



# Compatibilization of multicomponent composites through a transitioning phase: Interfacial tensions considerations

J. Justin Koh<sup>a,b</sup>, Xiwen Zhang<sup>b</sup>, Junhua Kong<sup>c</sup>, Chaobin He<sup>a,c,\*</sup>

<sup>a</sup> Department of Materials Science and Engineering, National University of Singapore, 9 Engineering Drive 1, 117576, Singapore

<sup>b</sup> Singapore Institute of Manufacturing Technology, Agency for Science, Technology and Research (A\*STAR), 73 Nanyang Drive, 637662, Singapore

<sup>c</sup> Institute of Materials Research and Engineering, Agency for Science, Technology and Research (A\*STAR), 2 Fusionopolis Way, Innovis #08-03, 138634, Singapore

## ARTICLE INFO

### Keywords:

Compatibilization  
Interfacial energy  
Polymer composites  
Poly(lactic acid)  
Starch  
Toughening

## ABSTRACT

A novel interfacial compatibilization technique for incompatible polymer blends or composites is proposed, in which a transitioning layer was introduced between the matrix and the dispersed phase of the otherwise incompatible components. The transitioning phase should have good interactions with both the components, resulting in lower interfacial energy between the phases. Theoretically, it is hypothesized that if the sum of the interfacial tension between the transitioning phase and both the components of the composite is smaller than the interfacial tension between the two components, the encapsulation of the dispersed phase by the transitioning phase is spontaneous, which will lead to better interphase interfacial interactions. Since this compatibilizing technique relies purely on judicious selection of a polymer with suitable surface energy as the transitioning layer, no tedious chemical synthetic processes are required. To illustrate the proposed technique, incompatible Poly(lactic acid)/Thermoplastic Starch (PLA/TPS) blend is compatibilized with Poly(butylene succinate) (PBS) as the transitioning layer in this paper. With PBS encapsulating the dispersed TPS phase, PLA/PBS/TPS 60/10/30 wt% demonstrate a better mechanical synergy, with significant improvement in strength, ductility and toughness as compared to PLA/TPS 70/30 wt%. This technique can also be applied to design other multicomponent blends or composites.

## 1. Introduction

The concept of polymer blends provides a wide range of opportunities and possibilities to create new polymeric materials that can incorporate the strength or mitigate the weakness of each individual components. One of the earliest successfully commercialized polymer blend is poly(phenylene oxide)/poly(styrene) system where the two miscible components produce a blend that has a balanced thermal, mechanical and chemical properties [1]. Subsequently, more polymer blends with properties suitable for specific applications are commercialized [2]. However, many of the polymers are thermodynamically immiscible hence phase separation occurs between the different components. Such systems can be treated as composites where their interphase interface is crucial. Incompatible blends or composites with poor interphase interfacial adhesion often results in poor mechanical properties. Therefore, interfacial compatibility between the different components is a key aspect in engineering polymer blends.

Throughout the years, many compatibilizing strategies have been discovered and utilized to improve the compatibility of composites. Some common technique include chemically crosslinking the different

phases [3–6], adding of copolymer filler with different segments compatible to the different separate phases [7,8] and surface modification of the one component (usually the filler or minority component) to make it compatible with the other [9–14]. Unfortunately, while many of these strategies are effective, they involve substantial synthetic processes that diminishes the feasibility of commercialization.

This paper provides a novel and systematic approach to compatibilize binary incompatible blends/composites by manipulating the interphase interactions by introducing a transitioning phase. The theory and basis of the technique is described in detail in the Theory Section. To the best of the author's knowledge, this is the first deliberate attempt to compatibilize blends using such strategy, even though there may be multicomponent blends that exhibit such mechanism by chances [15,16].

The strategy is further supported by using the PLA/Starch as an example. Both PLA and Starch are bio-based, biodegradable as well as biocompatible, hence are suitable for packaging, disposable, and biomedical applications. Unfortunately, incompatibility between PLA and Starch, leads to poor interfacial adhesion, defects and inadequate stress transfer between the two phases, which often results in a weaker and

\* Corresponding author. Department of Materials Science and Engineering, National University of Singapore, 9 Engineering Drive 1, 117576, Singapore.  
E-mail address: [msehcnus.edu.sg](mailto:msehcnus.edu.sg) (C. He).

more brittle PLA/Starch blend as compared to pure PLA. Throughout the years, many toughening strategies have been successfully applied to PLA/Starch blend [17]. In this paper, three biodegradable polyesters with different surface energy and interaction characteristics, namely Poly(butylene succinate) (PBS), Poly( $\epsilon$ -caprolactone) (PCL) and Poly(hydroxybutyrate-co-hydroxyvalerate) (PHBV), were investigated as potential transitioning phase between PLA and Thermoplastic Starch (TPS) to demonstrate the proposed interphase compatibilizing strategy.

## 2. Theory Section

The essence of the technique is to introduce a transitioning layer between the two otherwise incompatible components of the composite. This transitioning phase should have strong interactions with the two components of the composites, such that there is better interfacial adhesion and mechanical synergy between the phases, facilitating stress transfer. Also, the strong interaction should result in low interfacial tension between the transitioning phase and the two components of the composite, such that a thermodynamically spontaneous mechanism will take place for the transitioning phase to encapsulate the dispersed phase within the matrix. In addition, the lower interfacial tensions between the different phases aid in stabilizing the dispersion, by lowering the driving force for them to agglomerate.

