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Compatibilization of PBT–PPE blends using low molecular weight epoxy

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Abstract

Compatibilization of blends of an immiscible polymeric system consisting of poly(butylene terephthalate) (PBT) and poly(phenylene ether) (PPE) by a set of low molecular weight epoxy, varying in chain flexibility and functionality, was studied in terms of morphology and mechanical properties. Evidences, such as reduction of epoxide peak in FT-IR spectra and viscosity and glass transition temperatures higher than PBT of melt-blended materials of PBT–epoxy indicated formation of PBT–epoxy copolymers, which in turn compatibilized and stabilized the morphology in PPE–PBT–epoxy blends. Significant improvements in tensile and impact strengths resulted.

Keywords: Compatibilization; Epoxy; Copolymer

1. Introduction

Compatibilization of an incompatible blend system consisting of poly(butylene terephthalate) (PBT) and poly(phenylene ether) (PPE) was investigated in the present study using a set of low molecular weight thermosetting epoxy resins. Although both PPE and PBT possess some useful properties, their uncompatibilized blends do not exhibit many interesting properties [1]. PBT offers strong impact properties and excellent electrical resistivity. A relatively lower glass transition temperature, however, has a negative impact on the dimensional stability under varying thermal conditions. Also, its lower melt viscosity and high crystalline density results in poorly defined sample shapes during certain processing operations. PPE, on the other hand, is a tough engineering polymer with high glass transition temperatures, low thermal expansion coefficient, and low mold shrinkage, leading to good dimensional stability of molded parts [2], although its impact properties are poor and it is rarely used without some impact modifiers, one example being high impact polystyrene.

Till date only a few studies have focused on improving compatibility of these two polymers [3–5]. In each case, compatibilizer molecules were selectively used so that they contained two functional parts to interact with each polymer phase, e.g. styrene–glycidyl methacrylate copolymer [3], copolymers containing polystyrene blocks and methacrylate end groups [5], and block copolymers of polystyrene and polycarbonate [6]. In all these cases, styrene blocks produced miscibility with PPE-phase and carboxylic ester groups participated in transesterification reactions with the –COOH end groups of PBT.

In this study, low molecular weight epoxy molecules, containing at least two epoxide functional groups, were considered as compatibilizers instead of functional copolymers. The chemical reactions between the epoxide groups and the unreacted –COOH groups of polyesters and polyamides have been known in the literature and led to the development of compatibilized blends of polyesters and polyamides with other polymeric materials [7–13]. The miscibility of PPE with low molecular weight epoxy was also exploited in a series of recent studies in developing polymeric composites of glass and carbon fibers [14–19]. The miscibility of PPE and epoxy, however, gradually decreases with crosslinking reactions of epoxy, i.e. with the conversion of epoxide groups [17–19]. It is, therefore, imperative to have unreacted epoxide groups in the mixtures if effective compatibilization of PBT–PPE blends is desired. In the present study, we exploited these features of miscibility and reactions of epoxide functionalities, respectively, with PPE and –COOH groups of PBT to develop compatibilized PBT–PPE blends. Excess amounts of epoxy than required by stoichiometry were used.

Compatibility of both PBT- and PPE-rich blends was investigated, although the mechanical properties of only the PBT-rich blends were fully characterized in this study. The effects of flexibility of chains and the reactivity of

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Table 1
Important properties of epoxy resins and equivalent weight ratios with PBT (Product Literature, Shell Chemical Company, 1998)

Epoxy resin	Equivalent weight[a]	Equivalent weight ratio, epoxy:PBT[b]	Viscosity (Poise)	Density (g cm^{-3})	Melting.point (°C)	Boiling point (°C)
EPON® 828	185–192	0.007	110–150 at 25°C	1.16	–	315
EPON® 832	400–425	0.015	350–600 at 25°C	1.07	–	–
EPON® SU-8	192–230	0.072	10–60 at 130°C	1.2	82	–

[a] Grams of resin containing one gram equivalent of epoxide.

[b] PBT equivalent weight is 26,200.

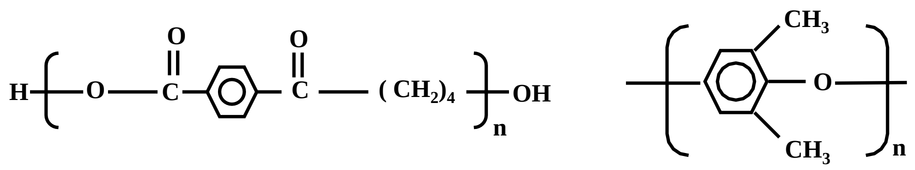
epoxy groups on morphology and mechanical properties were studied using two bifunctional epoxies differing in epoxy equivalent weights (EEW) and one octa-functional novolac-type epoxy. The stability of dispersed phase domains upon annealing indicated the effectiveness of epoxy as compatibilizer.

2. Experiments

2.1. Materials

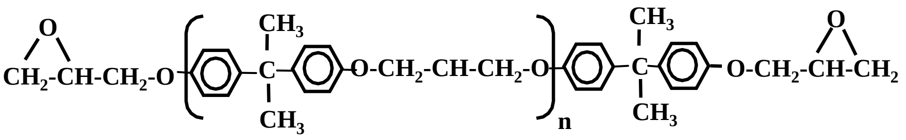
PBT in the form of VALOX 315 pellets and PPE in the form of unmodified powder were obtained from GE Plastics. The properties of PBT, supplied by GE Plastics, are as follows: weight average molecular weight (M_w), 26,320; specific gravity, 1.31; tensile strength (ASTM D638), 52 MPa; notched Izod impact strength (ASTM D256) at room temperature, 50 J m^{-1}; glass transition temperature (T_g, DSC), 43°C; melting temperature (DSC), 220–225°C; and concentration of –COOH and –OH, respectively, are 38 and 22 meq kg^{-1}. Molecular weight (M_w) and T_g of PPE were evaluated in our laboratory: $M_w = 25,200$ (GPC); $T_g = 210$–215°C.

Three different epoxies were used as prospective compatibilizers — EPON® 828, 832, and SU-8, all supplied by Shell Chemical Company. EPON® 828 and 832 are bifunctional and EPON® SU-8 is an octa-functional epoxy. EPON® 828 and SU-8 have almost the same EEW. Molecules of EPON® 832 are more flexible than EPON® 828. Table 1 lists important properties of the epoxies used and the equivalent weight ratios of epoxy components and PBT.

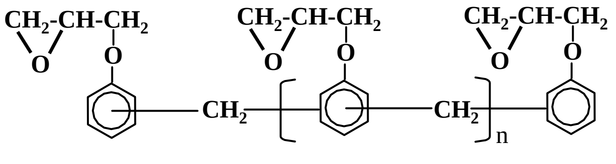


Polybutyleneterephthalate (PBT)

Polyphenyleneether(PPE)



EPON 828: Diglycidyl ether of bisphenol A (DGEBA); n=0.1



Novolac epoxy resin, e.g., EPON SU-8 with n=6

Fig. 1. Chemical structures of PBT, PPE, and EPON® 828 and SU-8 showing –COOH end group of PBT and epoxide groups of epoxies.

