup

High-pressure FIM experiments were carried out using a 50-ton Arburg Allrounder 270/320C injection molding machine with a 30 mm screw diameter, equipped with MuCell technology. In high-pressure FIM, the nucleated cells at the gate and during injection dissolve back into the melt under the high cavity pressure. Therefore, foaming initiates from a homogeneous melt/gas mixture as a result of the melt shrinkage during solidification [49]. This is when the PS/CF melt has settled in the mold with no more motion and, thus, the fiber displacement is only affected by the cell growth. A rectangular mold cavity with nominal dimensions of 135 mm × 111 mm × 3.2 mm, fed by a fan gate, was used to make the samples. A prism-insert visualization mold was employed to record the foaming phenomena occurring inside the mold cavity by using a camera (CVM10 camera from JAI and a magnifying lens from Navitar) connected to the computer. More details about the visualization system are given elsewhere [48]. The amount of the CO_2 blowing agent, used in the PS melt was adjusted to such a reduced content that a very low cell density was obtained. This assured that the individual cells could be visually captured and tracked during the cell growth, without any cell-to-cell interactions. Image processing was carried out using ImageJ software, National Institutes of Health, US. To analyze the degree of fiber orientation, the fiber angle and the cell size were measured at each