

# Studies on Morphology and Mechanical Properties of PP/HIPS Blends from Postconsumer Plastic Waste

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**ABSTRACT:** The aim of this work was to study the compatibilizing effect of the triblock copolymer poly(styrene-*b*-ethylene-*co*-butylene-*b*-styrene) (SEBS) on the morphology and mechanical properties of virgin and recycled polypropylene/high-impact polystyrene (PP/HIPS) blends. The components of the blend were obtained from municipal plastics waste (MPW), with the PP obtained from blue mineral water bottles, symbolized as PP<sub>b</sub>, and the HIPS from disposable cups. These materials were preground, washed only with water, dried with hot air, and ground again (PP<sub>b</sub>) or agglutinated (HIPS). Blends of PP<sub>b</sub> and HIPS in three weight ratios (6:1, 6:2, and 6:3) were prepared, and three concentrations of SEBS (5.0, 6.0, and 6.7% w/w) were used for investigations of its compatibilizing effect. Scanning elec-

tron microscopy (SEM) showed that SEBS reduced the diameter of HIPS dispersed particles that were globular and fibril shaped, along with improving the adhesion between the dispersed phase and the matrix. On the other hand, SEBS interactions with PP<sub>b</sub> and HIPS influenced the mechanical properties of the compatibilized PP<sub>b</sub>/HIPS/SEBS blends. The optimal concentration of SEBS was 5 wt % for application to composite films with similar characteristics to synthetic paper. © 2002 Wiley Periodicals, Inc. *J Appl Polym Sci* 87: 747–751, 2003

**Key words:** recycling; polymer blends; polypropylene; high-impact polystyrene; compatibilizer

## INTRODUCTION

Over the last few years the volume of municipal plastics waste (MPW) has largely increased, creating a critical problem for both today's society and future generations. Any possible alternative solution must necessarily pass by improved techniques of recycling that would be directed at least toward adequately using these materials.<sup>1,2</sup>

A substantial part of MPW is composed of mixed polymers from unexpurgated monocomponents or laminates and from composite materials. The processing of plastics mixtures for recycling has been attempted with some success; however, poor mechanical properties and uncertain economic values can limit more versatile recovering.<sup>3,4</sup> Polyolefins, polyethylene terephthalate (PET), and polystyrene (PS) as well as HIPS are among the most common plastics waste because they are among the most frequently used commercial plastics in our daily lives as well as in industries.<sup>5</sup> Recycling mixed plastics residues as blends is attractive from both the academic and indus-

trial points of view because of enhanced impact strength, dimensional stability, stress cracking, and improved processibility, which has been studied only for virgin blends.<sup>6–9</sup>

The heterogeneous composition of plastics waste has been a technical problem. Without the addition of a compatibilizing agent, the properties of blends are usually inferior because of the lack of compatibility of the different polymers.<sup>4,9,10</sup> Compatibilizers must be added to the blends for them to have good mechanical characteristics, in particular, impact strength. Styrene-butadiene or styrene-ethylene-butylene block copolymers are usually used as compatibilizers.<sup>7,9,11</sup> In an immiscible polymer blend such as PP/PS<sup>6,7</sup> and PP/HIPS blends,<sup>9,11</sup> adhesion between the phases is very weak in most cases.

Horák et al. (1996) studied the compatibilizing of HIPS and PP blends with di-, tri-, and pentablock types of styrene/butadiene copolymers.<sup>11</sup> Multiblock copolymers showed greater improvement in impact strength and elongation at break in comparison with the diblock copolymer. Recently, Melo et al. (2000)<sup>9</sup> studied the effect on the mechanical and morphological properties of a PP/HIPS (70:30) blend of the concentration of styrene-*b*-ethylene-*co*-butylene-*b*-styrene copolymer (SEBS) used as a compatibilizer. They found that an admixture of the SEBS copolymer led to a decrease in the average size of the dispersed PS particles and that the best overall properties were obtained at a 5% content.

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Santana et al. (2001)<sup>12</sup> investigated the degradative effects in the reprocessing of PP/HIPS blends from postconsumer plastic waste selectively collected through flow properties. They showed that these blends presented melt flow index (MFI) values higher than their individual components, indicating a light thermal-mechanical degradation, probably because they were submitted to an additional extrusion process.