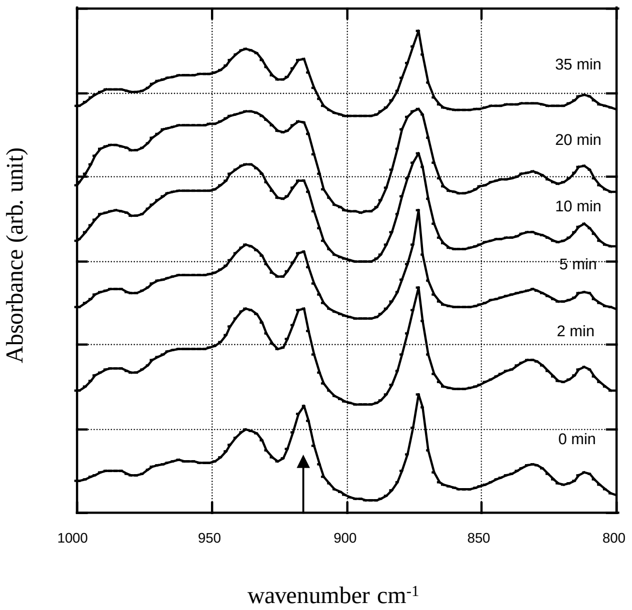


Fig. 2. FT-IR spectra for PBT–EPON® 828 mixtures as function of reaction time. PBT and epoxy were present in the weight ratio of 70:5. The reaction was carried out at 250°C.

The chemical structures of some of the components are shown in Fig. 1. Chloroform and hexafluoroisopropanol (HFIP), acquired from Aldrich Chemicals, were selected as solvents of PBT and epoxy mixtures.

2.2. Solution blending

Solution blended samples of PBT and epoxy were used to study PBT–epoxy reactions by FT-IR and to evaluate glass transition temperature (T_g) of the blends before and after reactions. The blends contained 70 parts by weight of PBT and 2, 5, and 10 parts by weight of epoxy. The epoxy content in these blends was much more than required by stoichiometry (Table 1). A solvent composed of 75% by weight of chloroform and 25% by weight of HFIP was used to dissolve the materials at room temperature. The solutions were cast into thin films on Teflon® trays and dried in vacuum oven for 24 h at room temperature.

2.3. Melt blending

PPE, PBT, and epoxy were melt-blended in a batch mixer, Brabender Plasticorder, at 250°C and 50 rpm. The components were added at various times during mixing, although the maximum time of mixing never exceeded 35 min. PBT-rich blends contained 70 parts PBT and 30 parts PPE; PPE-rich blends, on the other hand, consisted of 30 parts PBT and 70 parts PPE, all by weight. The epoxy content was varied between 2, 5, and 10 parts by weight.

Table 2
Glass transition temperatures of PBT–epoxy blends given in °C before and after reactions at 250°C for prescribed times. For reference, PBT glass transition temperature is 43°C. The compositions are in parts by weight

Blend	Rection time				
	0 min	5 min	10 min	20 min	35 min
PBT:EPON® 828 = 70:2	35	37	39	42	45
PBT:EPON® 828 = 70:5	29	34	37	39	41
PBT:EPON® 828 = 70:10	33	28	32	36	34
PBT:EPON® 832 = 70:2	40	45	47	48	50
PBT:EPON® 832 = 70:5	39	40	45	42	46
PBT:EPON® 832 = 70:10	36	39	40	41	43
PBT:EPON® SU-8 = 70:2	44	46	46	47	50
PBT:EPON® SU-8 = 70:5	44	45	46	47	48
PBT:EPON® SU-8 = 70:10	42	45	46	47	46

3. Results and discussion

3.1. PBT–epoxy and PPE–epoxy reactions

The first sign of PBT–epoxy reaction was observed when T_g values of PBT–epoxy blends were compared before and after the reactions, as shown in Table 2. The PBT and epoxy components of the solution-blended solid films were allowed to react in an oven at 250°C for periods of time varying between 2 and 35 min.

Unreacted blends of PBT with EPON® 828 and 832 showed decrease in T_g compared to that of neat PBT, indicating some degree of mutual solubility between the components, as reported in the literature [20–22]. The drop in T_g was most noticeable in the case of EPON® 828. Almost no changes in T_g — before and after reactions — was observed in the case of SU-8, indicating poor miscibility with PBT.

The values of T_g of blends of EPON® 828 and 832 increased with reaction times. This data indicated the possibility of PBT–epoxy reactions, which was investigated further using FT-IR and ARES Rheometrics rheometers.

A Nicolet Magna-550 FT-IR, Series II machine was used to generate FT-IR spectrum of solution-blended thin films of PBT–epoxy before and after reactions. PBT–epoxy blends were allowed to react with each other for various times between 2 and 35 min. PBT was characterized by wave numbers of 1720 and 1268 cm^{-1} indicating, respectively, the C=O and C(=O)–O stretching of aliphatic ester group and the peak at 727 cm^{-1} corresponds to the out of plane C–H bending of substituted benzene ring in PBT [23–24]. The epoxide peak at 917 cm^{-1} and the peaks at 825 and 1031 cm^{-1} corresponding to substituted aromatic groups characterized epoxies [25]. The peak and the area under the curve for epoxide linkage were observed to decrease over time indicating conversion of epoxide groups, as presented in Fig. 2 for PBT–EPON® 828 blends. Similar trends were found in the case of EPON® 832 and SU-8, although DSC studies did not reveal increase of T_g in the case of SU-8.

The decrease in epoxy peak in Fig. 2 can be attributed to the

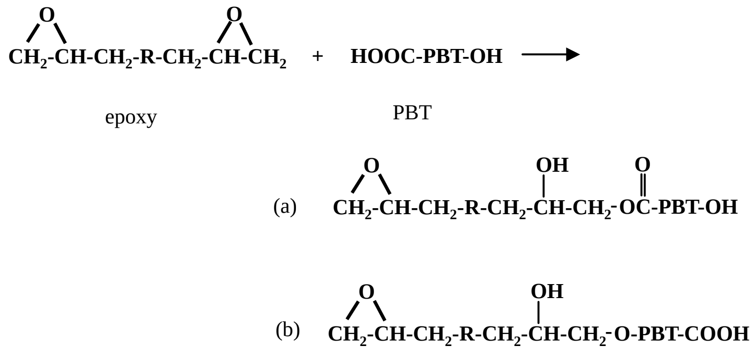


Fig. 3. Possible reactions between PBT and epoxy for copolymer formation. (a) –COOH and epoxide groups (b) –OH and epoxide groups.

following: (1) evaporation of epoxy, (2) homopolymerization of epoxy at the reaction temperature, and (3) PBT–epoxy reactions. To investigate the extent of evaporation, a thermogravimetric analysis (TGA) of solution-blended samples was carried out at 250°C in a stream of air for a period of 35 min, the longest time used in the reactions, to explore the possibility of loss of epoxy due to evaporation. A maximum loss of 2% based on epoxy weight was observed during this period of time, which was very small compared to ~60% conversion over a period of 35 min. The conversion of epoxy was calculated based on the area under the curve of the epoxy peak at 917 cm^{-1}. A recent study [26] discounted the possibility of homopolymerization of EPON 828 during heating at a scan rate of 10°C min^{-1} from room temperature to 450°C. It is, therefore, believed that the reduction in epoxy peak in Fig. 2 was mainly due to conversion of epoxide groups during reaction with PBT. Fig. 3 presents two possibilities of such reactions between –OH and –COOH groups of PBT and epoxide groups, although Fig. 3(a) is more likely due to higher reactivity of carboxylic acid group than primary alcoholic –OH group.

