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Preparation of composite dispersed phase morphologies in incompatible and compatible blends during melt processing

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Most processing/morphology studies of multi-phase polymer blends have been concerned with controlling the size and shape of the dispersed phase. The dispersed phase is generally a pure homo- or copolymer (apart from low levels of interfacial modifier). This paper describes the preparation during melt processing of a complex polymer blend morphology known as a composite dispersed phase system. Microscopically this structure can be seen to be composed of three parts: two distinct phases with sub-inclusions within one of the phases. This system is a type of blend within a blend. Various microscopic techniques are used to show that a composite multi-phase morphology can be prepared in an incompatible polypropylene/polycarbonate (PP/PC) blend as well as in a compatible polyethylene copolymer/polyamide blend. This structure has been generated at two compositions for polypropylene/polycarbonate through melt blending. At 50% volume fraction (near the region of dual-phase continuity), simultaneous addition of components results in co-continuous polypropylene and polycarbonate phases with the latter containing small PP spherical sub-inclusions. At 25% PC (volume fraction) the generation of a composite dispersed phase in a polypropylene matrix is obtained by imposing phase inversion followed by controlling the mixing time. The morphology in this case consists of a polypropylene matrix, a polycarbonate dispersed phase and spherical polypropylene sub-inclusions within the dispersed polycarbonate. Partial stabilization of the composite morphology in incompatible blends with mixing time can be obtained through control of the viscosity of the dispersed phase. Polyethylene copolymer/polyamide blends have also been prepared by the phase inversion process and show that strong interfacial interactions between the polyamide sub-inclusions and polyethylene copolymer dispersed phase material results in complete stabilization of the composite dispersed phase morphology with very high retention of sub-inclusions persisting even after long mixing times.

1. Introduction

Polypropylene/polycarbonate blends are highly incompatible and have been chosen in our laboratory as a model system to investigate the effect of various processing factors on morphology [1–4]. Favis and Chalifoux [1, 2] studied the effect of composition of the blends and viscosity ratio of the components on the size of the dispersed phase. The results obtained indicate that the size of the minor phase is strongly affected by these two factors. Most peculiar was the observation of an unexpected composite droplet type morphology at a PC volume concentration of 50%. The morphological structure was basically described as being composed of a matrix and a minor phase with small droplets dispersed in the latter. This type of structure is similar to that of some rubber based blends such as high impact polystyrene (HIPS) albeit the route by which it was obtained is completely different. In the case of HIPS, an elastomeric polymer such as polybutadiene is dissolved at a concentration of 5–10% in styrene to form a homogeneous solution which is subsequently polymerized. If careful experimental conditions are met, the well known salami-like HIPS structure is produced [5]. The material is composed of a polystyrene matrix and rubber minor phase. The latter contains small polystyrene sub-inclusions [6]. It is believed that this particular structure gives rise to a marked improvement in the impact strength of the rubber modified polystyrene [7]. The inclusion of very small particles of the same chemical nature as the matrix inside the dispersed phase also tends to raise the apparent volume fraction of the

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TABLE I Characteristics of polypropylene, polycarbonates, nylon and Surlyn materials

Sample	Melt[a] Index ASTM (g 10 min^{-1})	Density[a] at 250 °C (g cm^{-3})	Torque at 5 min/mixing at 250 °C (N m)	η' 100 rad s^{-1} (Pa s × 10^{-2})	G' 100 rad s^{-1} (Pa × 10^{-4})	M_n[a] (g mol^{-1})	M_w[a] (g mol^{-1})
PP-1	12	0.75	3.9	1.6	1.3	31 300	200 000
PC-1	18	1.08	26.9	17.9	8.1	12 300	26 500
PC-2	9	1.08	37.6	21.9	–	13 200	30 300
PC-3	6	1.08	43.5	26.4	15.7	–	30 000–32 000[b]
Nylon	–	0.96	5.7	2.44	0.434	–	–
Surlyn	1	0.74	14.3	5.0	4.6	–	–

[a] Obtained from the supplier.

[b] M_{visc}.

latter with respect to that of the matrix. By this process one can achieve the desired volume fraction with less dispersed phase which, in the case of HIPS is the more expensive material.

A similar morphological structure has also been observed in thin films of solution cast polymer blends [8, 9]. Groeninckx *et al.* [8] used transmission electron microscopy (TEM) to study the morphology of solution cast polycarbonate–polystyrene blends. For the 25/75% (wt) PC/PS concentration they observed that small PS particles are occluded in the dispersed PC phase. Trent *et al.* [9] made similar observations with a 10/90% (wt) polystyrene/polymethylmethacrylate films cast from toluene. The films were stained by a 0.5% (wt) solution of ruthenium tetroxide in distilled deionized water to enhance the phase contrast between the phases during observation by TEM. The staining did not affect PMMA while PS appeared as spherical dark droplets. The TEM observations revealed, however, that the dark regularly shaped PS droplets contained irregularly shaped sub-micron PMMA domains.

