

Editorial corner – a personal view

Producing green composites via polymer blending

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The rising environmental concern and the implementation of new rules and regulations are pushing researchers and industries to develop and produce eco-friendly products. One of the major growth areas is in the field of green composites, whereby natural fibers are used as reinforcement in combination with biopolymers derived from both renewable and non-renewable resources. While natural fibers are low-cost and environmentally superior alternative to synthetic fibers, biopolymers that are derived from renewable resources such as polyhydroxybutyrate (PHB), polylactide acid (PLA) and polycaprolactone (PCL), poly(butylene adipate terephthalate) (PBAT) and biobased poly(butylene succinate) (Bio-PBS) and their copolymers are promising eco-friendly alternatives for the petroleum-based polymers. However, the applications of these biopolymers on their own are considerably limited because of their high cost and imbalance properties associated with low strength, high brittleness (partly due to the poor interfacial adhesion between hydrophobic matrix and hydrophilic natural fiber), and poor processability. Various approaches have been adopted to improve the composites' strength via; optimizing filler-related properties (such as size and distribution), adding compatibilizer to improve the interfacial adhesion, and application of fiber surface modification. Usually, these techniques can effectively improve the mechanical strength and stiffness of the composites. However, the composite toughness remains the same

and far from satisfactory. So the question remains, is there any better approach that can be adopted in producing green composites with well-balanced properties (strength, stiffness, and toughness) without compromising their biodegradability?

One of the possible way is by modifying the matrix phase via incorporating of soft component. This will not only improve the matrix toughness but also compensate the brittleness attributed from the fibers. This soft component will improve the energy absorbing capability of the composites.

The introduction of soft component to a polymer could be achieved by copolymerization and/or blending techniques. In context aliphatic biopolymers, random copolymerization with soft component via biological route can improve both toughness and biodegradability by lowering stereo regularity as well as crystallinity. For biosynthetic polymer like PHB, the copolymerization via biological route using comonomer unit like hydroxyvalerate (HV) produced product with significant improvement in flexibility and toughness (<https://doi.org/10.1007/978-3-642-17997-6>). However, the copolymerization via biological route is relatively complex, while the recovery process is also expensive and require a large quantity of solvent.

Alternatively, the easier and faster technique to improve the toughness of a biopolymer is by blending with other flexible biopolymers. From the economic point of view, polymer blending is a cost-effective

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and versatile method for tailoring properties of a polymer that is less time-consuming than producing a new polymer through synthesizing routes. Currently, there are many commercially available biopolymers that can be mix-and-match to produce a polymer blend with desired properties. As an example, PHB naturally is a brittle polymer with moderate strength and low elongation at break. Their high price, low degradation temperature, and narrow processing window limit their applications in various fields. Thus blending of PHB with other biopolymers such as PCL, PBS and PBAT could provide a viable solution provided some issues such as blend component miscibility and compatibility, natural fiber distribution/dispersion, fiber-blend matrix interfacial adhesion could be resolved. This can be overcome by introducing an appropriate compatibilizer or coupling agent or via in-situ compatibilization. However, in the context of green composites, the attempt to produce a compatibilizer that can synergistically compatibilized the blend components and at the same time providing adequate adhesion to the natural fibers could be a challenging part. Compatibilizers such as glycidyl methacrylate (GMA) grafted biopolymer (*e.g.*, PBS-*g*-GMA and PLA-*g*-GMA), maleic anhydride (MA) grafted biopolymer (*e.g.*, PLA-*g*-MA, PHB-*g*-MA, and PBS-*g*-MA), and acrylic acid grafted biopolymer (PBSA-*g*-AA) have been proven effective in achieving the target.

In the context of blend matrix composites, the presence of second matrix component will make the compatibilisation attempt to be more challenging. Considering the complexity and heterogeneity nature of the polymer blend-based composites, one might expect the compatibilizing effect to be of limited success. On the contrary, the presence of cellulose and hemicellulose in natural fibers will provide possible reactive

sites for the formation of strong linkages with the functional groups in the compatibilizer, resulting in the formation of a better blend matrix and fiber interaction. Compatibilizers such as PLA-*g*-MA, PBS-*g*-MA and PCL-*g*-MA consist of bifunctional groups that can react with all component in the blend composites. Its polymer chain can physically entangle and cohere with the host polymer chains, whereas its anhydride functional group could interact with the hydroxyl groups of natural fiber (<https://doi.org/10.3390/polym10040382>). In addition, the secondary interaction may also be formed via a hydrogen bonding between the oxygen atom of compatibiliser anhydride group and the natural fiber hydrogen atom or vice versa (<https://doi.org/10.1080/09276440.2018.1439622>). Besides, in the case of biopolymer blend matrix the anhydride functional group of the compatibilizer could also interact with the hydroxyl end groups of the guest polymer via esterification reaction during the melt blending process (<https://doi.org/10.1002/vnl.21828>). Thus remarkable enhancement in stiffness, strength, and toughness could be expected, which may not be observed in the blends in the absence of natural fibers. Further enhancement could perhaps be achieved via dual compatibilization or toughening-compatibilization synergism mechanism. The blend components are first reactively linked with peroxides or isocyanates coupling agents or via the introduction of block graft copolymers containing blocks that miscible with both blend components. Subsequently, they can be compatibilized with anhydride- or epoxide-based compatibilizers to improve the blend matrix and natural fibers' interfacial adhesion. So there are still various ways to be explored in our endeavor to produce cost-effective, eco-friendly, and well-balanced properties green composites via polymer blending in the future.