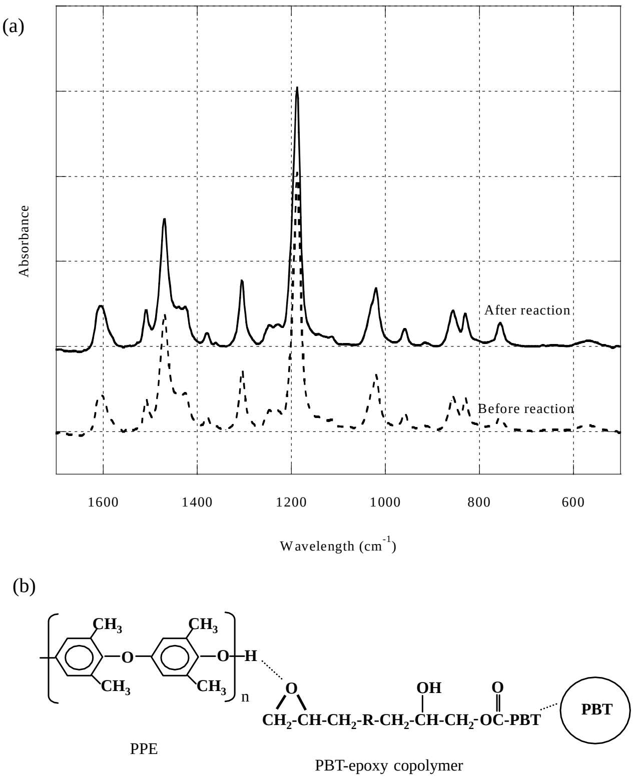


Fig. 4. (a) FT-IR spectra of reacted and unreacted blends of PPE (30 parts)–EPON® 828 (10 parts). (b) Possible compatibilization route between PBT and PPE by PBT–epoxy copolymer.

PPE and epoxy components were solution-blended in chloroform and allowed to react at 250°C for 35 min under nitrogen environment. Solution-casted films of the reacted materials were analyzed using FT-IR for possible reactions between phenolic –OH groups of PPE and epoxide groups of epoxy. Fig. 4(a) shows comparison of FT-IR spectra of samples before and after the reaction, normalized to the same sample thickness. It is apparent that negligible reactions occurred between PPE and EPON® 828 in 35 min at 250°C. Consequently, a possible route to compatibilization can be proposed, whereby the PBT–epoxy copolymers form energetic interactions, such as hydrogen bonding, with the phenolic –OH groups of PPE, as shown in Fig. 4(b).

3.2. Rheology and mixing torque

Indirect evidence of PBT–epoxy copolymer formation was also obtained from evaluation of shear viscosity of pre-reacted PBT–epoxy and PBT–epoxy–PPE blends and from mixing torque measured during melt-blending of PPE, PBT, and epoxy. Reacted PBT–epoxy (EPON® 828) mixture with 2 phr epoxy showed higher viscosity than PBT, possibly due to copolymer formation as alluded to in Fig. 3, although lower viscosities were obtained for blends with 5- and 10 phr epoxy, probably due to excess epoxy (Fig. 5). The same trend is reflected in three-component PBT–epoxy–PPE blends as presented in Fig. 6. It is interesting to note in Fig. 5 that PBT–epoxy reactions did not take PBT-phase viscosity anywhere near the PPE-phase viscosity. Also, the miscibility of a small fraction of epoxy with PPE should not make PPE-phase viscosity comparable to that of PBT-phase. Therefore, phase deformation and breakup of dispersed phase in PBT–epoxy–PPE

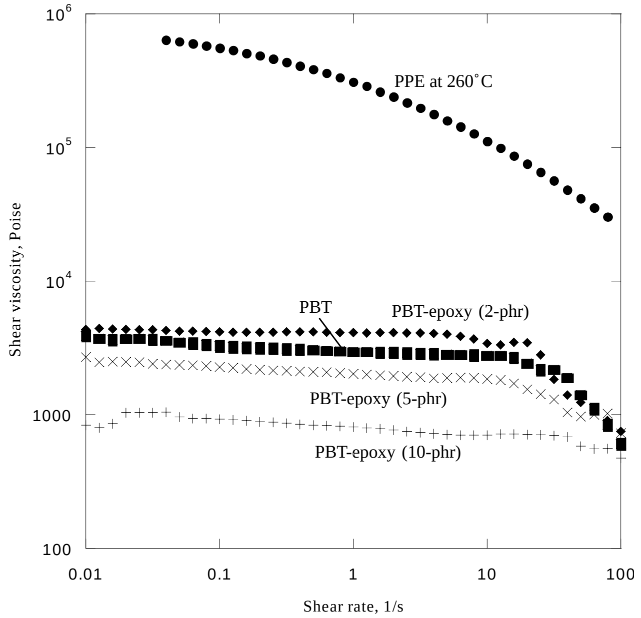


Fig. 5. Shear-viscosity of pre-reacted PBT–epoxy (EPON® 828) mixture measured at 250°C using plate–plate configuration. PBT–epoxy mixtures were prepared by melt-blending for 35 min.

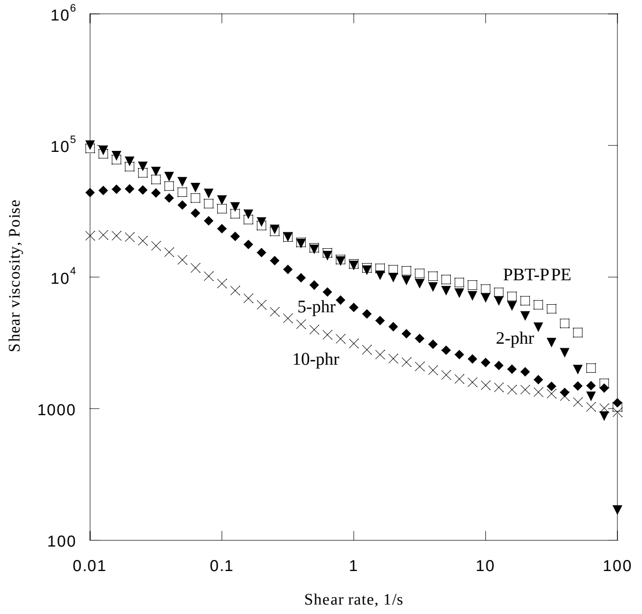


Fig. 6. Shear-viscosity of melt-blended (for 35 min) PBT–PPE–epoxy mixture measured at 250°C using plate–plate configuration. Mixtures contained 70 parts PBT, 30 parts PPE and 0, 2, 5, and 10-parts by weight epoxy (EPON® 828).

systems proceeded the same way as in PBT–PPE systems. However, PBT–epoxy copolymers produced stable morphology in the ternary system due to reduced coalescence as will be seen in Figs 8–13.

The values of mixing torque were found to be higher for three-component blends than PBT–PPE blends, when the blends contained 2 phr EPON® 828, 2- and 5 phr EPON® 832, and 2-, 5-, and 10 phr of EPON® SU-8 (Fig. 7). Increased torque values in the case of EPON® SU-8 are thought to be due to bulkiness of the chains of this octafunctional, novolac type epoxy. The trends for PPE-rich blends were similar although the torque values were much higher due to PPE continuous phase.

In all cases, the epoxy amounts were in excess of the equivalent weight ratios as presented in Table 1. Therefore, not all epoxy molecules were used in such reactions and the residual amounts of epoxy caused decline in the values of torque and shear viscosity, for example, in the case of 5 and 10 phr EPON® 828.

