

Morphology development in immiscible polymer blends: initial blend morphology and phase dimensions

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Abstract

The morphology of blends of polystyrene and polyethylene prepared by single screw extrusion and static mixing is shown to consist initially of sheets, which subsequently break up into droplets or threads. The rapid decrease of phase dimensions during blending can quantitatively be described by deformation of sheets in shear flow and not by deformation of droplets into threads. The final phase dimensions are found to be largely determined by the sheet thickness at the onset of break-up of the sheets. The type of morphology obtained after break-up can change during further processing without changing the phase dimensions significantly. At high values of the capillary number threads remain stable, whereas at low values droplets are formed. The final phase sizes appear to be almost independent of the capillary number. © 1999 Elsevier Science Ltd. All rights reserved.

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1. Introduction

The blending of polymers is an effective way of obtaining materials with a desired set of properties. Most polymers are immiscible, and blending usually leads to heterogeneous morphologies. The type of morphology and the phase dimensions determine the properties of a blend [1–3]. In order to be able to control the blend properties, the morphology development during processing should be understood. Most studies on polymer blending have been focused on elementary processes such as drop (de)formation, thread break-up and coalescence [4–15]. Less work has been done on the morphology development starting from the initial pellets [16–20]. Generally it is observed that a quick morphology change and a rapid decrease of phase dimensions occur in the initial stages of mixing [21]. Scott and Macosko [19] found that the initial morphology obtained by blending in a batch mixer consisted of ‘sheets’ of the dispersed phase. Lindt [17] and Sundararaj et al. [18] showed that this ‘sheeting’ mechanism also occurred in a single screw and twin screw extruder. The sheets break up as a result of the interfacial disturbances, eventually leading to a morphology of dispersed droplets or threads.

It will be shown in this article, that phase size reduction in single-screw extrusion proceeds through the formation of sheets, which eventually break up into threads or droplets. The measured phase dimensions can quantitatively be described by deformation of sheets rather than by deformation of droplets. It will be shown further that independent of the capillary number, the phase dimensions do not significantly decrease any further after sheet break-up, whether or not the blend is passed through a static mixer or just through neutral elements. The type of morphology obtained, however, does depend on the capillary number which governs the stability of the structures formed after sheet break-up.

2. Theory

The prediction of morphology and phase sizes obtained by the blending of immiscible polymers is difficult because of the complicated nature of the blending process. Several theories have been proposed for various blending mechanisms and for specific parts of the total process. So far, only a few theories have appeared pertaining to a ‘sheeting’ mechanism. In this section the traditional approach to a prediction of phase sizes in polymer blending will be compared to a prediction of the phase sizes based on a ‘sheeting’ mechanism.

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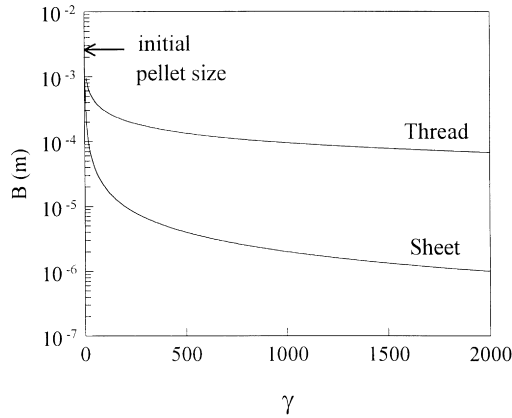


Fig. 1. The diameter of the thread and the thickness of the sheet in the case of affine deformation of a pellet of 3 mm thickness into a thread (Eq. (3)) or a sheet (Eq. (6)), respectively.

2.1. Droplets and threads

The classical approach to predicting the phase sizes in heterogeneous polymer blends is based on the elementary process of break-up of isolated droplets in a flow-field of a matrix phase, governed by the Capillary number:

$$Ca = \frac{\tau R}{\sigma} = \frac{\eta_m \dot{\gamma} R}{\sigma}, \quad (1)$$

which is the ratio of the deforming stress $\tau = \eta_m \dot{\gamma}$ (with η_m the viscosity of the matrix phase and $\dot{\gamma}$ the shear rate) and the interfacial stress σ/R (with σ the interfacial tension and R the droplet radius). Above a critical value, Ca_{crit} , the shear stress overrules the interfacial stress and the droplet is stretched until it breaks. The resulting droplet diameter B_{min} is:

$$B_{min} = \frac{2Ca_{crit}\sigma}{\eta_m \dot{\gamma}}. \quad (2)$$

Grace [22] showed that for break-up of Newtonian droplets in stationary flow, Ca_{crit} depends on the type of flow and the viscosity ratio, $p = \eta_d/\eta_m$ (η_d being the viscosity of the dispersed phase). Grace's observations are valid only for isolated droplets, whereas during mixing, a large number of droplets are close together which can coalesce leading to larger phase sizes. Furthermore, his values for Ca_{crit} give only the conditions for stability of a droplet in stationary flow. In reality, droplets will be subjected to non-stationary deformation, and at sufficiently high values of Ca , droplets will be stretched into long threads, which eventually will break up into a line of droplets [8,10]. As a result of the elongation the diameter of the extending droplet decreases and, consequently, the local capillary number decreases until the critical value for break-up, $Ca_{crit} = 1$, of the extended thread is reached [10]. The extended thread will then break up according to the Raleigh mechanism, and a line of droplets will be formed

with diameters approximately twice the thickness of the breaking thread.

The phase size thus obtained depends on the total deformation of the original droplet or pellet. The diameter of the thread, B_{thread} , depends on the total amount of imposed strain ($\gamma = \dot{\gamma}t$) according to Ref. [4]:

$$\frac{B_{thread}}{2R_0} = (1 + \gamma^2)^{-1/4}, \quad (3)$$

where R_0 is the radius of the original droplet, assuming affine deformation. This affine deformation is in shear possible only if $Ca/Ca_{crit} > 2$ [9]. In elongational flow, affine deformation occurs only if $Ca/Ca_{crit} > 5$ [10]. In most mixing devices the flows are dominated by shear as elongation is difficult to sustain for long times. For this reason and for reason of our experimental conditions only shear flow is considered in this article.

The diameter B_{thread} , as a function of γ is shown in Fig. 1. This figure demonstrates that the phase dimensions of 0.1–10 μm typically found after mixing are not predicted by Eq. (3) starting from the typical initial pellet size (3 mm). Delamare [14] used an initial droplet size of 0.53 μm , which was found after the melting zone in order to fit his results without explaining the use of this initial size. The typical phase dimensions found after blending can in special cases be explained by using the concept of reorientations [4]. By reorienting and folding the extending droplet ("bakers" transformation) the thread diameter will be given by:

$$\frac{B_{thread}}{2R_0} = \prod_{j=1}^n (\gamma_j)^{-1/2} \quad (4)$$

in which n is the number of reorientations and γ_j is the amount of strain between two reorientations. If, for example $\gamma = 2$ between each reorientation [9], 20 reorientations are necessary to arrive at $B_{thread} = 3 \mu\text{m}$, starting from 3 mm. The "bakers" transformation is, however, realised only in well designed static mixers and in certain dynamic mixers such as co-rotating twin-screw extruders. Single-screw extruders, however, are inferior for creating these reorientations. Yet, the phase dimensions after single-screw extrusion are usually in the range of 0.1–10 μm [17,18] and a different mechanism is required for explaining these observations.

Break-up of threads according to the Raleigh mechanism will occur only if the residence time of the thread is larger than the break-up time. This break-up time t_b can be estimated using Tomotika's theory [25]:

$$t_b = \frac{\eta_m B_{thread}}{\sigma \Omega_m} \ln\left(\frac{0.81 B_{thread}}{2\alpha_0}\right), \quad (5)$$

where α_0 is the amplitude of the initial disturbance, usually estimated between 10^{-7} and 10^{-9} m [23,24] and Ω_m is a function of the viscosity ratio [25]. Eq. (5) should be used appropriately. It is valid only for Newtonian systems in quiescent condition, which is usually not the case in

polymer blending. Stretching has a stabilizing effect against break-up of the liquid thread, leading to thinner threads [24].

The prediction of the phase sizes according to Eqs. (2)–(4) if applicable at all, will be invalidated even more if coalescence of the droplets occurs, leading to an increase of the average phase size. Coalescence will occur only if the concentration of droplets is sufficiently high and if the droplet diameter does not exceed a critical diameter [10].

2.2. Sheets

The theories briefly described above are valid for dispersed droplets and threads only and do not include the mechanism of their initial formation. In the initial stages of blending a quick morphology change occurs involving the formation of striated structures [12,16–20]. Plochocki [12] postulated an ‘abrasion’ mechanism for this morphology change. The group of Macosko [18–20] showed that a ‘sheeting’ mechanism is causing this morphology change, where pellets are stretched into ribbons or ‘sheets’ upon melting. Sundararaj et al. [20] presented a map showing the different conditions, where either ‘sheets’ or threads may be formed using the ratio of normal stresses and the Deborah number as parameters. It was shown that sheets can easily be formed in shear flow and that threads, however, are formed only at low shear rates ($<5 \text{ s}^{-1}$). Sheets are not transformed into threads because the interfacial forces are far too low under most circumstances to change the shape of the cross-section of the sheets significantly [8,20].