The composite dispersed phase structure is also common in some biological and physical systems as has been reviewed by Eastmond and Phillips [10]. The authors show that the internal structure of white fat cells taken from a rat mesentery and the structure of an assembly of soap bubbles have strikingly similar morphological features as HIPS and concluded that interfacial tension is responsible for their presence. Dokic and Sherman [11] studied the effect of temperature variation on phase inversion of concentrated oil/water emulsions (70/30 wt %) stabilized by various emulsifiers. They show that, when the temperature induced phase inversion was successful, the development of a multi-phase water/oil/water emulsion structure in which water droplets are occluded within the oil drops precedes phase inversion.

Although it has been reported for some polymer pairs, the presence of a composite dispersed phase morphology has not been significantly discussed in polymer blend studies, apart from high-impact polystyrene. It has been generally described as a curious observation. Little work has been done to examine the potential of producing this morphology during melt processing and few studies have been carried out to evaluate the parameters which could influence composite multi-phase formation.

The work done by Van Oene [12] relates sub-inclusion formation for polystyrene/polymethylmethacrylate blends to the minimum surface free energy which depends on interfacial tension, elasticity ratio and on the radius of droplets and sub-inclusions. No generalized attempt was made to control the phenomena. Berger *et al.* [13] assumed that sub-inclusion formation in polyethylene terephthalate/polyamide-6 blends was related to an elasticity difference between both phases and evaluated its sign from the number average molecular weight difference. In one case, a high viscosity ratio was the source of composite dispersed phase formation. Ban *et al.* [14] observed a high sub-inclusion concentration concurrently with a decrease in droplet diameter when using a modified poly(ethylene-copropylene) (EPR) for reactive processing with nylon as compared to normal EPR. The results were explained by steric stabilization of the nylon sub-inclusions preventing coalescence with the matrix. No comment was made on the formation process. Favis and Lavallée [15] also reported the presence of a composite multi-phase for polypropylene/polycarbonate blends during melt processing.

In this article, a general process is shown for the formation of composite dispersed phase morphologies independent of surface tension, solubility or elastic effects.

2. Experimental procedure

2.1. Materials

The blends investigated in this study were prepared from commercial grades of bisphenol-A polycarbonate (pellets), Merlon M-39, Merlon M-40 and Makrolon 3103, obtained from the Mobay Corporation; polypropylene (powder), Pro-Fax 6301 obtained from Himont; nylon-6, Capron 8202 obtained from Allied Engineered Plastics and an ionomer, Surlyn 9020 obtained from Dupont. The properties of the resins, as specified by the suppliers, are summarized in Table I which also gives the dynamic viscosity, η', and the storage modulus, G', at 100 s^{-1}, at 250 °C under a strain of 10%. Both η' and G' were measured under dry nitrogen using a rheometrics mechanical spectrometer, RMS, model 605, equipped with a parallel plate fixture with a gap between the plates of 1.2 mm. Table I indicates that the PP sample is significantly less viscous than the PC samples. As for the storage modulus, which is an indication of the degree of elasticity of a given polymer, Table I shows that the PC samples have a higher storage modulus. Nylon and Surlyn both show a relatively low viscosity and storage modulus, nylon being slightly less viscous than Surlyn.

2.2. Mixing conditions

For the PP/PC blends care must be taken to dry the samples since polycarbonate can pick-up moisture fairly easily and undergo hydrolytic degradation during processing. They were therefore kept for about 20 h under vacuum at 95 °C. They were then taken out from the oven and added to the polypropylene samples only at the start of the mixing process in a Brabender chamber. The polypropylene in powder form was stabilized beforehand with 0.2% antioxidant. In order to minimize thermal degradation of polypropylene the temperature inside the chamber was maintained at 250 °C. Furthermore, all experiments were carried out under dry nitrogen. The roller blade attachment normally used for viscous materials was run at a constant speed of 50 r.p.m. After blending, the melt was rapidly shaped into a block inside a rectangular mould using a hydraulic press. The samples were stored for analysis at room temperature. The mixing process was done in three different ways: (a) For a 50/50% PP/PC blend (volume fraction), the materials were first dry blended in the desired proportion then introduced in the heated chamber and mixed for about 10 min; (b) For a 25/75% PC/PP concentration, the material (PP) destined to be the matrix in the final blend was blended as a fine dispersion in polycarbonate (80/20% vol PC/PP) for approximately 5 min. The rest of the polypropylene was then quickly added to the melt mixture to provoke phase inversion. The mixing was stopped either at the end of the melting process or after 5 additional min, and (c) the same process was used, but the concentration before inversion was 87/13% vol PC/PP in order to maximize the sub-inclusion concentration. In that case the mixing was stopped either 1, 3 or 6 min after the last addition of PP. Different matrix viscosities were studied.

