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Viscometric Investigation of Compatibilizer Effect on Polyvinyl chloride/Polystyrene Blends

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Abstract: In this study, polyvinyl chloride (PVC) and polystyrene (PS) polymers, which do not form a compatible blend with each other in certain concentration ranges and are widely used on an industrial scale, were used. The concentration range in which these two polymers do not form a compatible blend were determined viscometrically, and maleic anhydride-styrene (MAS) copolymer was added as a compatibilizing agent in this concentration range. The concentration range in which the polymer pair did not form a compatible blend was determined as 1.5 and 2.0 g dL^{-1}. In this concentration range, 5.0 and 10.0 % MAS copolymer was added to the polymer couple and its compatibilizing effect was investigated. It was determined by viscometric method that the MAS copolymer was a good compatibilizer in the range where the polymer pair did not form a compatible blend.

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Polivinilklorür/Polistiren Karışımlarına Uyumlaştırıcı Etkisinin Viskozimetrik İncelenmesi

Anahtar Kelimeler
Maleik anhidrit,
Polistiren,
Polivinilklorür,
Uyumlaştırma

Öz: Bu çalışmada belirli derişim aralıklarında birbirleriyle uyumlu karışım oluşturmayan ve endüstriyel ölçekte yaygın kullanıma sahip polivinilklorür (PVC) ve polistiren (PS) polimerleri kullanıldı. Bu iki polimerin uyumlu karışım oluşturmadığı derişim aralığı viskozimetrik yolla belirlenerek bu derişim aralığında maleik anhidrit-stiren (MAS) ko-polimeri uyumlaştırıcı ajan olarak eklendi. Polimer çiftinin uyumlu karışım oluşturmadığı derişim aralığı 1.5 ve 2.0 g dL^{-1} olarak saptandı. Bu derişim aralığında polimer çiftine % 5.0 ve 10.0 oranlarında MAS ko-polimeri eklenerek uyumlaştırıcı etkisi araştırıldı. Eklenen ko-polimerin PVC/PS polimer çifti için iyi bir uyumlaştırıcı olduğu belirlendi.

1. INTRODUCTION

A polymer blend is analogous to metal alloys, in which at least two polymers are combined to create a new material with different physical properties. Mostly, the polymer blends are immiscible in nature while some are miscible. In the immiscible polymer blends, the polymers exist in different phases, whereas single phases are observed in miscible or homogeneous polymer blends due to having the same chemical structure [1]. Polymer blends can be classified according to the physical properties of their constituents as plastic, thermoplastic, elastomer, and so on. However, since the ultimate properties of a polymer blend will depend on the final morphology, miscibility and phase behavior of polymer blends needs to be taken into account. In the other words, polymer blending can be practicable only when the selected polymers are miscible or can be made miscible by a selected compatibilizer [2,3]. Blends are usually made in two ways. The first way is to dissolve the two polymers in the same solvent and then evaporate the solvent. When the solvent has completely evaporated; If the two polymers are miscible, a homogeneous blend is observed, if not phase separations are observed. The second way is to heat the two polymers together above the glass transition temperatures of both polymers. This is often done in extruders [4].

Compatibilizers are utilized in polymer chemistry to improve the mechanical properties of multicomponent polymer blends. The process of compatibilization is meant to reduce polymer particle interfacial tension, facilitate chain dispersion, stabilize morphology against severe melt processing conditions, and enhance adhesion between two phases [5]. There are compatibilizers with

different blending mechanisms. Examples of reactive compatibilizers: acrylic functions grafted on polyolefin, polyethylene and PP, allow compatibilization with PAs, EVOH, polybutylene terephthalate (PBT), PET. Acrylic functions are often maleic anhydride, glycidyl methacrylate. Such compatibilizers are marketed, for example, as Amplify GR-MA (Dow), Elvaloy PTW and Fusabond (DuPont). Examples of nonreactive compatibilizers: Ethylene–ethylacrylate (EEA) copolymers for PP/PA recycling, ethylene–butylacrylate (EBA), and ethylene methacrylate (EMA) copolymer for compatibilization of PP, PE, PBT, PA, ABS, PC. PMMA or polystyrene grafted on PP (Interloy) can compatibilize polypropylene with PMMA, SAN, ASA, ABS, PVC, PC, PPE. Acrylic–imide copolymers (Paraloid) can compatibilize PPE/PA, PC/PE. Styrenic block copolymers can compatibilize PP/HDPE, PPE/PA, olefins and styrenics SB, PS, ABS [6].