3.3. Blend morphology

The stability of dispersed phase domains of PBT-rich and PPE-rich blends was inferred from a comparison of SEM pictures taken before and after annealing at 250°C for 2 h. The cold fractured samples did not produce expected contrast between the phases in PBT-rich blends. We, therefore, resorted to solvent etching of PBT layers using HFIP–chloroform mixtures. As chloroform is also a good solvent for PPE, some dispersed PPE domains also dissolved during etching, leaving behind some holes. In the case of PPE-rich blends, the continuous PPE-phase was removed by Soxhlet extraction using chloroform and the dispersed phase domains of PBT were recovered using thimbles of ultra fine pore size. Representative SEM micrographs are shown in Figs. 8–13.

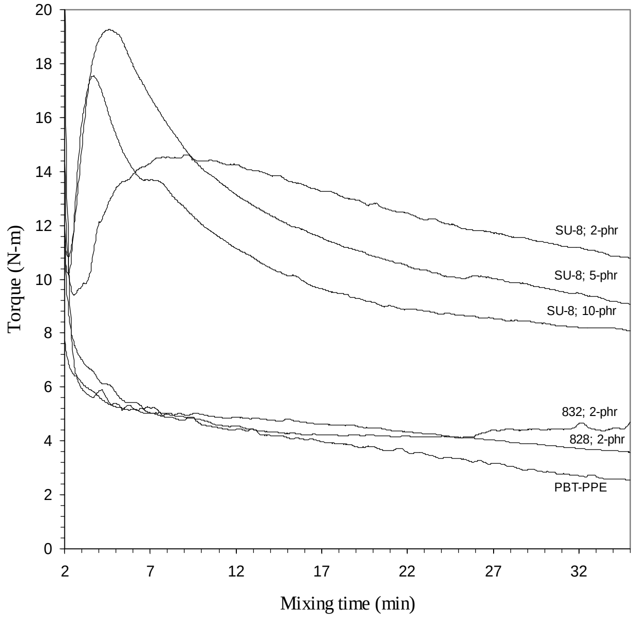


Fig. 7. Variation of torque with time during melt-blending of PBT, PPE, and epoxy mixtures. PBT–PPE = 70:30 parts by weight. Melt-blending was carried out at 250°C and 50 rpm. EPON® 828, 832, and SU-8 are represented in the legend as 828, 832, and SU-8 respectively. Epoxy amounts are given as 0, 2, 5, and 10 phr.

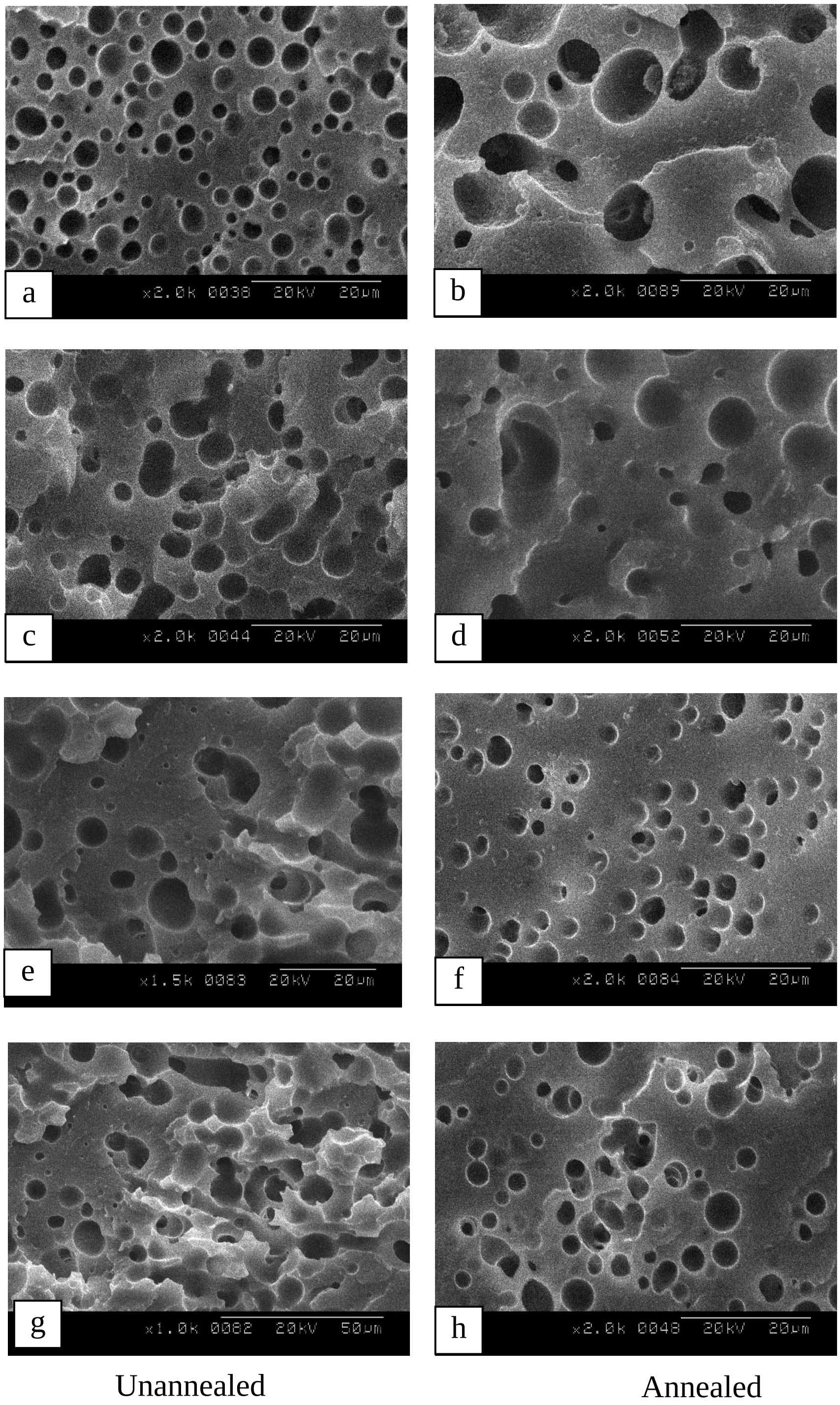


Fig. 8. SEM micrographs of PBT-rich blends with 70 parts PBT and 30 parts PPE. EPON® 828 was used as compatibilizer. Left and right columns represent, respectively, unannealed and annealed samples. (a)–(b): No epoxy, (c)–(d): 2 phr epoxy, (e)–(f): 5 phr epoxy, (g)–(h): 10 phr epoxy.

3.3.1. PBT-rich blends

In the absence of epoxy, typical dispersed PPE domains grew substantially upon annealing (Fig. 8a–b). The same was observed for blends containing 2 phr EPON® 828, Fig. 8(c–d). These observations imply that 2 phr EPON® 828 did not act as an effective compatibilizer. A better phase stability was observed with 5 and 10 phr of EPON® 828, Fig. 8(e–h). In view of the torque values in Fig. 3 and SEM pictures of Fig. 8, it can be intuitively argued that enough unreacted epoxide groups were not available in blends containing 2 phr EPON® 828 to interact with the PPE domains.

Blends containing EPON® 832 showed better phase stability to annealing than EPON® 828 (Fig. 9) and the smallest PPE domains were obtained for 2 phr epoxy.