The nylon-Surlyn blends were prepared using a similar procedure. Since nylon is also moisture sensitive, similar drying conditions were applied. The volume fraction of nylon before inversion was 20% vol (25% wt) and the blending time of this first dispersion was 20 min. More nylon was then added to provoke phase inversion and blended 3 more min (final concentration of Surlyn 30% vol). A blend with the same final concentration was prepared without inversion using a blending time of 3 min.

2.3. Morphological analysis

2.3.1. Optical microscopy (OM)

Optical observations of the polymer phases were made using a Leitz/Wetzlar, Dialux 20, in the bright field mode. The specimens were microtomed with a diamond or a glass knife mounted on a microtome (Reichert-Jung Supercut) at room temperature. An optimum specimen thickness of 2 µm was selected as lower thicknesses were not amenable to an easy and smooth cut.

2.3.2. Scanning electron microscopy (SEM)

The specimens observed were either fractured in liquid nitrogen or plane-faced using the Reichert-Jung Supercut microtome. After coating the desired surface with a gold–palladium alloy, the observations were carried out with a Jeol JSM-T220 scanning electron microscope operated at a voltage of 15 kV to avoid charging problems and the likelihood of artifacts.

2.3.3. Transmission electron microscopy (TEM)

Some specimens were also observed using the transmission electron microscope Phillips M100 at an accelerating voltage of 80 kV and the transmission electron microscope Hitachi H9000 at an accelerating voltage of 100 kV. Thin sections (90 nm) were microtomed using an ultramicrotome (Ultracut E from Reichert-Jung) and subjected to a staining procedure with ruthenium tetroxide as described by Trent *et al.* [9]. The 0.5% RuO_4 solution was used as-received from the manufacturer and the time of exposition to the RuO_4 vapours was about 30 min. Because polycarbonate and nylon have a higher affinity for RuO_4 than polypropylene or ionomer, they appear darker in the micrographs.

3. Results and discussion

3.1. Generation of composite multiphase morphology at or near region of dual phase continuity

From the previous investigation of the effect of composition on phase size in PC/PP blends [2], it was pointed out that the composite multi-phase morphology was observed at 50% (vol) PC. It was further noted that this structure was obtained near the phase inversion region. In order to confirm the presence of this structure a sample (50% vol fraction) was prepared according to method (a) (simultaneous addition) outlined in the Experimental section and was subsequently observed using a variety of techniques. The specimen of Fig. 1a was plane-faced; it shows two distinct phases with poor adhesion at the interface. Small spherical particles or their cavities are visible in one of the phases. These particles can clearly be seen in the specimen of Fig. 1b which has been subjected to a liquid nitrogen fracture. The composite morphology of one phase is evident. We can also infer from the two micrographs that, at this concentration, the composite phase is mainly made up of elongated structures and few droplets and that the over-all morphology is close to the co-continuous type. It can be seen in Fig. 1c that the observed composite elongated structures are made up of PC. A thin section of the sample was mounted on a copper grid and exposed to RuO_4 vapours for about 30 min [16]. Trent *et al.* [9] have already shown