The preparation of polymer blends and the use of these blends in different application areas are closely related to the miscibility of polymers. In this way, blends were formed by using miscible polymers and used for different purposes [7-11]. In order to form compatible polymer blends with each other, there must first be secondary interactions between them, such as dipole-dipole interactions or hydrogen bonds. However, most polymers are not miscible because they cannot interact with each other in this way. For polymers that do not mix with each other to become miscible, the interactions mentioned here must occur [12].

Interactions and interfacial adhesion between polymers with dissimilar chemical structures cannot be effective enough to form blends. Therefore, such polymers must be compatibilized in order to form miscible blends [13,14]. Compatibilization is the addition of a small amount of an interfacing agent to immiscible polymer blends. These agents are called compatibilizers. A compatibilizer is a polymer that, when added to an immiscible polymer pair, increases the degree of compatibility between the polymer pair. Such compatibilizing polymer may be a homopolymer as well as usually a co- or terpolymer. The compatibilizer reduces the interface tension by strengthening the adhesion between the polymers in the blend [15]. For PS and PVC; although both are vinylic polymers, interfacial adhesions of benzene and chlorine side groups are high. Therefore, they do not form compatible blends and a compatibilizer is required.

Polystyrene and PVC polymers do not form compatible blends [16]. The thermal properties of polystyrene are relatively good. It is also a well-known commercial polymer that is radiation resistant. Polystyrene is produced and used in a wide variety of forms. The benzene ring in its chain provides flame resistance and solvent resistance. The polystyrene used in the industry is a highly tactical polystyrene and has an amorphous structure. Its UV radiation resistance and insolubility in many solvents cause environmental problems [17]. PVC polymer is one of the most important industrial polymers that is widely used commercially and preferred in many materials. Polyvinylchloride polymer also has a large

commercial use like polystyrene. It is a thermoformable, mostly amorphous polymer with a linear chain [3].

An additional method of polymer modification is through blending; a procedure that involves the mixing of the main polymer in interest with other components to provide a composite mixture or a blend is used, with improved properties [18]. Blending of polymers is becoming increasingly important in different applications to enhance properties, improve processing, or decrease the cost of production [19]. Blending may also involve the addition of a different class of materials to polymers, such as compatibilizers [20-22]. Polymer blending is also achieved through the mixing of two or more different polymers in the liquid phase [23]. Mixing polymers of different classes may result in miscible blends [24]. Polymer blend miscibility is assessed by viscometry, which also provides information about the interactions of the polymers with each other through blending [25,26].

Although there are different techniques to determine whether polymers are miscible or not, the viscometric method, which can provide important information about polymer-polymer miscibility, still remains a simple and inexpensive method. In this respect, it will continue to be a preferred method in laboratories in examining the behavior of polymer blends. It seems that the Krigbaum-Wall equation will continue to be the most widely used equation for those who are looking for an inexpensive and easy method to evaluate the results obtained by the viscometric method of polymer blends.

Non-miscible polymer pairs were compatibilized using various type of compatibilizers and were studied. There are studies showing that the mechanical and thermal properties of polymer blends compatibilized using compatibilizers are improved [27-29]. In most studies, co- or terpolymers based on maleic anhydride have been used as compatibilizing agents [30-34, 17]. A maleic anhydride based compatibilizer is able to reduce the interfacial tension. As the result of this phenomenon, the interfacial between both phases is strong and the direct effect correspond to the improvement of compatibility [35]. In this research article authors used maleic anhydride-styrene (MAS) for compatibilization of PVC/PS blends. MAS is a reactive synthetic random copolymer made up of styrene and maleic anhydride. Copolymers with maleic anhydride make the blend system miscible by establishing secondary chemical interactions between immiscible polymer pairs. Because of its high reactivity and efficiency, it is widely used as a compatibilizer with most of the blend systems [36].

Many studies have been carried out for blending of PS and PVC [16, 37,38]. In these studies, different copolymers containing maleic anhydride were used and remarkable results were obtained.

In this study, solutions containing 1:1 by weight of polyvinyl chloride (PVC) and polystyrene (PS) with concentrations of 0.5, 1.0, 1.5 and 2.0 g.dL^{-1} were prepared. In the solution medium, it was determined by viscometric method at which concentrations a compatible

blend was formed or not. The concentration range in which this polymer pair did not form a compatible blend was found to be 1.5 and 2.0 g.dL^{-1}. Then, the compatibilizing effect was investigated by adding 5 and 10% by mass of MAS copolymer to the solution containing an equal weight of polymer pairs in this concentration range. As a result, it was found that MAS copolymer can be used as a compatibilizer for PVC and PS blends at 1.5 and 2.0 g.dL^{-1} concentrations.