In the shear flow the thickness of the sheets decreases until the interfacial disturbances will initiate break-up of the sheets. The decrease in thickness of the sheets can be calculated as a striation thickness [4]. Starting from a sphere with initial radius R_0 a ribbon (‘sheet’) will be formed in shear flow having a width equal to the radius of the original sphere, but having a much smaller thickness B_{sheet} . The length of the ribbon after deformation is given by $L = 2R_0(1 + \gamma^2)^{1/2}$ if $\gamma \gg 1$ [4]. Its volume is $V = 1/4\pi(2R_0)B_{\text{sheet}}L$ which should be equal to the volume of the original sphere $V = 1/6\pi(2R_0)^3$. This leads to the following expression for B_{sheet} if $\gamma \gg 1$:

$$\frac{B_{\text{sheet}}}{2R_0} = \frac{2}{3}(1 + \gamma^2)^{-1/2}. \quad (6)$$

The sheet thickness, B_{sheet} , as a function of the overall deformation is shown in Fig. 1. From this figure it can be seen that the sheet thickness decreases much more rapidly with the deformation than the thread diameter does.

At a certain thickness the sheets will start breaking up by hole formation, which may be caused by interfacial instabilities which occur in layered structures during flow [26] or by impurities in the blend [18]. The holes will grow due to the interfacial forces and finally the holes will coalesce leaving a network of ligaments. This network can break up into threads or droplets [17], depending on the blending conditions. The phase dimensions of the resulting structures is of

the same order of magnitude as the final sheet thickness. Eq. (6) can, therefore, be used to predict the final phase sizes. From Fig. 1 it can be seen that the typical phase sizes of 0.1–1 μm found after blending are indeed predicted by Eq. (6).

It can be concluded that a ‘sheeting’ mechanism can explain the quick change in phase dimensions found in different mixing methods [12,16–20]. Shear flow is indeed an effective way of dispersing, because the reduction in striation thickness is concluded by break-up of the sheets into dispersed structures. The phase size reduction by way of deformation of threads, which finally break up into droplets is much less effective.

A further deformation of droplets after sheet break-up does not lead to the formation of new sheets, because at these small phase sizes the interfacial forces are of such a magnitude that droplets will be deformed into threads or will hardly be deformed at all. Deformation of these droplets into threads which subsequently break up is not very effective for size reduction as was shown earlier, so the phase dimensions are expected to stay approximately constant after sheet break-up. This will be shown below.

3. Experimental

Blends of polyethylene (PE) and polystyrene (PS) were made using different processing methods. A single screw extruder was used to study the sheet formation in a mixer that is inferior in achieving reorientations. Hence, an effective mixing was not expected. The effect of subsequent further mixing was studied by feeding the extrudate to a static mixer. A co-extrusion set up feeding both components to a static mixer separately was used to exclude the influence of the melting process on the morphology development and to create layered structures over a large range of phase sizes. In addition to the static mixer a variable number of ‘neutral’ elements were used, which only extend the residence time and the total amount of imposed shear. With these elements the morphology development after sheet break-up was studied.

For the single screw extrusion a Collin Laboratory single screw extruder equipped with a transport screw ($D = 20 \text{ mm}$, $L/D = 20$) was used, feeding a static mixer which contains an adjustable number (0–11) of static mixing elements. For the co-extrusion set-up [27] using the same static mixer, the Collin extruder, described above, and a Händle extruder ($D = 17 \text{ mm}$, $L/D = 20$) were used for feeding PE and PS separately to the static mixer, respectively. The PS melt is injected centrally in the PE stream as a thread with a diameter of approximately 2 mm just in front of the static mixer. The composition of the PS/PE blends can be varied by changing the screw rotational speed of the Händle extruder. The static mixing elements used in both types of the experiments were of the type Ross ISG (15 mm diameter), which contains four channels with a diameter of

Table 1
Trade name and shear viscosities at $\dot{\gamma} = 22 \text{ s}^{-1}$ of the polymers used

Polymer	Grade	η (Pa s) ($T = 200^\circ\text{C}$)	η (Pa s) ($T = 250^\circ\text{C}$)
PS	Hostyrene N2000 (SHELL)	780	160
PE1	StamylanLD 2100TN00 (DSM)	1860	1210
PE2	StamylanLD 2102TN26 (DSM)	960	540

2.7 mm. The average shear rate in these channels was estimated to be about 22 s^{-1} . The polymers and their viscosities are listed in Table 1. Blends of 5 and 17.5 wt.% PS in PE1 were used to study the sheet formation as a function of the number of mixing elements with both processing methods described above at 200°C . The extruded blends were quenched in water.