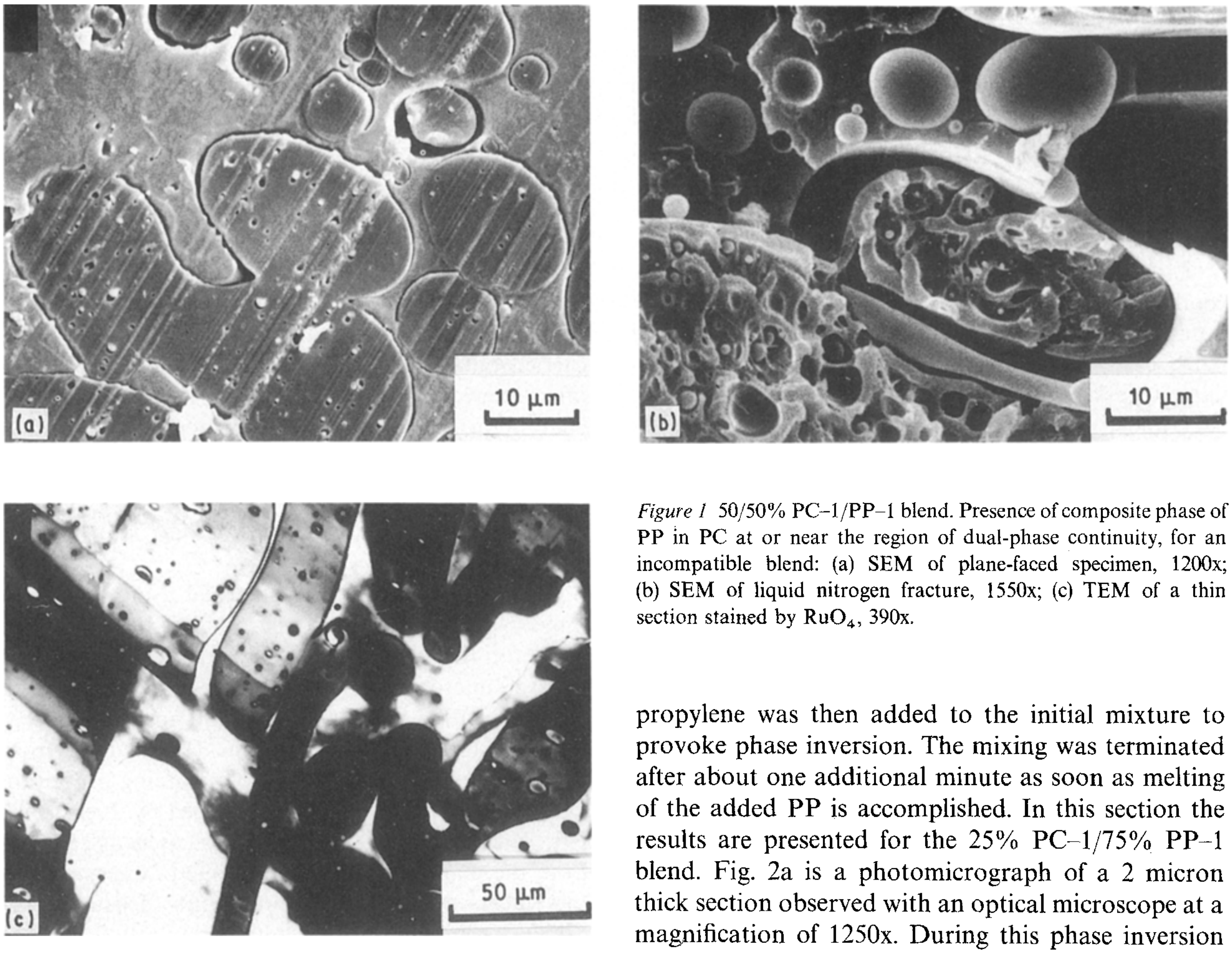

Figure 1 50/50% PC–1/PP–1 blend. Presence of composite phase of PP in PC at or near the region of dual-phase continuity, for an incompatible blend: (a) SEM of plane-faced specimen, 1200x; (b) SEM of liquid nitrogen fracture, 1550x; (c) TEM of a thin section stained by RuO_4, 390x.

that polycarbonate can be more effectively stained than PP and tests in this laboratory confirmed this conclusion. Two thin sections of pure PC and PP were simultaneously exposed to RuO_4 vapours for 2 h; at the end of the test, the PC specimen was entirely black while the PP specimen was very slightly altered. Fig. 1c indicates, therefore, that the composite elongated phase shown previously is the PC phase. These results confirm, using a variety of techniques, the presence of the composite multi-phase morphology in 50/50% PC/PP blends (vol fraction) reported previously. They indicate that the blend is composed of distinct regions of polypropylene and polycarbonate with the latter containing polypropylene sub-inclusions.

3.2. Generation of a composite dispersed phase morphology at low PC composition

Since a composite multi-phase morphology was observed near or at the region of dual-phase continuity, it was believed that this morphology could potentially be produced at lower concentrations by selectively invoking rapid phase inversion and subsequent control of mixing time. The method used is outlined in the Experimental section as method (b) (two step mixing). The mixing in this case was carried out as follows: all the polycarbonate and a portion of PP (in order to obtain a 20% volume fraction of PP) were dry blended and introduced in a Brabender chamber for a mixing period of 5 min. The remaining polypropylene was then added to the initial mixture to provoke phase inversion. The mixing was terminated after about one additional minute as soon as melting of the added PP is accomplished. In this section the results are presented for the 25% PC–1/75% PP–1 blend. Fig. 2a is a photomicrograph of a 2 micron thick section observed with an optical microscope at a magnification of 1250x. During this phase inversion process some of the initial PP remains as sub-inclusions within the more viscous polycarbonate dispersed phase as can be seen in the photomicrograph of Fig. 2a. If the mixing is allowed 5 more minutes after the end of the melting peak of PP, the PP sub-inclusions within the PC phase are almost non-existent as is illustrated in Fig. 3a. A closer view of the situation is shown in Figs 2b and 3b where two TEM micrographs illustrate the difference between the results obtained with the two mixing processes. In both micrographs PC appears as the dispersed darker phase. Within the composite phase of the specimen obtained by the first mixing process, PP is seen as small white domains. The dark lines crossing the PC droplet are marks left by the microtome's knife. No such marks are observed on the whiter PP matrix because PP is a much softer material.