2. MATERIAL AND METHOD

2.1. Chemicals

Polyvinyl cloride (PVC): The PVC used in this study is a polymer produced by Petkim(Turkey) under the name Petvinyl and with code number P38/74.

Polystyrene (PS): PS used in this study is a polymer produced by Petkim(Turkey) under the name Petren and with code number K-560.

Tetrahydrofuran (THF): The THF used in this study is of analytical grade and is Lab-Scan branded and produced by Labscan Ltd.

Maleic anhydride-Styrene (MAS) copolymer: This copolymer was synthesized in our laboratory.

Styrene: Analytical grade, a Sigma-Aldrich product.

Benzene: Analytical grade, a Sigma-Aldrich product.

Azobisisobutyronitrile (AIBN): % 99 purity, a Merck product.

1-Propanol: Analytical grade, a Merck product.

2.2. Experimental

In this study the miscibility and immiscible concentration range of the polymers were determined. Since PS, PVC and MAS polymers are all soluble in THF, polymer solutions were prepared using THF.

2.2.1. Synthesis of MAS copolymer

With a molar ratio of 1:1; 10 mL of styrene and 9.8 grams of maleic anhydride were taken into a reaction flask. Monomers were dissolved by adding 50 mL of benzene. 0.02 g of AIBN initiator was added to this solution and reacted for 90 minutes at 65 °C. The copolymer was precipitated by adding propyl alcohol to the resulting viscous solution. The copolymer dried at room temperature.

2.2.2. Viscometric measurements

In this study the flow times of pure polymer and polymer solutions were measured to calculate the miscibility parameter. Relative (ηr), and specific (ηsp) viscosities of each solution were calculated. These measurements were made with the Cannon-Fenske type viscometer.

All polymer solutions were prepared in 25 mL volumes, in concentrations and ratios where they were immiscible.The prepared solutions were poured into the viscometer and the flow times were recorded.

The interaction parameters of a polymer blend is defined by Krigbaum and Wall by Equation (Eq.1 and 2) [39].

$$b^{id}{}_{blend} = b_{AA}W^2{}_A + b_{BB}W^2{}_B + 2b^{id}{}_{AB}W_AW_B \qquad (1)$$

$$b^{id}{}_{AB} = \sqrt{bAA \; x \; bBB} \qquad (2)$$

where : b_{AA}, b_{BB}, b_{AB} are the interaction parameters of first polymer, second polymer and polymer blend respectively. W represents the weight fraction of the polymers.

The interaction parameters (b) of polymers can be determined using dilute solutions, and this parameter can be used as a criterion for miscibility. This method is based on comparing ideal (b^{id}) and experimental interaction (b^{exp}) parameters. It is possible to calculate these two values from the reduced viscosity value ($[\eta]$) – concentration (C) graph [40].

$$\frac{\eta sp}{c} = [\eta] + bC \qquad (3)$$

$$\Delta b_{blend} = b^{exp}{}_{blend} - b^{id}{}_{blend} \qquad (4)$$

When Δb is positive;
Polymers are said to be miscible. However, if it is negative, it is assumed that the polymers are not compatible.

From the viscometric flow times of the polymers and blends all the parameters mentioned above are calculated and given in the Tables 1, 2, 3 and 4.

3. RESULTS AND DISCUSSION

Table 1. Calculated $\eta_{sp/c}$ values of polymers

C /g dL^{-1}	$\eta_{sp/c}$ values	
	PVC	PS
0.5	1.314	1.016
1.0	1.449	0.986
1.5	2.090	1.441
2.0	2.594	1.527

Table 2. Calculated ideal interaction parameters (b^{id}) of PVC, PS and PVC/PS

C / g dL^{-1}	$b_{11 \,(PVC)}$	$b_{22 \,(PS)}$	$b^{id}{}_{(PVC/PS)}$
0.5	1.360	0.250	0.583
1.0	0.817	0.094	0.277
1.5	0.970	0.330	0.595
2.0	0.980	0.319	0.557

Table 3. Calculated experimental interaction parameters (b^{exp}) of PVC/PS blends

C / g.dL^{-1}	b^{exp}
0.5	0.861
1.0	0.528
1.5	0.347
2.0	0.281

Table 4. Calculated miscibility criterion (Δb) of PVC/PS blends

C /g.dL^{-1}	Δ_b
0.5	0.278
1.0	0.251
1.5	-0.248
2.0	-0.276

Table 5. Flow times of solutions added MAS copolymer

C /g dL^{-1}	Flow times / s		
	% 0 MAS	% 5 MAS	% 10 MAS
1.5	1200	1330	1391
2.0	1621	1760	1864