3.1. Stability experiments

Blends were made in the Collin extruder at 200°C , which fed the static mixer containing one Ross ISG element. The stability of these blends during further processing was studied by extending the static mixer with an adjustable number of ‘neutral’ elements with different lengths: 1, 2, 4, 6 and 7 cm. The elements contain four channels parallel to each other with the same diameter as the channels in the Ross ISG elements. The Ross ISG elements (Fig. 2(a)) enforce a ‘bakers transformation’, whereas the ‘neutral’ elements (Fig. 2(b)) only extend the residence time at a constant shear rate. By a combination of these elements the static mixer can be extended by 1–27 cm. These lengths correspond to an extended residence time of about 1–23.4 s. Two systems were studied: system 1, which contains 17.5 wt.% PS in PE1 and system 2, which contains 17.5 wt.% PS in PE2. Systems 1 and 2 were processed at 200 and 250°C in these ‘neutral’ elements, respectively, with the objective of comparing the stability of the morphology at different levels of viscosities.

3.2. Characterization

The extruded strands were broken in liquid nitrogen and extraction of the PS phase was performed in a Soxhlet extraction apparatus with 2-butanone during 3 days. This was sufficient for complete removal of the soluble fraction. In the case of sheets 100% of the PS phase can be extracted

as a result of the continuity of the sheet phase. If the sheets break up leading to a dispersion, less PS can be extracted. In the case of fibres, more PS can be extracted than in the case of droplets as will be shown later.

A scanning electron microscope (Philips XL 20) was used to study the morphology and the phase dimensions after extraction. Samples were fractured cryogenically prior to extraction. The reported droplet radii, fibre diameters and sheet thicknesses given are the results of 50–100 measurements of the holes resulting after extraction. In order to determine the stability of the fibres during flow, the morphology parallel to the flow direction was studied. In these cases the sample surfaces were smoothed parallel to the flow direction by using a diamond knife under liquid nitrogen. These smooth surfaces enable the determination of the length of the fibres.

4. Results and discussion

4.1. Single screw extrusion

The morphology found at the end of the extruder was an ‘onion’-like structure with rather irregular ‘sheets’ of 1–4 μm thickness and also with some fibres and droplets of the same size. A typical example of this structure is given in Fig. 3(a) and (b), showing the morphology both parallel and perpendicular to the flow direction of a blend of 17.5 wt.% of PS in PE1. The extraction of these blends resulted in the removal of 90–95% of the PS-phase indicating a large degree of continuity of this phase. From these experiments it appears that a ‘sheeting’ mechanism occurs in single-screw extrusion. The average shear rate in these experiments (20 s^{-1}) exceeded the critical value for sheet formation of 10 s^{-1} given by Sundararaj [20]. The width of the sheets (10–100 μm) appears to be smaller than the width of the original pellets (3 mm). Apparently, some break-up of the sheets already took place, which is not surprising considering the complex flow history of the pellets in the extruder. This may also explain the observed fibres and droplets.

The blend leaving the extruder was fed to a static mixer, and no further decrease in phase sizes was found except for the first mixing element, as is shown in Fig. 4. Increasing the number of mixing elements up to 10 did not result in smaller phase dimensions despite the extra shear and reorientations imposed by the static mixer. An example of the morphology of the blend after 10 mixing elements, both parallel and perpendicular to the flow direction is shown in Fig. 3(c)

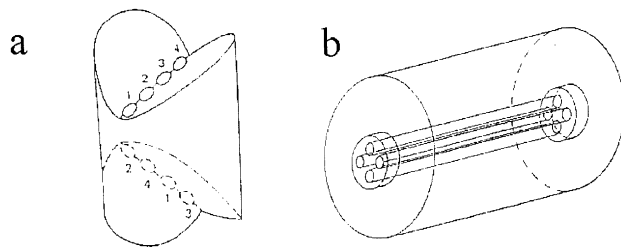


Fig. 2. The elements used in the static mixer: (a) Ross ISG and (b) ‘neutral’ elements.