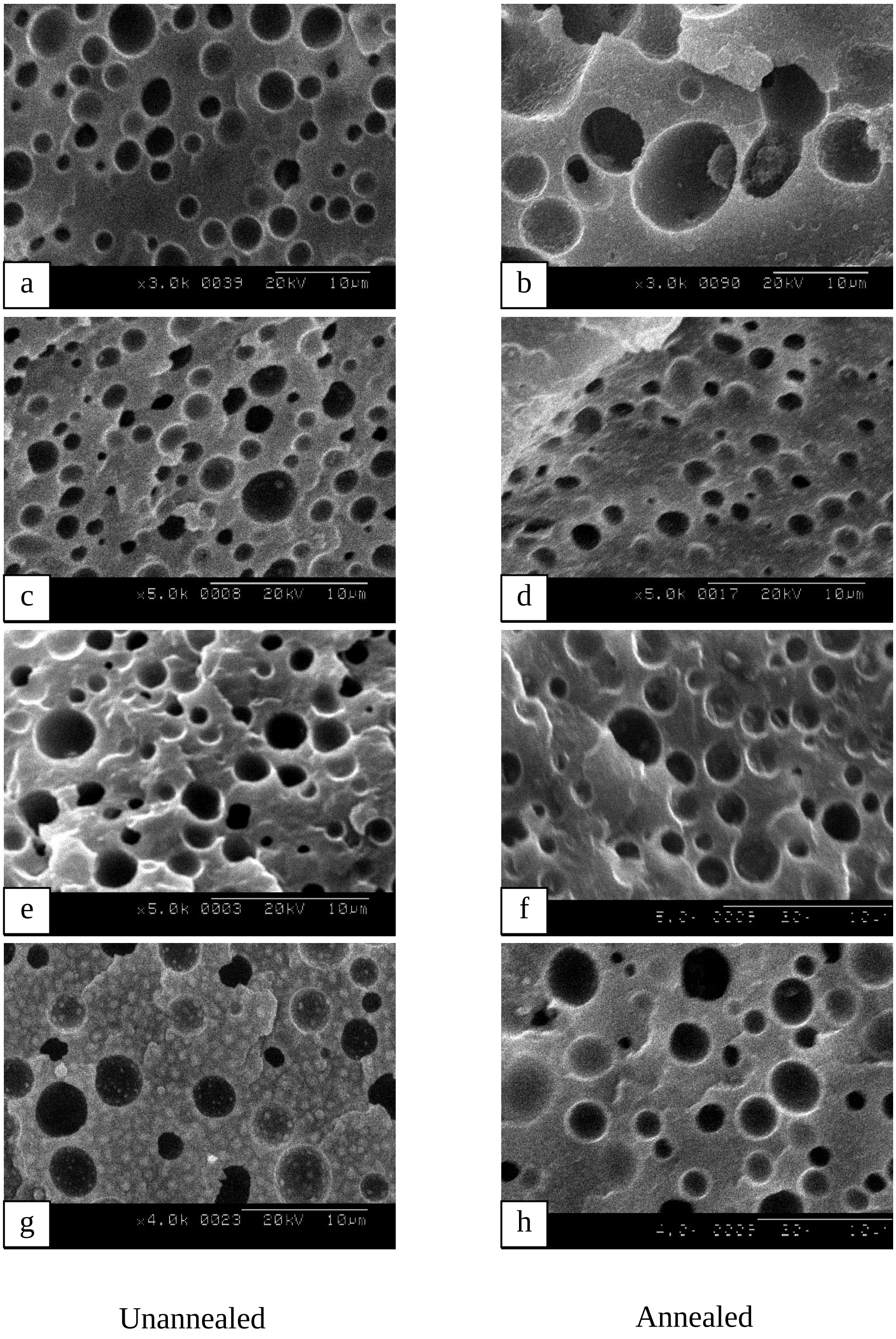


Fig. 9. SEM micrographs of PBT-rich blends with 70 parts PBT and 30 parts PPE. EPON® 832 was used as compatibilizer. Left and right columns represent respectively unannealed and annealed samples. (a)–(b): No epoxy, (c)–(d): 2 phr epoxy, (e)–(f): 5 phr epoxy, (g)–(h): 10 phr epoxy.

Such phase stability at low EPON® 832 content can be explained on the basis of reduced reactivity of epoxide groups due to bulkiness of the chains; longer chains of EPON® 832 — twice as long as EPON® 828 — made it less mobile to diffuse to the site of –COOH groups. Consequently, in a given reaction time, more unreacted epoxide groups remained in the case of EPON® 832 to interact with the PPE phase and provide phase stability.

The argument of epoxy mobility to the reaction site, i.e. to the site of –COOH groups can be further extended to explain phase stability against annealing in blends containing SU-8. Morphology was unstable to annealing for 2 and 5 phr SU-8 epoxy cases, although, some stability was observed in the case of 10 phr SU-8 (Fig. 10). Even though SU-8 and EPON® 828 have similar EEW, a typical chain of SU-8 is about four times bulkier than a chain of EPON®

