

Abstract

Along the path towards developing polymeric materials that exhibit superior mechanical strength, composite materials—primarily glass fiber reinforced—have played the most significant role. In order to effectively act as reinforcement, fibers within the composite must be able to transfer stresses from the polymer matrix. The effectiveness of stress transfer depends on many factors, but the phenomena occurring at the fiber-polymer matrix interface play a key role.

In this dissertation, polymer blends of polypropylene with poly(butylene terephthalate) were prepared as a matrix basalt-fiber-reinforced, thermoplastic composites. However, these two polymeric materials are thermodynamically immiscible (depending on their molecular masses, the difference between their solubility parameters is 5–6 MPa^{1/2}). As a result, the blends were compatibilized using two copolymers (thermoplastic elastomers) that differed in chemical structure, segment arrangement, and molecular weight. Two different mechanisms of compatibilization efficiency were proposed and confirmed by differential scanning calorimetry, thermal analysis of dynamic mechanical properties, thermogravimetric analysis, mechanical testing, scanning electron microscopy, and dielectric spectroscopy. Importantly, the compatibilization mechanisms relate directly to the structural architecture (random vs. triblock copolymers) of the compatibilizers and the volume of the interphase formed between the components of the polymer blends. Crucially, apart from the chemical nature of the copolymers, their arrangement in the interfacial region (organization of blocks/segments) was even more important. The random, experimental poly(butylene terephthalate-*r*-butylene dilinoleate) copolymer (PBT-DLA) allowed for more efficient interphase coverage ("stitching" type mechanism), while the commercial poly(styrene *b*-ethylene/butylene-*b*-styrene) triblock copolymer (SEBS) penetrated significantly deeper into both phases of the blended polymers ("stapling" type mechanism). Both compatibilization strategies yielded comparable results, but the PBT DLA copolymer has the crucial advantage over commercial petrochemical compatibilizers that it can be obtained from 100% renewable monomers.

In the second stage of the work composites, with chopped basalt fibers as the reinforcing phase, were prepared from the compatibilized polymer blends. Two types of basalt fibers were used, modified with two different types of sizings to increase adhesion between the reinforcement and polymer matrix. The reference sizing is a commercial preparation optimized for engineering thermoplastic materials, such as polyesters, polyurethanes, and polyamides. The experimental sizing, on the other hand, is based on polypropylene grafted with maleic anhydride and was developed for polyolefins.

The key aspect—the presence of interfacial interactions at the fiber-polymer matrix interface—was quantified by measuring the adhesion of the fiber to the polymer material, using the pull-out method (pulling a single fiber out of the polymer matrix). The obtained results were correlated with the mechanical properties of the composites in quasi static tensile tests, three-point bending tests, and impact tests. The phenomena occurring in the interfacial region were also described and attributed to van der Waals interactions, polar interactions or hydrogen bonds formation, depending on the sizing type. The hypothesis regarding the mechanisms responsible for the behavior of the materials at the interface between (single) fiber and polymer matrix, was confirmed by scanning electron microscopy.

However, to further confirm the research hypothesis concerning thermoplastic blend composites, while taking into account the entire volume of the composite, additional research on the adhesion of fibers to the polymer matrix was required. For this purpose, for the first time for these types of materials, the non-destructive technique of broadband dielectric spectroscopy was used to explain, describe, and quantify the phenomena occurring at the fiber-polymer matrix interface. Collectively, the results confirmed that the overall adhesion of fibers to the polymer matrix is influenced by the synergistic interactions of functional groups present in the polymer matrix, as a result of their compatibilization, and on the fiber surface, due to fiber sizing.

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