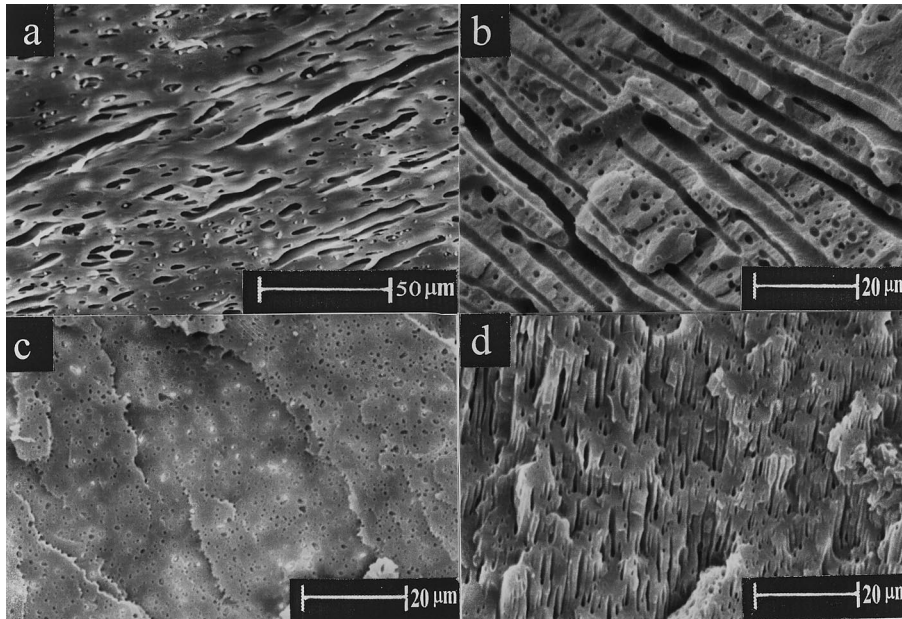


Fig. 3. Morphology of the blend with 17.5 wt.% PS in PE1: (a) and (b) represent the morphology at the end of the extruder screw parallel and perpendicular to the original flow-field, respectively; (c) and (d) represent the morphology after 10 mixing elements parallel and perpendicular to the original flow-field, respectively (after extraction of the PS phase).

and (d). The original sheet-like morphology has broken up into a fibrillar structure in this example. Depending on the capillary number break-up into droplets can occur as well. This will be discussed below.

The thickness of the sheets of 1–4 μm found after extrusion is in agreement with Eq. (6). According to this equation a sheet thickness of 2 μm will be formed from pellets of 3 mm at an overall deformation in the extruder of approximately $\gamma = 1000$. If a mechanism of thread formation and break-up occurred, fibres and droplets would have been found with a diameter of approximately 100 μm (Eq. (3)). This was not observed.

4.2. Co-extrusion

A sheet morphology was obtained by blending the molten

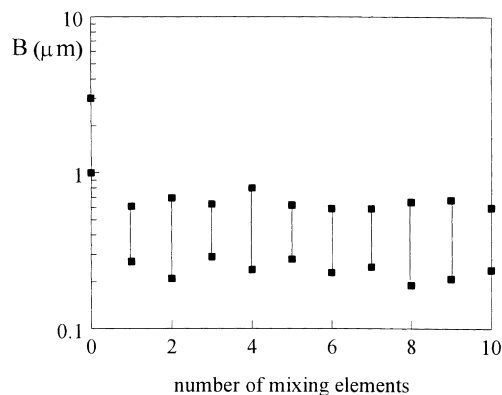


Fig. 4. The minimum and maximum phase dimensions of the PS phase as a function of the number of mixing elements following single screw extrusion.

polymers in the Ross static mixer if the number of mixing elements was smaller than 7. Using a larger number of mixing elements resulted in morphologies with dispersed threads or droplets. Sheets were obtained with a large distribution in thickness and with large disturbances on the surface. An example is shown in Fig. 5. The sheet thicknesses as a function of the number of mixing elements, determined from scanning electron micrographs, are given in Table 2. These thicknesses decrease rapidly with the number of mixing elements until sheet break-up occurs and a dispersed morphology is obtained with a constant phase size. This is shown in Fig. 6. The measured sheet thickness can be compared with the striation thickness as expected in the Ross static mixer [28]:

$$B_{\text{striation}} = \frac{0.002}{2(4^N)} \quad (7)$$

with N the number of mixing elements and 0.002 the initial thickness of the PS phase in m. Eq. (7) gives a fair description of the measured phase sizes for the mixing elements 1–6, for larger numbers of mixing elements sheet break-up occurs and the phase sizes remain constant.