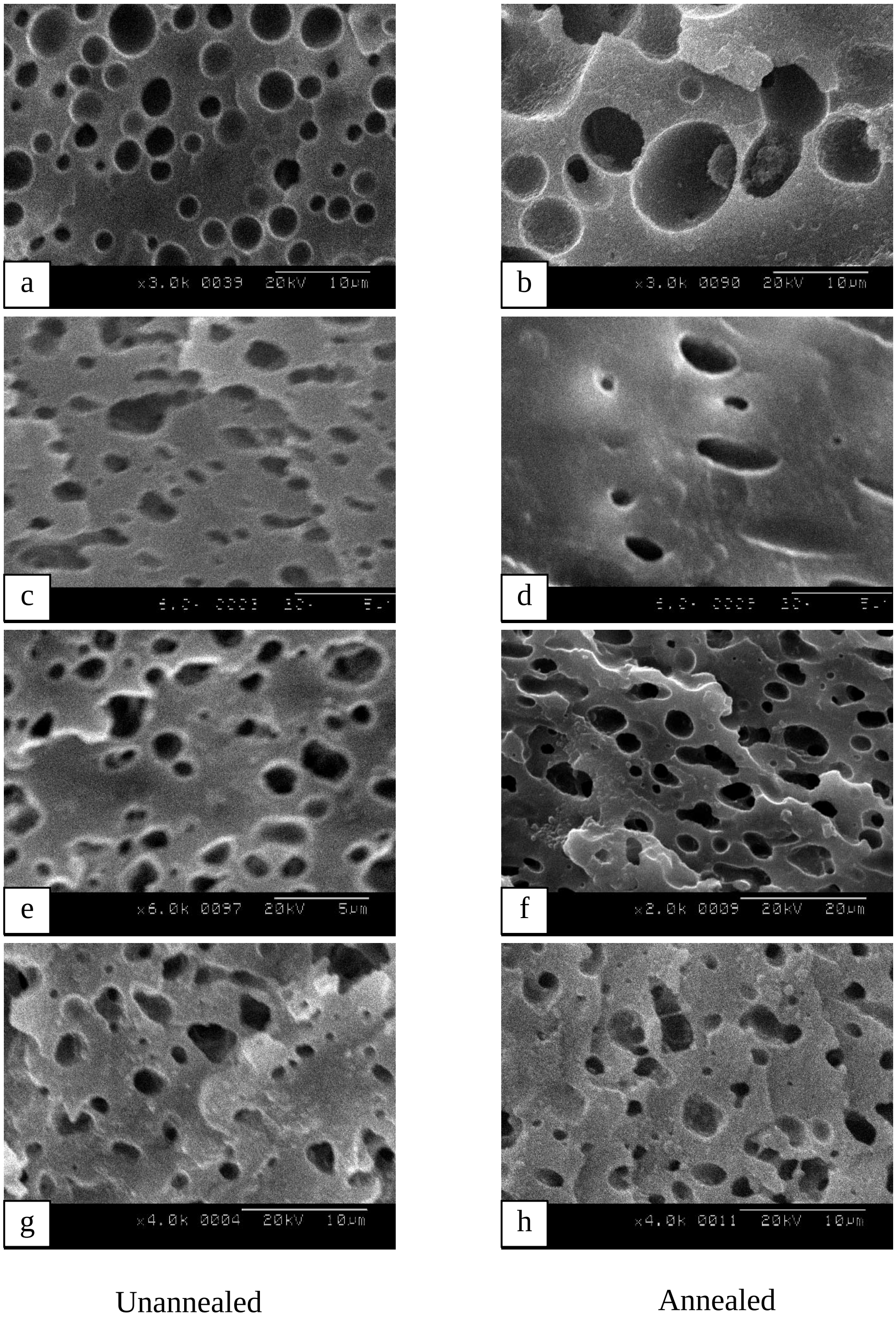


Fig. 10. SEM micrographs of PBT-rich blends with 70 parts PBT and 30 parts PPE. EPON® SU-8 was used as compatibilizer. Left and right columns represent, respectively, unannealed and annealed samples. (a)–(b): No epoxy, (c)–(d): 2 phr epoxy, (e)–(f): 5 phr epoxy, (g)–(h): 10 phr epoxy.

828, due to octa-functionality of the former. Therefore, the conversion of epoxide groups in the case of SU-8 can be expected to be smaller, leading to the formation of smaller number of PBT–epoxy copolymer molecules to act as compatibilizer.

3.3.2. *PPE-rich blends*

In PPE-rich blends, stable morphology was obtained only in the case of 10 phr EPON® 828 (see Fig. 11). It was observed earlier that EPON® 832 stabilized the morphology in PBT-rich blends (Fig. 9), although it failed to sustain phase stability in PPE-rich blends (Fig. 12). Unstable morphology was also obtained with SU-8, as is shown in Fig. 13. The loss of compatibilization in PPE-rich blends containing EPON® 832 presented an anomalous situation, an intuitive explanation of which is being offered here. The loss of compatibilization can be thought of either due to lower concentration of PBT–epoxy copolymers or due to

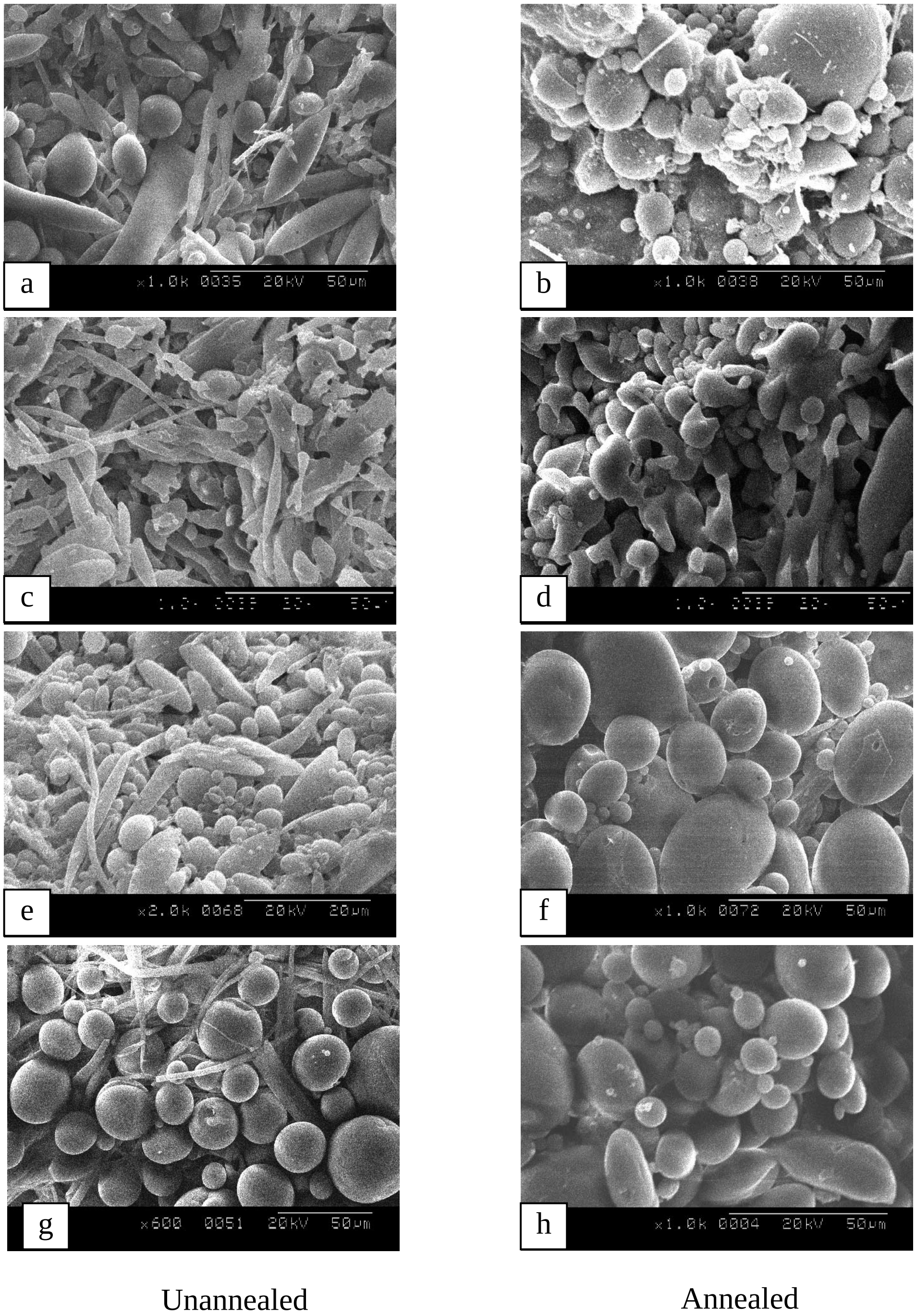


Fig. 11. SEM micrographs of PPE-rich blends with 30 parts PBT and 70 parts PPE. EPON® 828 was used as compatibilizer. Left and right columns represent, respectively, unannealed and annealed samples. (a)–(b): No epoxy, (c)–(d): 2 phr epoxy, (e)–(f): 5 phr epoxy, (g)–(h): 10 phr epoxy.

lower concentration of unreacted epoxide groups, the latter responsible for interactions with the PPE phase. As is apparent, these two effects oppose each other. A lower concentration of PBT–epoxy copolymer was a possibility in PPE-rich blends due to lower concentration of PBT itself. On the other hand, lower concentration of residual unreacted epoxide groups might be possible due to substantial reactions between –COOH end groups of PBT and epoxide groups as PBT: epoxy weight ratios in PPE-rich blends were much higher than in PBT-rich blends. A plausible explanation can, therefore, be based on greater reduction of concentration of epoxide groups in PPE-rich blends.