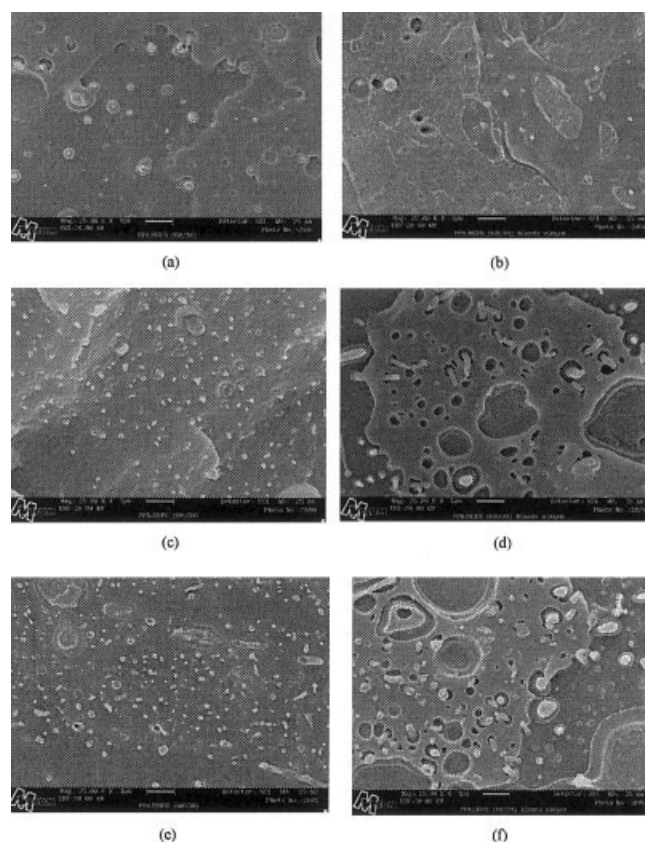
In this article we report on a study of the compatibilizing effects of SEBS in postconsumer and virgin PP/HIPS blends, whose aim was to find specific correlations between morphological and mechanical properties of those compatibilized blends.

## EXPERIMENTAL

The components of the virgin blend were polypropylene: H-603 (OPP), with an MFI of 1.5 g/10 min, for mineral-water-bottle and high-impact polystyrene (HIPS); and 4300 (BASF), with an MFI of 1.6 g/10 min.

The components of the recycled blend were obtained from selectively collected plastic waste in Federal University of São Carlos, São Paulo State, SP, Brazil. Polypropylene (with an average MFI of 2.0 g/10 min) came from 0.5-, 1-, and 3.5-L mineral water bottles (PP<sub>b</sub>), and high-impact polystyrene (HIPS), with an average MFI of 2.3 g/10 min, from disposable cups. These materials were preground (average size ~ 10 cm × 5 cm) in a knife mill (Kie model MAK 250), washed only in water, and dried with hot air to 50°C<sup>13-15</sup> in a centrifuge system of washing/drying.<sup>16</sup> Then the PP<sub>b</sub> was ground again to flakes and the HIPS agglutinated (Lombard model 162).<sup>15</sup> PP<sub>b</sub>/HIPS blends were prepared in three weight ratios—6:1; 6:2, and 6:3—in the presence of, respectively, 5%, 6%, and 7% of the styrene-*b*-ethylene-*co*-butylene-*b*-styrene triblock copolymer Kraton G (Shell Chemical Co., Ohio), used as a compatibilizer agent. Processing the mixture of the PP<sub>b</sub>/HIPS blends was done in a single-screw Gerst extruder (model 25 × 24D) at 130 rpm and with temperature profile of 180°C, 190°C, and 200°C in the three sections.