The break-up of the sheets is illustrated by the results of extraction of the PS-phase. In the case of sheets 100% of the PS phase can be extracted, because the sheets form continuous structures. This continuity is destroyed if the sheets break up into a dispersion of either threads or droplets, leading to a lower fraction of the PS that can be extracted. The extraction results shown in Fig. 7 confirm that the sheets break up after six mixing elements. The phase dimensions of the resulting fibres or droplets remain remarkably constant even after a high number of mixing elements

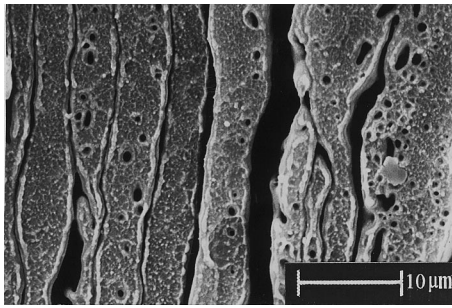


Fig. 5. Sheets of PS in PE1 prepared by co-extrusion after extraction of the PS phase (after extraction of the PS phase).

(Table 2 and Fig. 6). The type of morphology, however, can vary as will be shown in the next section.

4.3. Morphology development after sheet break-up

Break-up of blend morphologies under conditions of flow was studied by extending the static mixer with ‘neutral’ elements as described in Section 3. Two blends were compared: system 1, consisting of 17.5% PS in PE1, and system 2 consisting of 17.5% PS in PE2. Both the blends exhibit a fibrillar morphology after passing the single-screw extruder and the static mixer (containing one mixing element) as shown in Figs. 8(a) and 9(a). The fibres in blend system 1 did not break up during the flow through the complete ‘neutral’ section (residence time is 23 s) as shown in Fig. 8(b) and confirmed by the extraction results in Fig. 10. The fibres in blend system 2, however, broke up and droplets were formed as shown in Fig. 9(b). This break-up is confirmed by the decreasing amount of extractable PS shown in Fig. 10.

The relevant parameters, calculated capillary numbers and calculated break-up times for both systems are given in Table 3. In system 1, $Ca = 2$ and break up did not occur, although the process time exceeded the calculated break-up time, whereas in system 2, $Ca = 0.8$ and break-up of the fibres

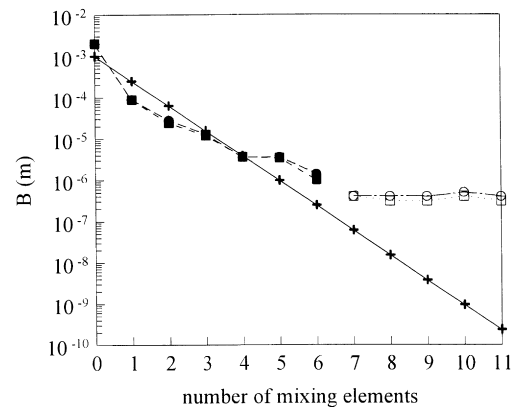


Fig. 6. Phase dimensions of the PS phase in PE1 as a function of the number of Ross ISG mixing elements after coextrusion; +: striation thickness (Eq. (7)); ■ and ●: thickness of sheets of 5 and 17.5 wt.% PS, respectively; □ and ○: diameters of threads/droplets of 5 and 17.5 wt.% PS, respectively.

took place within 4 s. It appears that the shear stabilizes the fibres if $Ca > 1$, whereas break-up occurs if $Ca < 1$.

The phase dimensions of these systems are given in Table 4. The phase dimensions (fibre diameter and droplet diameter) did not change significantly in either blend system on passing the ‘neutral’ elements. This is analogous to the behaviour in the ‘active’ mixing elements after break-up of the sheet structure as shown in Figs. 4 and 6. The average value of L/B in system 1 remains approximately 30 and the diameter of the fibres did not change significantly as shown in Table 4. The values of L/B in system 2 decreased rapidly because of the break-up of the fibres as shown in Table 4. It was found that after break up, the droplets were a factor of 1.6 as thick as the original fibre. These droplets, formed after break up, did not deform into a long thread anymore.

5. General discussion

Sheets are formed in the initial stage of polymer blending. This is found in processes where blending and melting

Table 2

Sheet thickness and particle size as a function of the number of mixing elements in case of blends with 5 and 17.5 wt.% PS in PE2 prepared by co-extrusion

Number of elements	5 wt.% PS		17.5 wt.% PS	
	Sheet (μm)	Particles (μm)	Sheet (μm)	Particles (μm)
0	≈ 2000	–	≈ 2000	–
1	88.6 ± 19.1	–	89.9 ± 21.0	–
2	23.8 ± 15.9	–	28.8 ± 21.8	–
3	11.7 ± 10.0	–	12.9 ± 11.5	–
4	3.6 ± 3.5	–	3.6 ± 3.6	–
5	3.4 ± 3.0	–	3.7 ± 3.5	–
6	1.0 ± 0.8	0.3 ± 0.2	1.4 ± 1.0	0.4 ± 0.1
7	–	0.4 ± 0.3	–	0.4 ± 0.2
8	–	0.3 ± 0.2	–	0.4 ± 0.2
9	–	0.3 ± 0.1	–	0.4 ± 0.3
10	–	0.4 ± 0.2	–	0.5 ± 0.3
11	–	0.3 ± 0.2	–	0.4 ± 0.2