3.4. Tensile and impact properties

The test specimens for tensile and impact properties were prepared at 250°C by compression molding of melt-blended samples. The total time of compression molding was about

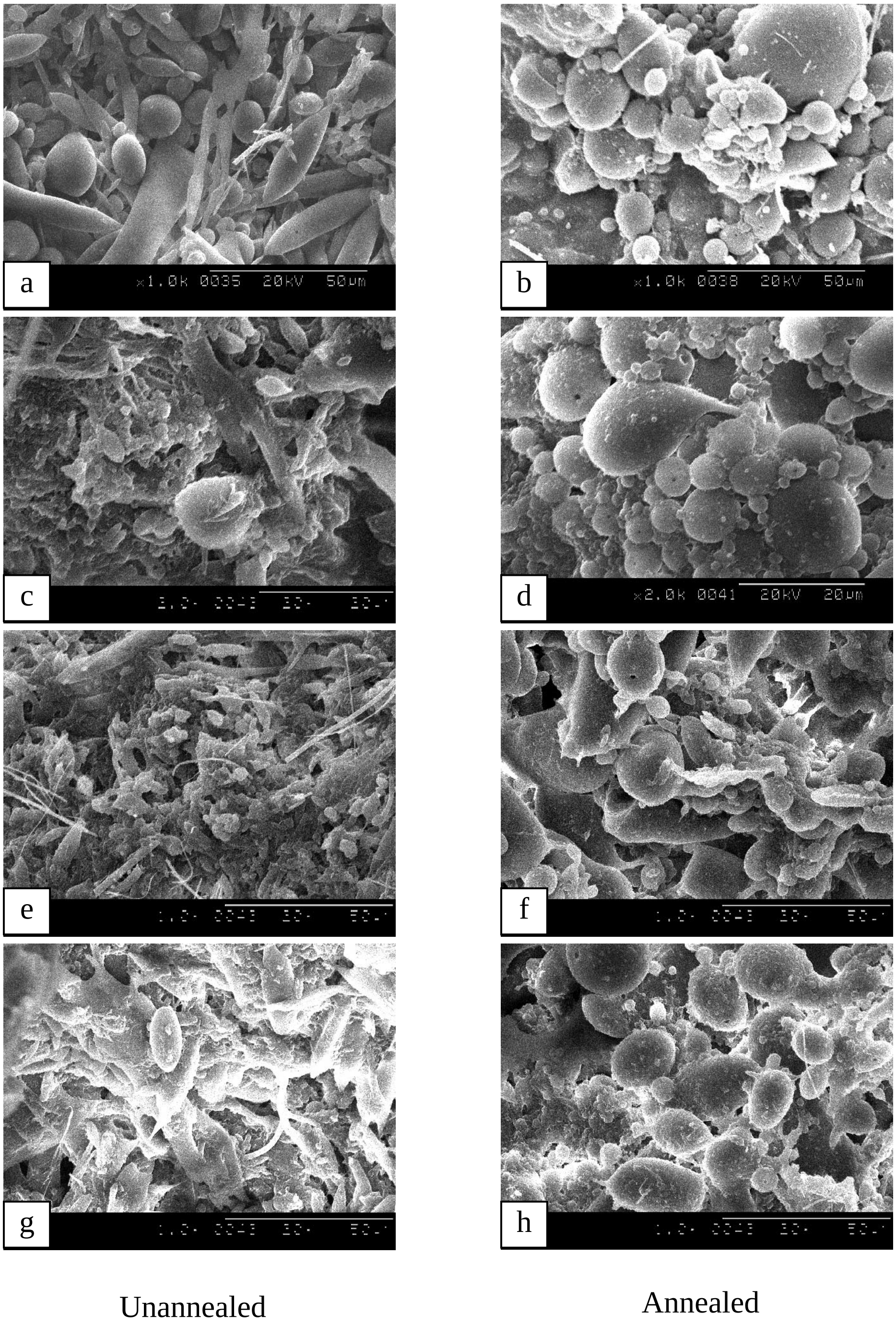


Fig. 12. SEM micrographs of PPE-rich blends with 30 parts PBT and 70 parts PPE. EPON® 832 was used as compatibilizer. Left and right columns represent, respectively, unannealed and annealed samples. (a)–(b): No epoxy, (c)–(d): 2 phr epoxy, (e)–(f): 5 phr epoxy, (g)–(h): 10 phr epoxy.

10 min. Some test specimens were annealed at 250°C for 2 h to study the effects of annealing on tensile and impact properties. As seen earlier, PPE-rich blends did not show phase stability, except with 10 phr of EPON® 828. Therefore, we report only the mechanical properties of PBT-rich blends. Also, the mechanical properties of PBT-rich blend with SU-8 were not evaluated as the blend was found to be unstable to annealing.

Figs 14 and 15 show the effect of epoxy content on tensile strength and tensile modulus. It is seen that annealing caused marginal weakening of modulus and strength, especially in the compatibilized blends. It is also shown in Fig. 14 that the tensile strength of uncompatibilized blend fell well below the corresponding strengths of the component polymers, indicating poor interfacial adhesion between the phases. EPON® 832 provided better interfacial adhesion than EPON® 828 as reflected in much improved values of tensile strengths in Fig. 14, although the modulus decreased