From the thermodynamic perspective, the Gibbs Free Energy of a three component immiscible system, is the sum of the free energy of the individual components and the interfacial energy of the component pairs, as described by the following equation:

$$G_{123} = V_1 G_1 + V_2 G_2 + V_3 G_3 + A_{12} \gamma_{12} + A_{13} \gamma_{13} + A_{23} \gamma_{23} \quad (1)$$

where 1 is the major component that forms the matrix, 2 and 3 being the minor components dispersed within 1.  $V_i$  and  $G_i$  represents the volume and free energy per unit volume of the component.  $A_{ij}$  and  $\gamma_{ij}$  represents the interface area and the interfacial tension between the component pair  $i$  and  $j$ . If component 3 has a low interfacial energy with component 1 and 2, such that,

$$\gamma_{12} > \gamma_{13} + \gamma_{23} \quad (2)$$

then there will be a strong driving force to eliminate the high  $A_{12} \gamma_{12}$  component in order to lower the free energy of the system. This will lead to a spontaneous mechanism for component 3 to encapsulate component 2, forming a transitioning phase between component 1 and 2 (Scheme 1). This mechanism has also been described by the Harkin's spreading theory [18]. The following is the Hobb's modified Harkins' equation [19]:

$$\lambda_{32} = \gamma_{12} - \gamma_{13} - \gamma_{23} \quad (3)$$

where  $\lambda_{32}$  is the spreading coefficient for situation when component 2 is

the core being encapsulated by component 3.  $\lambda_{32}$  must be positive for this situation to be true.

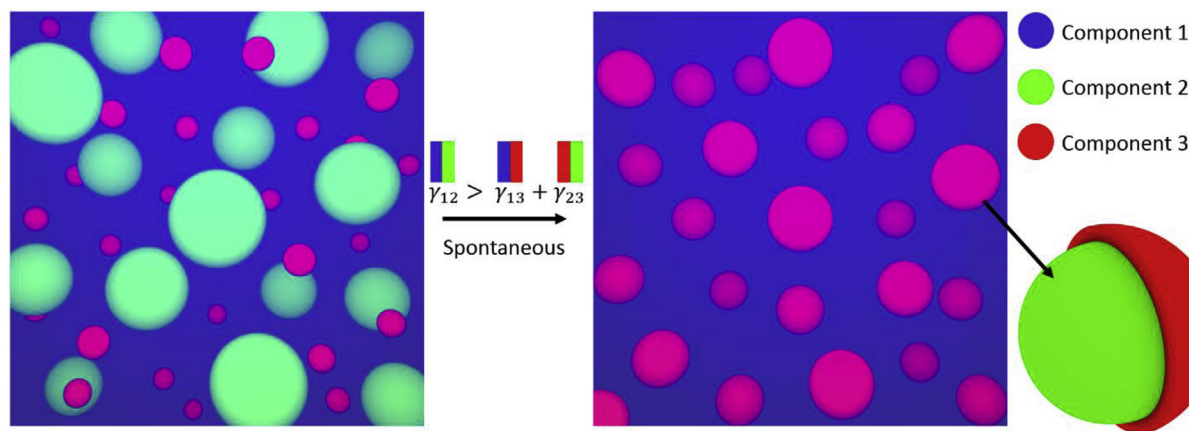
There are also concerns regarding the change in interfacial area (especially  $A_{13}$ ) after the encapsulation, resulting in inaccurate prediction of the encapsulation. This led Guo et al. [20] to develop another model that takes the effect of interfacial area into consideration for the thermodynamic equilibrium morphology. Despite the tedious effort, Guo et al. concluded that interfacial tension played the major role and that interfacial area is less significant. More importantly, Hobbs modified Harkin's equation has successfully predicted most ternary composite system for the encapsulation of the dispersed phase, including Guo's PMMA/PS/HDPE system.

Besides, there may also concerns regarding the competition from kinetics effects of melt blending, especially when the component 3 has a much higher viscosity than the dispersed phase. Although there are reports showing that the much higher viscosity of component 3 are not able to overcome the thermodynamic driving force as described by Harkins' spreading coefficient [21]. Reverse encapsulation of the component 3 by the dispersed phase have been reported [22] when the viscosity of the 3<sup>rd</sup> component is much higher than the dispersed phase (liquid crystalline polymers) and the driving force (spreading coefficient) is low. Since the illustration used in this study is TPS, which is a quasi-solid at the blending temperature, such rheological issues are not a concern.

Nonetheless, the key is to identify a 3<sup>rd</sup> component that has strong interaction, hence low interfacial tension, with both the matrix and the filler material of the composite. According the Fowkes [23], the surface tension (interfacial tension with air) of a substance comprise of several additive components, such as dispersion, inductive dipole, permanent dipole-dipole, hydrogen bonding interactions and etc. Thereafter, there are several theories established to simplify the estimation of surface tensions, for example, the Van Oss-Chanhury-Good (VCG) [24] and Owens-Wendt (OW) [25] methods. In this paper, OW the method is used due to its wide acceptance as well as its reproducibility. OW combines some of the interaction terms to resolve surface tension of a substance into two components: dispersive and polar interactions. The polar component can be taken to be the combination of inductive dipole, permanent dipole-dipole and hydrogen bonding interactions. This method is suitable for simple, non-aromatic and non-ionic polymeric materials.

To obtain surface tension experimentally, Young's equation relates the surface tension of a solid to the contact angle of a liquid, surface tension of the liquid and the interfacial tension between the liquid and the solid. The surface tension of probe liquids are available in literatures; in this paper Ström's data were used [26].

The two equations that are commonly used to estimate the



**Scheme 1.** Schematic illustration for spontaneous formation of a transitioning phase between dispersed and matrix phase due to lower interfacial energy resulting from strong supramolecular interactions.

Download English Version:

<https://daneshyari.com/en/article/7214193>

Download Persian Version:

<https://daneshyari.com/article/7214193>

[Daneshyari.com](https://daneshyari.com)