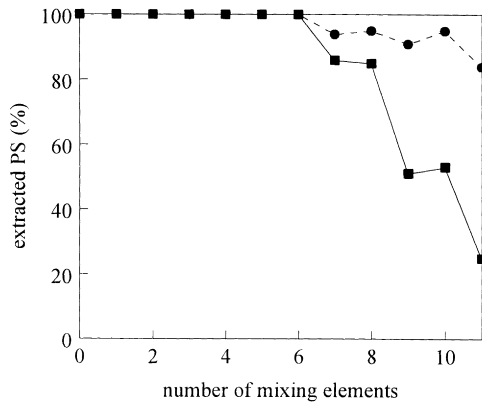


Fig. 7. Fraction of PS extracted from blends prepared by co-extrusion as a function of the number of mixing elements: ■ = 5 wt.% PS; ● = 17.5 wt.% PS.

proceed simultaneously, as was shown for blending of immiscible polymers in batch mixers [19], single screw [17] and twin screw extruders [18]. In these blending processes melting and mixing proceed concurrently and sheet formation can be seen as the consequence of the interaction of these process steps [16,18]. Yet, sheet formation need not necessarily be the consequence of a concurrent melting/mixing process, but may well be the consequence of the geometry of shear flow. Shear flow leads to an increase in the length of a fluid particle in the flow direction at sufficient large deformations, and to an associated decrease in thickness, leaving the width largely unchanged. Molten pellets are thus deformed into ribbons ('sheets'), when subjected to shear flow. This view is supported by

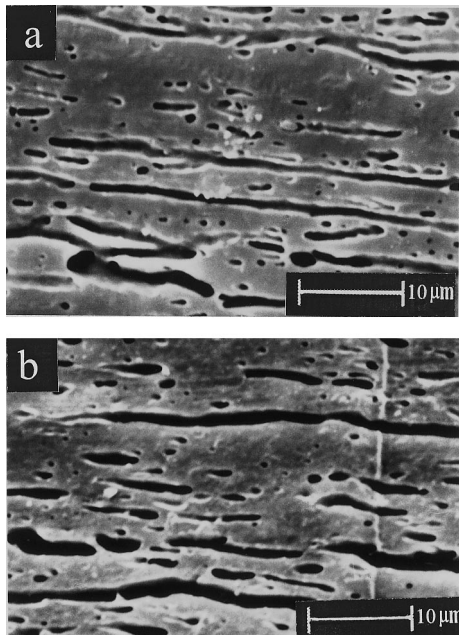


Fig. 8. Morphology of a blend of 17.5 wt.% PS in PE1 prepared by a single screw extrusion parallel to the flow direction: (a) after leaving the Ross ISG mixing element and (b) after 23 s ($\gamma = 515$) leaving the 'neutral' elements. The fibres were stable.

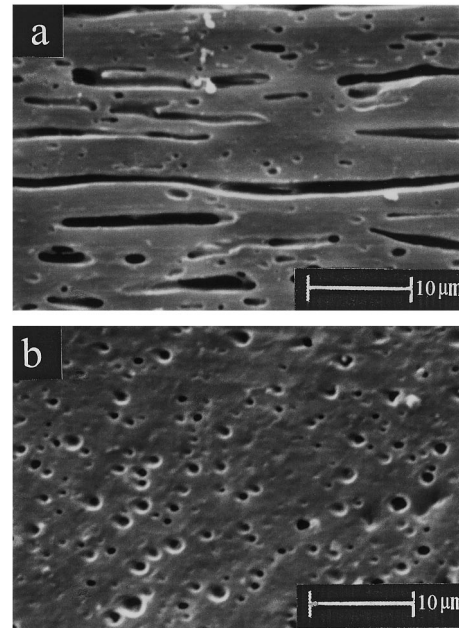


Fig. 9. Morphology of the blend of 17.5 wt.% PS in PE2 prepared by a single screw extrusion parallel to the flow direction: (a) after leaving the Ross ISG mixing element and (b) after 4 s ($\gamma = 100$) leaving the 'neutral' elements. The fibres break up.