A scanning electron microscope (SEM) LO model Stereoscan 440 was used for studying the morphology. Samples were fractured in liquid nitrogen and covered with gold before being examined under a microscope at an acceleration voltage of 20 kV. Specimens for the mechanical test were prepared in a PIC BOY injection-molding machine, model Boy 15.<sup>17,18</sup> Flexural and tensile (type I) tests were carried out with an Instron machine, model 1122. For the impact test notched Izod impact strength was measured with an impact-testing machine (CEAST, model Resil 25 P/N 6545.500) at room temperature based on the ASTM D256-93 standard; and variations in HDT were obtained in an apparatus for the deflection temperature test (CEAST,



**Figure 1** SEM micrographs of recycled and virgin PP/HIPS blends, respectively: B1 (6:1), B2 (6:2), and B3 (6:3) compatibilized with SEBS: (a and b) 7 wt % B1, (c and d) 6 wt % B2, and (e and f) 5 wt % B3, magnified 25,000 times.

model HDT 6 VIC) based on the ASTM 648-96 standard.

## RESULTS AND DISCUSSION

### Morphology study

SEM micrographs of binary PP/HIPS blends (6:1) containing 7 wt % of the SEBS compatibilizer are shown in Figure 1(a,b), for the recycled and virgin materials, respectively. First, we observed that the concentration of HIPS particles (dispersed phase) was low and that the presence of some holes as well as some pulled-out particles with droplet and thread forms originated by the stretching in the fracture. The presence of SEBS in excess<sup>9,11</sup> can also be observed in Figure 1(a). The compatibilizer is on the interface of PP and HIPS phases, which is more evident in Figure 1(a) than in Figure 1(b). A large size distribution is shown in both micrographs, with the particle dispersions of the virgin blends higher than those of the recycled blends.

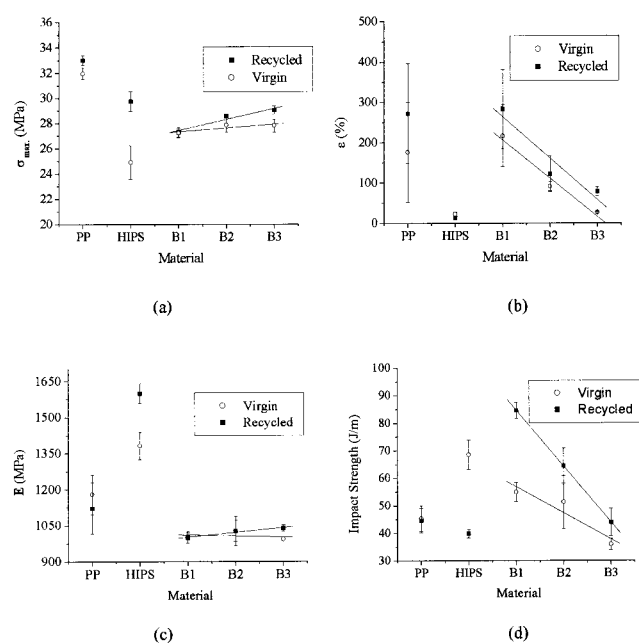
Figure 1(c,d) shows micrographs of fractured surfaces of PP/HIPS blends (6:2) containing 6 wt % of the SEBS compatibilizer. The size of the dispersed HIPS particles is reduced with a small decrease in the SEBS

triblock copolymer in both blends. The size reductions as well as the better adherence of the dispersed phase in the matrix especially are especially evident in the recycled blend rather than the virgin blend. The higher average interfacial area of the HIPS particles (diameters of submicron sizes 0.08–0.15  $\mu\text{m}$ ) is the reason for the more efficient interfacial activity of SEBS at this level of concentration. However, Figure 1(d) shows that the virgin blend had a higher concentration of holes and particles of HIPS being pulled by the stretching in the fracture. The weak adherence of virgin blends could be a result of the grade of HIPS and/or the conditions for the processing of the blend mixtures.