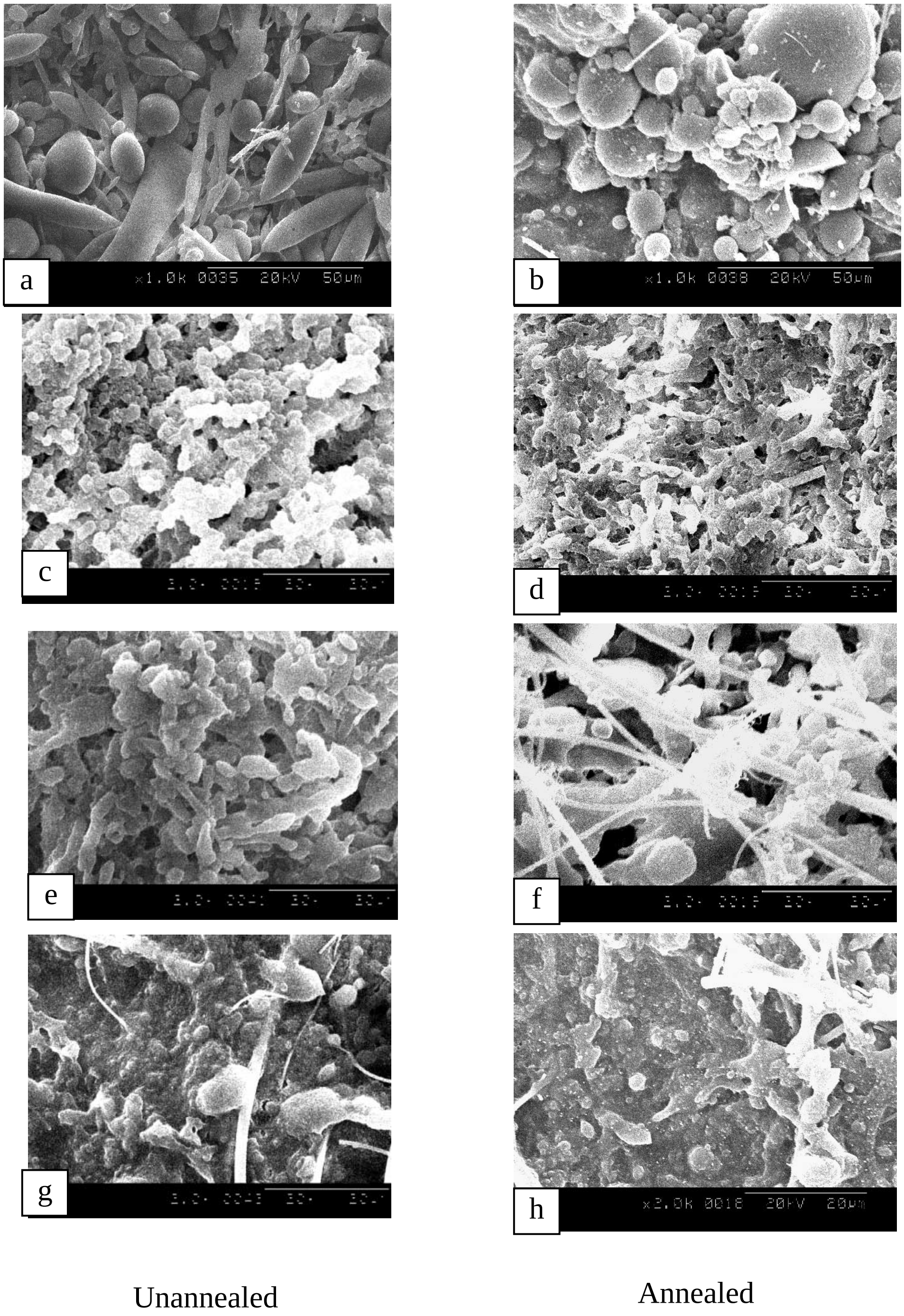


Fig. 13. SEM micrographs of PPE-rich blends with 30 parts PBT and 70 parts PPE. EPON® SU-8 was used as compatibilizer. Left and right columns represent, respectively, unannealed and annealed samples. (a)–(b): No epoxy, (c)–(d): 2 phr epoxy, (e)–(f): 5 phr epoxy, (g)–(h): 10 phr epoxy.

with epoxy content beyond 2 phr as shown in Fig. 15. This is probably due to longer, more flexible chains of EPON® 832 holding the interphase together.

Fig. 16 shows the variation of impact strengths with epoxy concentration in the blends. The impact strength of all blends containing EPON® 832 remained unaltered upon annealing. The impact strengths of PPE and uncompatibilized PPE–PBT blends can be considered nearly the same. In addition, the impact strength of blends with EPON® 828 can be considered unchanged from that of PBT–PPE blends. About four-fold increase in impact strength was, however, observed in blends containing 5 phr of EPON® 832. As mentioned previously, EPON® 832 has more flexible chains and its chain molecular weight is about twice as much of EPON® 828. Therefore, these flexible chains, residing at the interphase, hold the two phases together at higher loading by extending themselves. Lower impact strength at 10 phr EPON® 832 was due to the presence of

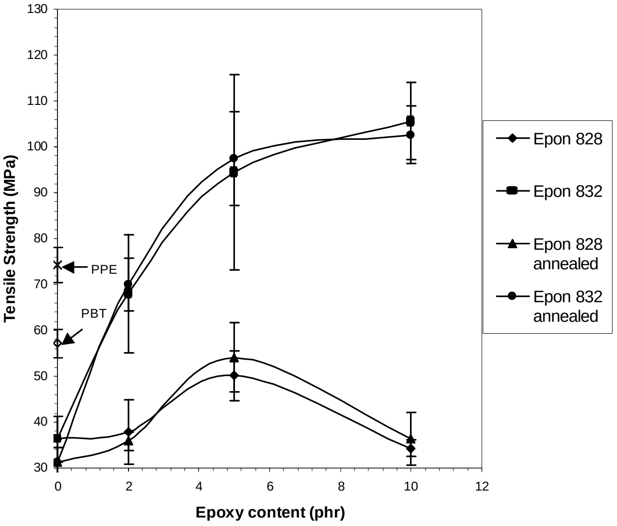


Fig. 14. Variation of tensile strength with epoxy content for PBT(70)–PPE(30) blends. Lines represent interpolation between experimental data points. The legends indicate the type of epoxy used.

higher amount of unreacted epoxy, which plasticized the phases, thereby weakened the inherent strength of the components.

4. Conclusions

Two bi-functional and one octa-functional epoxy were used in this study to compatibilize immiscible blends of PBT and PPE. Studies using DSC and FT-IR established reactions between –COOH groups of PBT and epoxide groups of epoxies. Increased torque values during blending of PBT–PPE–epoxy components also indicated PBT–epoxy reactions. The blend morphology and stability of morphology against annealing depended on epoxy content and the type of epoxy. It was found that a minimum of 2 phr of EPON® 832, 5 phr of EPON® 828, or 10 phr of EPON® SU-8 were required to effectively compatibilize the PBT-rich blends. The effects of compatibilization by epoxy were also reflected in tensile and impact strengths. The best improvements were obtained in blends containing EPON® 832, probably due to better chain flexibility of the molecules; a three-fold increase of tensile strength and a four-fold increase of impact strength over uncompatibilized blend were observed.

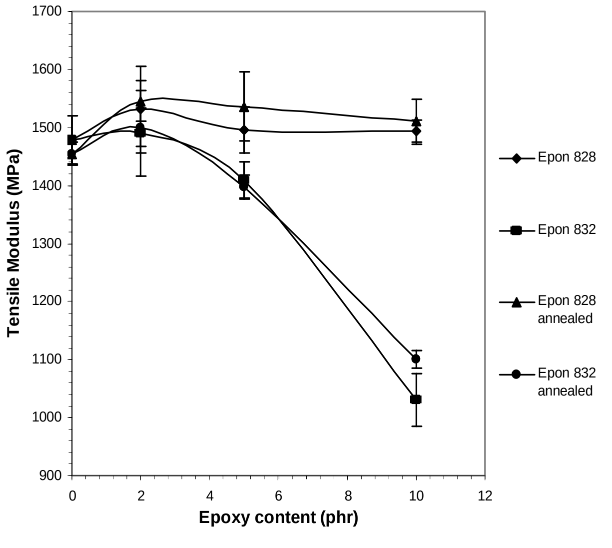


Fig. 15. Variation of tensile modulus with epoxy content for PBT(70)–PPE(30) blends. Lines represent interpolation between experimental data points. The legends indicate the type of epoxy used. Tensile modulus of PPE and PBT were found to be, respectively, 1700 and 1570 MPa.

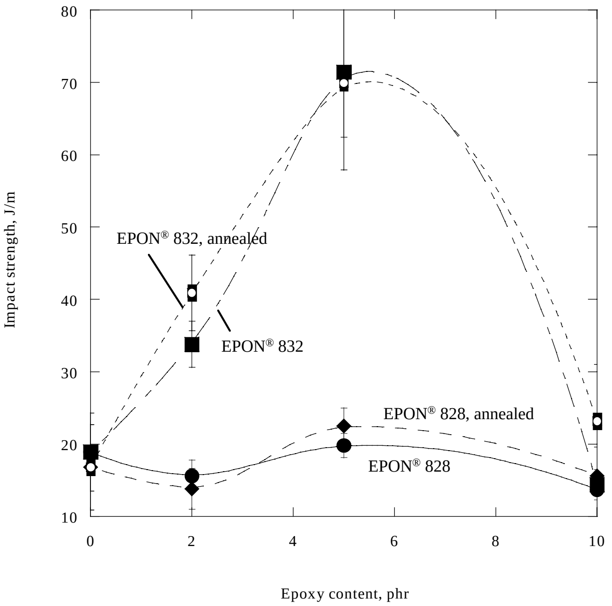


Fig. 16. Variation of impact strength with epoxy content for PBT(70)–PPE(30) blends. Lines represent interpolation between experimental data points. Impact strengths of neat PPE and PBT were measured to be 19.6 and 36.1 J m^{-1}, respectively.

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