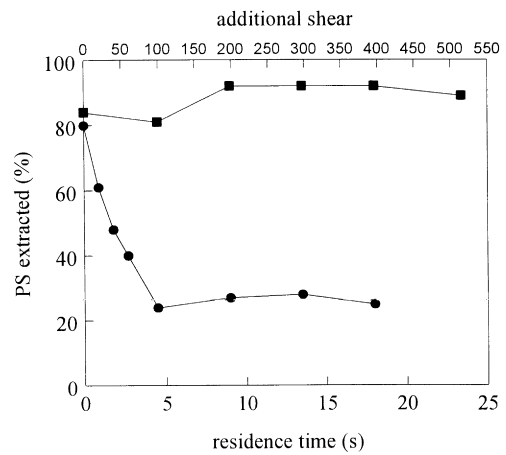


Fig. 10. Fraction of PS extracted as a function of the residence time in the 'neutral elements': ■ = system 1, ● = system 2. The fibres in blends of system 2 break up.

Table 3
Viscosity ratio, interfacial tension, phase dimensions and the resulting capillary numbers and theoretical break-up times (Eq. (5)) during processing in 'neutral' elements

System	p	σ (mN/m)	$B_{\text{fibre } t=0}$ (μm)	Ca	t_b (s)
1	0.4	4.5 [3]	0.4 ± 0.2	2.0	8
2	0.3	3.5^a	0.5 ± 0.2	0.8	3

^a Calculated using the theory described in Ref. [29].

Table 4

The average aspect ratio, L/B and diameter of the fibres (system 1) and fibres/droplets (system 2) as a function of the residence time (t) in 'neutral' elements

Flow time (t) (s)	Average extra shear	System 1—fibres for all t		System 2— fibres: $t < 1.8$ s; droplets: $t > 1.8$ s	
		L/B	B (μm)	L/B	B (μm)
0	0	30	0.4 ± 0.2	34	0.5 ± 0.2
0.9	20	—	—	31	0.5 ± 0.2
1.8	40	—	—	31	0.5 ± 0.2
4.5	100	26	0.7 ± 0.3	4	0.6 ± 0.2
9.0	200	24	0.6 ± 0.3	2	0.7 ± 0.2
13.5	300	39	0.5 ± 0.2	2	0.8 ± 0.3
18.0	400	31	0.6 ± 0.2	3	0.8 ± 0.3
23.4	515	39	0.6 ± 0.2	3	0.8 ± 0.3

our micrographic observations and by the good agreement between measured phase sizes and predictions based on Eq. (6).

As stated earlier, this mechanism of deformation of 'sheets' followed by break-up is an effective way of dispersing two polymers. It is much more effective in reducing dimensions of the dispersed phase than deformation of droplets into threads followed by formation of lines of smaller droplets. This latter mechanism involves two-dimensional reduction of lateral dimensions of the thread and implicitly requires elongational flow or large interfacial stresses in order to maintain the cylindrical shape. Of course, sheets can be formed only as long as the interfacial stresses are small compared to the deforming stresses.

As expected, sheets are formed by feeding the static mixer with separately prepared polymer melts. The measured layer thicknesses neatly follow the power law for the striation thickness (Eq. (7)) until break-up sets in. This method of producing fairly well defined sheets leads to the same sheet thickness for break-up and the same final phase dimensions as the single-screw extrusion experiments.

The final phase dimensions are determined by the break-up of the sheets. It appears that a critical sheet thickness for break-up exists, which probably is determined by the growth of disturbances on the sheet surface. The growth of such interfacial disturbances depends on rheological parameters and process conditions [30,31], a phenomenon which is not yet fully understood. If the critical sheet thickness could be predicted, the phase dimensions would be known a priori. From our results it appears that the critical thickness for sheet break-up is not dependent on the viscosity of the major component.

After sheet break-up, the phase dimensions do not change significantly whether or not the blend is passed through a static mixer or just through neutral elements. The type of morphology obtained after break-up of the sheets depends on the stability of the fibrous structures formed after sheet break-up. At high values of the capillary number threads are stable, whereas at low values the threads break up into droplets. The lateral dimensions of the threads and droplets

are approximately equal. The remarkable result is that the final dimensions obtained on blending are not dependent on the value of the capillary number, contrary to classical theories.

6. Conclusion

It is shown that a 'sheeting' mechanism occurs during the initial stage of mixing, both in single screw extrusion and static mixing. This mechanism leads to a rapid decrease of the phase dimensions as shown both experimentally and theoretically. After the break up no further significant decrease of the phase dimensions is found.

The type of morphology depends on the stability of the fibres formed after the sheet break up. If the capillary number is higher than one, a fibre/matrix morphology is formed if this capillary number is lower, break up occurs and a droplet matrix morphology is formed. The final phase sizes do not depend on the capillary number.

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