3.3. Role of the viscosity of the dispersed phase

Increasing the viscosity of the dispersed phase should result in a lower mobility for the sub-inclusions and hence a higher retention of sub-inclusions in the final product. Various blends were prepared using three polycarbonates with low, medium and high viscosities to test this hypothesis. Their characteristics are given in Table I. The blend concentration is 25/75% vol PC/PP. Polypropylene, at a volume fraction of PP of 13%, was first added to PC and blended for 5 min. The rest of the PP was then added and mixed for 1, 3 and 6 min. A typical torque/time curve is given in Fig. 4. The first peak corresponds to the melting of the

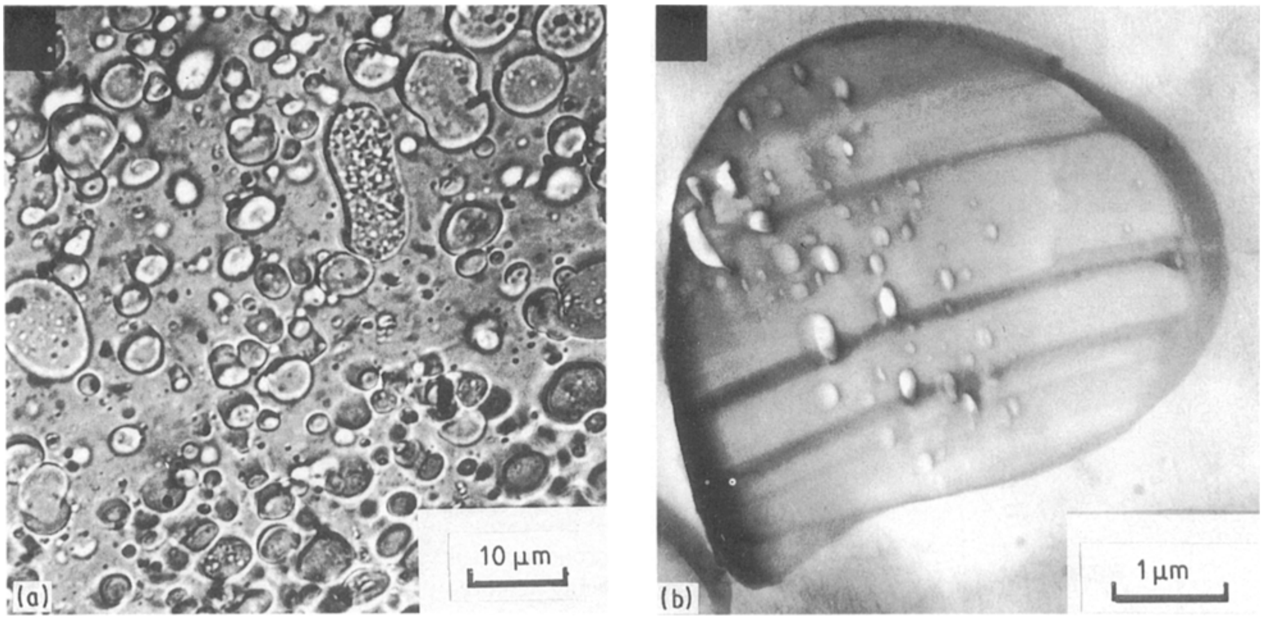

Figure 2 25/75% PC–1/PP–1 incompatible blend, blending stopped at the end of melting of 2nd addition of PP. Presence of a composite phase: (a) OM, 1250x; (b) TEM, 15000x.