When Table 4, which was created with the data in Tables 1, 2 and 3, is examined, it can be seen that PVC and PS mix with each other at relatively low concentrations. However, as the concentration increases, they begin to not mix (due to negative Δb). In order to better monitor this relationship, the calculated miscibility parameter (Δb) values for PVC/PVS blends are plotted against concentration and given in Figure 1. It is understood from Figure 1 that the PS/PVC blend does not miscible at 1.5 and 2.0 g.dL^{-1} concentrations. By taking note of these two experimentally determined concentrations, 5 and 10 percent maleic anhydride-styrene (MAS) copolymer was added to the solutions containing 1:1 PS/PVC couple by weight at these two immiscible concentrations. The flow times of these blends with compatibilizer added, measured by viscometer, are shown in Table 5.

By using the flow times in Table 5, the relative, specific and η_{sp}/c values of the solution which do not have compatibilizer and the solutions containing 5 and 10 percent MAS compatibilizer were calculated and given in Table 6. In order to determine whether the added MAS copolymer is a good compatibilizer, it is necessary to look at the interaction parameter (b^{exp} and b^{exp}) calculated via the data in Table 6 with equations 1, 2 and 3. Relevant calculations have been made and given in Table 7. When Table 7 is examined, it is seen that the b^{exp} values of the polymer blends increase as the MAS copolymer is added. However, b values alone are not sufficient as proof that polymers become miscible. For this reason, the miscibility criterion (Δb) should be checked at the relevant concentrations. For this, (Δb) values of the blends were calculated from equation 4 and given in Table 8. It can be read from Table 8 that Δb values of PVC/PS polymer couple increase as MAS copolymer compatibilizer is added. It can also be read from Table 8 that the miscibility increases with the addition of the compatibilizer at a relatively high value. In particular at the rate of 10 percent, the added compatibilizer made the immiscible PVC/PS couple miscible. Figure 2 shows the SEM images of PVC/PS and PVC/PS/MAS containing % 10 MAS blends. The SEM micrographs show clearly that the PVC/PS blend has a separate two-phase morphology.

Table 6. Different viscosity values of solutions added MAS copolymer

C /g dL^{-1}	Viscosity values								
	% 0 MAS			% 5 MAS			% 10 MAS		
	η_b	η_{sp}	$\eta_{sp/c}$	η_b	η_{sp}	$\eta_{sp/c}$	η_b	η_{sp}	$\eta_{sp/c}$
1.5	3.243	2.243	1.495	3.594	2.594	1.730	3.639	2.639	1.760
2.0	4.381	3.381	1.691	4.757	3.757	1.878	5.038	4.038	2.019

Table 7. b values of MAS added polymer couple solutions

C /g dL^{-1}	b^{exp} values			b^{id} values
	% 0 MAS	% 5 MAS	% 10 MAS	(from Table.2)
1.5	0.347	0.620	0.660	0.595
2.0	0.281	0.469	0.610	0.557

Table 8. Δb values of immiscible blends containing MAS

C /g dL^{-1}	Δb values		
	% 0 MAS (from Table 4)	% 5 MAS	% 10 MAS
1.5	-0.248	0.025	0.065
2.0	-0.276	-0.088	0.053

In particular, the PVC-PS blend reveals the polarity differences, and high interfacial tension induces the heterogeneity in the matrix. The appearance of voids and sporadic chain sizes and shapes is due to the inconsistency between the two incompatible polymers. In contrast, the (PVC/PS/MAS) blend has a single-phase morphology. Addition of MAS copolymer results in thermodynamic compatibility through addition of compatibilizer. The smooth surface with fewer fissures and better morphological characteristics is evidence of moved forward attachment between the two incompatible