In relation to the grade of HIPS (size and composition of styrene–butadiene S-B block copolymer), the incompatibility of polystyrene (PS) and polybutadiene (PB) is known, and a phase separation occurs shortly after the beginning of the polymerization leading to a typical polymeric oil/oil emulsion, with (PS + styrene) as the dispersed phase and (PB + styrene) as the continuous medium, which usually inverts during the course of polymerization. The final product, after complete polymerization of styrene, consists of a continuous elastomer phase, generally of spherical shape with typical occlusions of PS particles. There are typical morphologies using styrene–butadiene block copolymer of the styrene-butadiene-styrene (SBS) and butadiene-styrene-butadiene (BSB) types of various compositions. Onion skin (final composition: 75 wt % PS, 12.5 wt % S-B block copolymers with 48 wt % PS,  $M_n = 225,000$ ; 12.5% S-B block copolymer with 52 wt % PS,  $M_n = 78,000$ ) and core-shell (styrene in the presence of 10 wt % S-B copolymer; PS content of block copolymer 48 wt %,  $M_n = 225,000$ ) morphologies are known, in particular the last, and have already gained commercial importance because these types of HIPS provide toughness and transparency, which is not the case for classical elastomer-modified PS.<sup>19</sup> Therefore, in this work the virgin blends presented a morphology [Fig. 2(d,f)] similar to the onion and the recycled blend to the core-shell form [Fig. 2(c,e)].

On the other hand, Fortelný et al. (1996) studied the mixture processing condition of the blends and concluded that the particle size decreases with an increasing rate of mixing for the blends with a low content of the dispersed phase. This conclusion was followed by an analysis of theoretical expressions for particle size in a steady flow.<sup>20</sup> They showed that increasing viscosity of the PP matrix influences a decreasing of particle size of the dispersed phase.<sup>6</sup>

Recycled HIPS/PP blends B3 (6:3) and virgin with SEBS (5 wt %) showed similar characteristics of dispersion and adherence with the HIPS/PP blend (6:2), as shown in Figure 1(e,f). Good adherence of the dispersed phase in the matrix of the recycled blend was



**Figure 2** Mechanical properties: (a) tensile stress at maximum load,  $\sigma$ ; (b) tensile strain at break (elongation),  $\epsilon$ ; (c) tensile modulus,  $E$ ; and (d) impact strength.

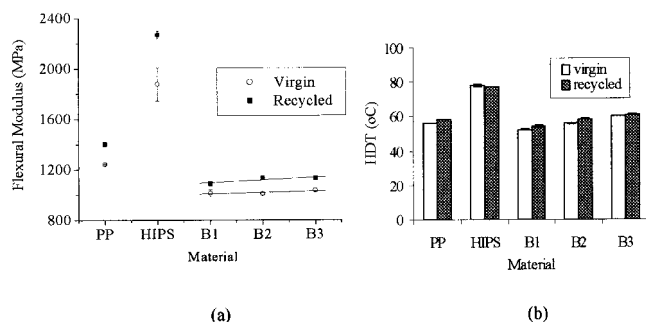
observed, and, again, the virgin blend showed weak adherence of HIPS particles in the matrix.

Despite the presence of some holes in the fracture surface in the recycled blends, these can be considered negligible. The B3 blend had a higher concentration of HIPS and a lower concentration of SEBS than the B2 blend. The dispersed-phase size of the B3 was reduced one more time, with average diameters of submicron sizes 0.04–0.12  $\mu\text{m}$ , leading to a higher average interfacial area of the HIPS particles.

### Mechanical properties

Figure 2 shows the mechanical properties of virgin and recycled PP and HIPS and of their binary PP/HIPS blends compatibilized with SEBS at 7 (B1), 6 (B2), and 5 (B3) wt %. Recycled PP presented higher tensile stress at maximum load (a) and elongation (tension strain) at break (b) than virgin PP, as well as with recycled and virgin HIPS. On the other hand, recycled HIPS presented higher tensile strength, tensile modulus, and impact strength (d) than did virgin HIPS H4300. The result can be attributed to the different grades of those polymers, that is, the virgin blend is a specific grade from BASF, whereas the recycled blend is comprised of a mixture of grades from various resin suppliers.

In relation to the blends, the optimization of the SEBS concentration (decreasing from 7 to 5 wt %) improved the interfacial adhesion (previously analyzed by morphological studies), and thus tensile stress would be increased, as shown in Figure 2(a).



**Figure 3** Thermal-mechanical properties: (a) flexural modulus,  $M$ , and (b) deflection temperature under flexural load.

Also, the recycled blends demonstrated slightly higher tensile stress and tensile modulus [Fig 2(a–c)] than did the virgin blends. But, on the other hand, the impact strength and elongation at break properties increased even after exceeding optimal concentration (5%). The reason for this is that the triblock copolymer preferentially occupies the interfacial area, forming a certain type of bond between the phases and thus increasing their adhesion.