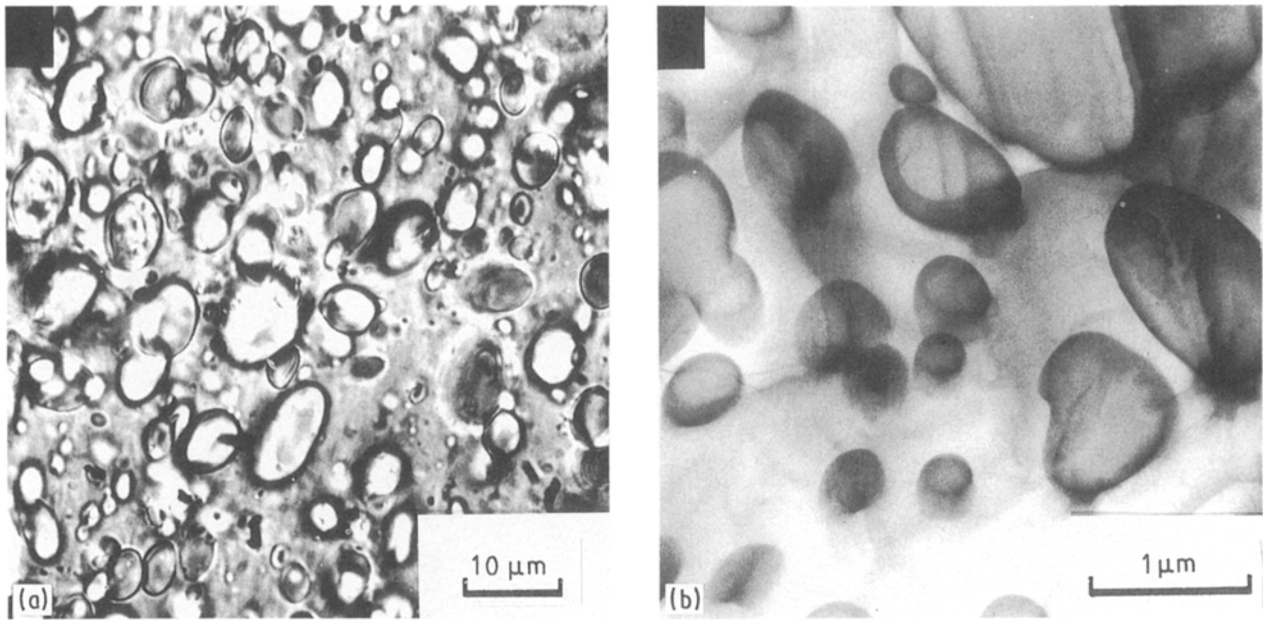

Figure 3 25/75% PC–1/PP–1 incompatible blend, blending stopped 5 min after melting of 2nd addition of PP. Absence of a composite phase: (a) OM, 1250x; (b) TEM, 21000x.

first dispersion and the second one to the melting of the second addition of PP (phase inversion). The arrows indicate times of blending previously mentioned. It should be noted that the steady state in torque is reached within one minute. TEM micrographs of these blends are shown on Fig. 5. As mentioned before, the concentration of sub-inclusions decreases with time. For two of the three blends studied (low and medium viscosities PC–1 and PC–2 as dispersed phase), there are no sub-inclusions (less than 1% retention as defined below) left after three minutes. After one minute of mixing, it can be seen that an increase in the viscosity of the dispersed phase results in an increase in the concentration of sub-inclusions, as expected. The PC–3 (high viscosity dispersed phase) is the only one to show sub-inclusions after 6 min, showing the increase in sub-inclusion stability with viscosity. Table II tabulates the percentage retention of sub-inclusions as a function of mixing time and viscosity of the dispersed phase. The percentage retention is defined as the volume fraction of PP sub-inclusions in the final material divided by the total volume fraction of PP in the initial dispersion before inversion. Volume fraction is equivalent to the area ratio as obtained by image analysis on TEM micrographs. Sub-inclusion retention below 1% was considered as zero retention.

With the PC–3 dispersed phase, the presence of sub-inclusions persists until roughly 20 min of mixing. These results have been summarized in Fig. 6, where the dispersed phase viscosity has been plotted against blending time. Blends exhibiting a composite dispersed phase morphology are represented by (○, □). From these values a boundary curve can be drawn showing limitations of the process in terms of minimum viscosity for a given time, and maximum time for a given viscosity.

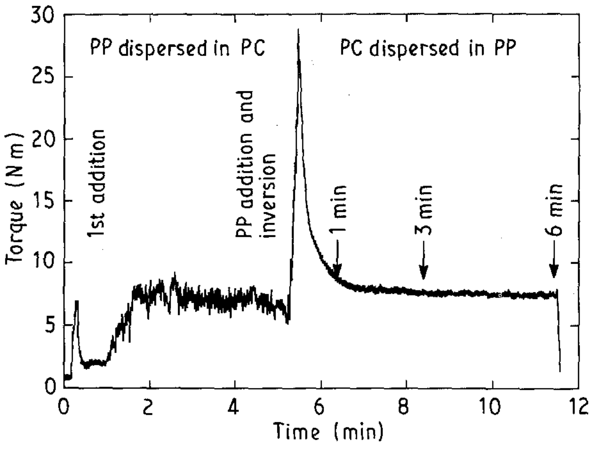

Figure 4 Typical torque versus time curve for phase inversion of an incompatible PC/PP blend.

3.4. Role of interfacial modification

It becomes apparent in the processing of composite dispersed phase morphologies that two aspects must be controlled. The first is the production of very fine sub-inclusions (relative to the final dispersed phase) and the second is the immobilization of those sub-inclusions during processing. Although the results with polycarbonate and polypropylene show how to control composite dispersed phase behaviour, blending time and viscosity limitations leave a narrow window for the preparation conditions. The use of an interfacial modifier should stabilize the first dispersion, producing fine particles (destined to become sub-inclusions in the final product) resulting in a higher volume fraction of sub-inclusions retained after phase inversion. As well, it would be expected that the range of processing times and material viscosities could be broadened since immobilization of sub-inclusions is being achieved primarily through interfacial chemical interactions.