polymers. This situation observed in Figure 2 showed that the MAS copolymer is a good compatibilizer for these two commercial polymers.
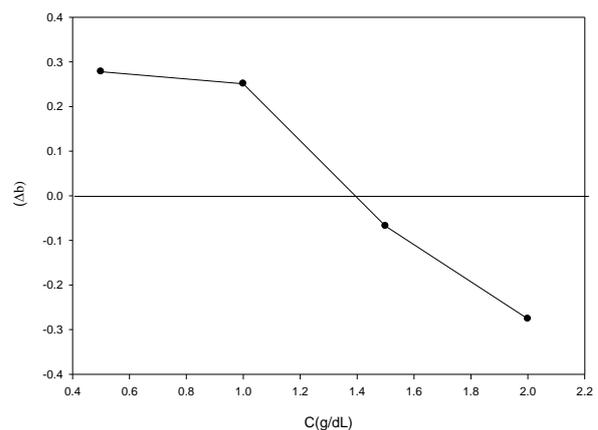
**Figure 1.** Change of miscibility parameter (Δb_{mix}) for PVC/PS blend
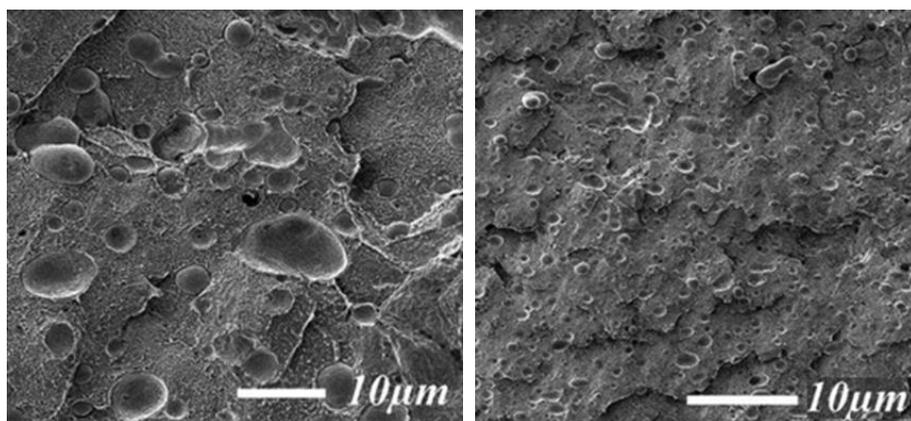
**Figure 2.** SEM Images of the polymer blends: (left) PVC/PS, (right) PVC/PS/MAS
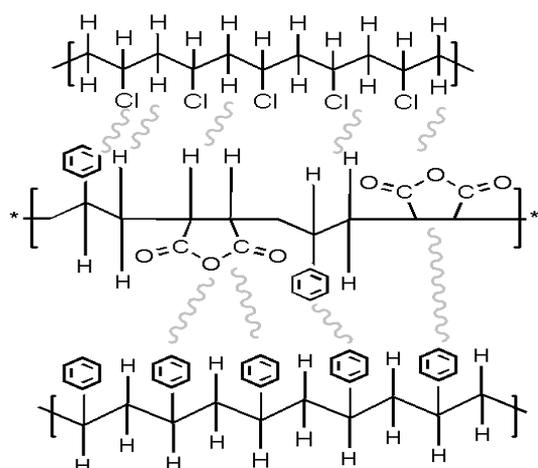
Figure 3. Possible secondary interactions between the polymers and compatibilizer

The factor that makes the two polymers miscible is largely the secondary interactions between the compatibilizer and the components of the blend. Maleic anhydride is an electron donor monomer. The anhydride groups in the compatibilizer make dipole-dipole interactions with the electron donor benzene rings in the polystyrene chain. They also interact with the hydrogen atoms in the PVC backbone due to the electron withdrawal of the -Cl group in the PVC chain. The benzene rings in the copolymer will also interact with the polystyrene chain due to the similarity in chemical structure and will strengthen the bridge established by the copolymer between the two polymers. Such possible secondary interactions are given in Figure 3.

As a result, it can be said that the MAS copolymer is a good compatibilizing agent like other similar copolymers [41-43] in preparing compatible blends of various commercial polymers.

This research have shown that improved compatibility and morphological properties of PS/PVC blends compatibilized by MAS copolymer. MAS copolymer has formed a superior adhesion between the PVC and the PS phases. Due to this effective compatibilization, this copolymer, which we synthesized in our laboratory, can be selected as a compatibilizer in applications where these two important commercial polymers are used.

4. CONCLUSION

As mentioned earlier in this article, compatibilized blending of polymers is one of the only implies of getting an assortment of physical and chemical properties from the constituent polymers. The pick-up in superior properties of polymers depends on their degree of compatibility or miscibility at an atomic degree. Moreover, blending of polymers is a productive approach to the accomplishment of modern combinations of wanted properties without having to synthesize new polymeric materials. This study, in which two very important commercial polymers such as PS and PVC are made into a compatible blend, will contribute to the literature in line with these purposes.

Further studies involving thermal, mechanical and durability testing of MAS-compatibilized PS/PVC blends can be performed to identify potential applications in which these blends could be used.

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