The result of this process was an improvement in the mechanical properties. After exceeding the critical concentration of the compatibilizer, the excessive copolymer dispersed in the bulk polymers contributed further to an enhancement of these mechanical characteristics.<sup>11</sup> Meanwhile, it should be noted that the impact strength was much improved by the best adherence and compatibilization of the dispersed phase on the matrix. In this case, it was observed, as shown in Figure 2(d), that the recycled blends had better impact strength than the virgin blends did. However, a decrease in the impact strength with an increase in the HIPS concentration in the blend was observed. This behavior can be explained by the results of the high tensile modulus [Figure 2(c)] and low elongation [Figure 2(b)] of HIPS.

As can be noted in Figure 2(a–d), all the mechanical properties of the virgin blends were lower than those of the recycled blends. This could be explained by the probable difference in distribution and concentration of each styrene and butadiene block in the copolymer<sup>19</sup> because virgin and recycled HIPS have different origins. The weak adherence of the dispersed phase (HIPS) in the matrix (PP) for the virgin material, shown by phase morphology analysis [Fig. 1(c,d) for the recycled and virgin blends, respectively] could explain these differences in mechanical properties.

Figure 3 shows the mechanical properties of PP and HIPS virgin and recycled blends and of their binary PP/HIPS blends compatibilized with SEBS at 7 (B1), 6 (B2), and 5 (B3) wt %. HIPS showed a higher flexural modulus [Fig. 3(a)] and HDT [Fig. 3(b)] than PP did. Also recycled PP, HIPS, and their blends showed a higher flexural modulus and HDT than the virgin blend.

Observe that flexural modulus ( $M$ ) and HDT values fell after exceeding a limiting compatibilizer concentration, which confirms the tensile stress ( $\sigma$ ) and tensile modulus ( $E$ ) results analyzed previously [Fig. 2(a,c)]. Therefore, there are some properties for which an excessive triblock copolymer concentration brought about observable changes in the property–concentration dependence.<sup>9,11</sup> Finally, we can conclude that knowledge of critical compatibilizer concentration is important for the development of multiphase polymer materials with well-balanced properties according to specific requirements.<sup>11</sup>

A better visualization of the thermal-mechanical properties is shown in Table I, where the B3 blend shows an increase in tensile stress ( $\sigma_{\max}$ ), tensile modulus ( $E$ ), flexural modulus, and HDT properties and a decrease in tensile strain at break ( $\epsilon$ ) and impact strength (RI) properties with an increase of the HIPS concentration and an decrease of the excessive concentration of SEBS. Despite a decreasing RI of B3 in rela-

**TABLE I**  
Thermal-Mechanical Properties of PP, HIPS, and Their Recycled and Virgin Blends

Material PP/HIPS	$\sigma_{\max}$ (MPa)	$\epsilon$ (%)	$E$ (MPa)	$M$ (MPa)	RI (J/m)	HDT (°C)
PP <sub>b</sub>	33,0	272,2	1122	1404	44,6	58,3
PP <sub>v</sub>	31,9	176,1	1179	1243	45,3	56,1
HIPS <sub>r</sub>	29,8	13,6	1599	2271	39,9	77,3
HIPS <sub>v</sub>	24,9	22,3	1382	1879	68,5	78,2
B1 <sub>r</sub> (6:1) with 7% SEBS	27,3	283,6	998	1090	84,6	54,3
B1 <sub>v</sub> (6:1) with 7% SEBS	27,2	216,5	1009	1014	54,9	52,3
B2 <sub>r</sub> (6:2) with 6% SEBS	28,5	121,9	1027	1137	64,5	58,4
B2 <sub>v</sub> (6:2) with 6% SEBS	27,9	90,9	1027	1009	51,3	55,9
B3 <sub>r</sub> (6:3) with 5% SEBS	29,1	77,8	1039	1132	43,9	61,3
B3 <sub>v</sub> (6:3) with 5% SEBS	27,8	26,3	993	1036	35,9	53,4

v: virgin, r: recycled

tion to the others blends, this resulted in higher recycled HIPS, similar to the recycled PP value.