In this study, a blend of nylon with an ionomer (copolymer of ethylene with methacrylic acid and isobutyl acrylate) was studied. Previous studies [17, 18] have shown that this blend is a two-phase system and that there is a covalent bond between the terminal

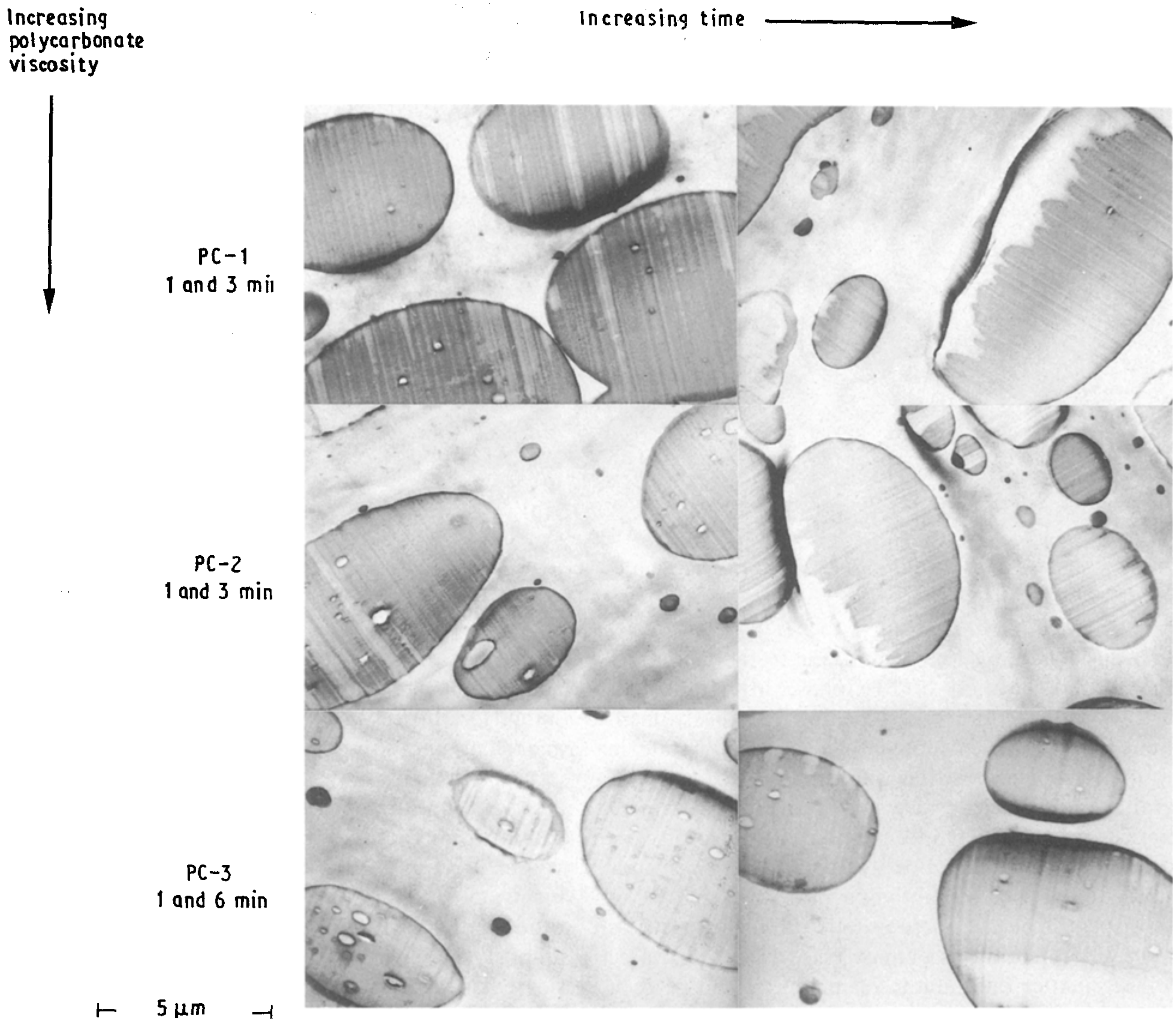

Figure 5 TEM of 25/75% PC/PP-1 blend. Dependence of composite dispersed phase morphology on increasing dispersed phase viscosity and time of mixing for incompatible blends, 5000x.

TABLE II Per cent retention of sub-inclusions

Polycarbonate \ Time (min)	1	3	6
PC–1	5	0 (< 1%)	0 (< 1%)
PC–2	6	0 (< 1%)	0 (< 1%)
PC–3	34	6	3

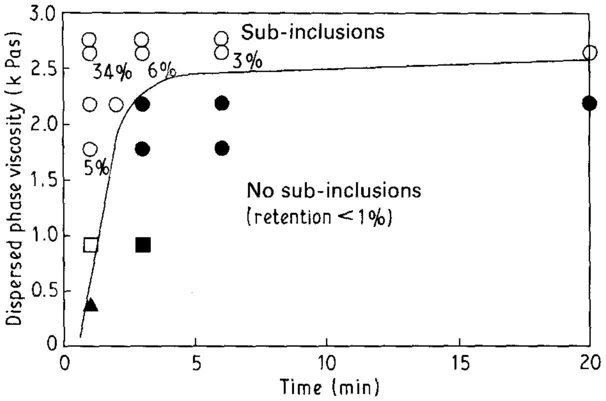

Figure 6 Viscosity/time limits required to form the composite dispersed phase in incompatible blends. The per cent retention of sub-inclusions is shown for selected points. (○, □) represent the presence of sub-inclusions; (●) PP/PC, (■) PA/PE, (▲) PA/PP represent the absence of sub-inclusions (< 1% retention).

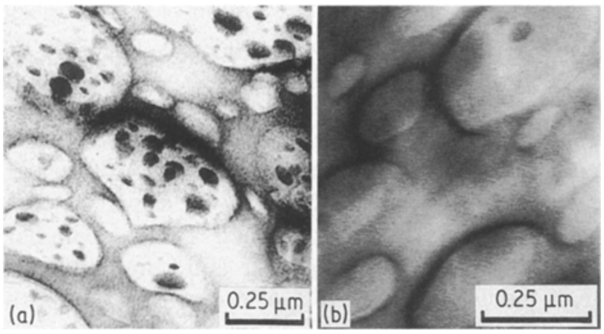

Figure 7 TEM of 30/70% ionomer/nylon blend. The influence of phase inversion is shown in forming a high concentration of sub-inclusions in the dispersed phase for compatible blends: (a) with phase inversion, presence of a composite dispersed phase; 100% sub-inclusion retention, 20% by volume of sub-inclusions relative to the dispersed phase, 42000; (b) without phase inversion, some spontaneous formation of composite morphology, but only 6% by volume of sub-inclusions relative to the dispersed phase, 59000x.

amine of the polyamide and the carbonyl of the copolymer. The ability of this ionomer to act as an interfacial modifier for nylon with polyethylene or polypropylene has been displayed and it has been shown in phase size/composition studies that the modified nylon/polyolefin blends show low rates of coalescence with composition [19]. The latter point indicates immobilization of the dispersed phase. Since the ionomer appears to experience stronger interactions with the nylon than with polyolefins [17], the nylon/ionomer blend was taken as a model system to investigate the role of good interfacial interactions on the formation of the composite dispersed phase morphology.

Several experiments were carried out to illustrate the applicability of the phase inversion process to the case of compatibilized blends. In the first experiment the predispersion contained 20% nylon/80% ionomer (by volume). A long blending time before inversion (20 min) was used to ensure good adhesion between both phases. Addition of an excess of nylon to provoke phase inversion was carried out, such that, the final product contained 30% ionomer/70% nylon (volume). Three minutes of mixing were carried out after phase inversion. The electron micrograph in Fig. 7a illustrates the significant volume of nylon sub-inclusions retained in the ionomer dispersed phase. In fact, calculation of the volume per cent of nylon present as sub-inclusions in the final product indicates that all the nylon present as droplets in the predispersion were retained as sub-inclusions after phase inversion. One hundred per cent sub-inclusion retention was achieved (i.e. 20% vol of nylon sub-inclusions based on the dispersed phase). In the second experiment the blending time after inversion was increased to 20 min. In this case sub-inclusion retention decreased slightly to 85% (17 vol % of sub-inclusions relative to the dispersed phase). In the third case a similar blend of 70% nylon/30% ionomer was prepared by simultaneous addition (no predispersion or phase inversion). This blend which was mixed for 3 min was found to result in approximately 6% (Fig. 7b) by volume of nylon sub-inclusions relative to the dispersed phase. Approximately three and a half times fewer sub-inclusions were obtained than in the previous experiments, thus showing the importance of the phase inversion step.

Parameters such as time-controlled phase inversion, the viscosity of the dispersed phase and chemical immobilization of sub-inclusions can be used to generate a composite dispersed phase morphology in incompatible and compatible blends during melt mixing. This approach is significantly different from other studies in which the sub-inclusion morphology is produced spontaneously [12, 20].

4. Conclusions

This study outlines an approach for generating novel composite multi-phase morphologies during melt processing. The results confirm the spontaneous formation of a composite multiphase morphology for a 50/50% incompatible blend of polypropylene and polycarbonate after melt blending near the phase inversion region. The morphology consists of co-continuous polypropylene and polycarbonate phases with the latter containing spherical polypropylene sub-inclusions. A composite dispersed phase morphology may also be prepared reproducibly and throughout the sample at a lower PC concentration ($\Phi_{PC} = 0.25$) by selectively imposing phase inversion and controlling the time of mixing. The morphology in this case consists of a polypropylene matrix, a polycarbonate dispersed phase and spherical polypropylene sub-inclusions within the dispersed polycarbonate. The control of the dispersed phase viscosity can improve the retention of the composite morphology with mixing time in incompatible systems. Studies on polyamide/polyethylene copolymer systems show that an interfacial reaction between sub-inclusions and dispersed phase results in complete stabilization and high retention of the sub-inclusions even for a relatively low dispersed phase viscosity and long mixing times. These preliminary results on model incompatible and compatible systems indicate the potential of preparing a composite dispersed phase morphology through melt blending, independent of solubility considerations. This technique could potentially be used to produce multicomponent blends where the physical properties of the dispersed phase itself can be closely controlled as a discrete blend. These results should be applicable to a wide range of systems.

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