### CONCLUSIONS

This study compared the compatibilization efficiency of SEBS copolymer as well as HIPS content in virgin and recycled PP/HIPS blends with high rigidity and better mechanical properties because this blend will be used as raw material for preparing composite films with characteristics similar to synthetic paper.

Morphological studies provided evidence for the interfacial activity of block copolymers attributable to a stronger interaction of SEBS between the dispersed phase and the matrix, reducing average particle size of the HIPS and increasing interfacial adhesion between both phases.

The B3 recycled blend PP/HIPS (6:3) with 5% wt of SEBS presented the highest reduction in size, with average diameters of submicron sizes 0.04–0.12  $\mu\text{m}$ , leading to a higher average interfacial area of the HIPS particles.

An excessive triblock copolymer concentration brought about observable changes in the property–concentration dependence; therefore, critical compatibilizer concentration is important for the development of multiphase polymer materials with well-balanced properties according to specific requirements.

In this work the optimal concentration of SEBS in PP/HIPS blend (6:3) was found to be 5 wt % based on an improvement in the majority of their thermal-mechanical properties.

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### References

- Eyer, C. V. *Qualidade Ambiental*; ED. ABDR: SP-Brazil, 1995.
- Judd, A. H. In *Defense of Garbage*; Greenwood Publishign: Westport, CT, 1993.
- Ehrig, R. J. *Plastics Recycling*; Hanser: New York, 1992.
- Park, H. D.; Park, K. P.; Cho, W. J.; Há, C. S.; Kwon, S. K. *Polym Recycling* 1996, 2, 277.
- Instituto de Pesquisas Tecnológicas. "Lixo Municipal—manual de Gerenciamento Integrado," Publication IPT 2163, SP, Brazil, August 1996,; p 172.
- Fortelný I, Michalková, D.; Mikesová, J. *J Appl Polym Sci* 1996, 59, 155.
- Radonjic, M. V.; Smit, I. *J Appl Polym Sci* 1998, 69, 2625.
- Há, C. S.; Park, H. D.; Cho, W. J. *J Appl Polym Sci* 2000, 76, 1048.
- Mélo, T.; Carvalho, L.; Calumby, R.; Brito, K.; D'Almeida, J.; Spieth, E. *Polímeros: Ciencia e Tecnologia* 2000, 10(2), 82.
- Fortelný, D., Michalková, D.; Hromadková, J.; Lednicky, F. *J Appl Polym Sci* 2001, 81, 570.
- Horák, Z.; Fort, V.; Hlavatá, D.; Lednicky, F.; Vecerka, F. *Polymer* 1996, 37(1), 65.
- Campomanes, R. S.; Manrich, S. PP/HIPS blends from post-consumer plastic waste selectively collect In 6° Congresso Brasileiro de Polímeros—ABPol, and IXth International Macromolecular Colloquium; Gramado/RS, Brazil, November 2001.
- Trevizan, D.; Campomanes, R. S.; Manrich, S. Otimização dos Parâmetros do Sistema de lavagem/Secagem dos copos de HIPS Provenientes da Coleta Seletiva do Campus, VII Congress of Scientific Initiation—CIC, UFSCar/São Carlos,SP, Brazil, August 23–25, 2000, Anais TE450.
- Trevizan, D.; Campomanes, R. S.; Manrich, S. Estudo da Influencia do Tamanho dos Flakes de HIPS No Processo de Lavagem-Secagem, VIII Jornadas de Jovens Pesquisadores do Grupo de Montevideo; UFSCar/São Carlos, SP, Brazil, September, 28–30, 2000, Anais EM-30.
- Campomanes, R. S.; Trevizan, D.; Manrich, S. Avaliação da Reciclabilidade de HIPS Pós-Consumo Coletado Seletivamente, IX International Macromolecular Colloquium and VI Congresso Brasileiro de Polímeros; Gramado/RS, Brazil, November 11–15, 2001.
- Manrich, S.; Herrera, J. C.; Rossalini, A. C.; Acconci, C. Depósito de Patente, MU7901780, July, 16, 1999.
- American Society of Testing Materials. Standard test method for tensile properties of plastics/ D638–90, Philadelphia, PA, 1991, 8(1), 157.
- American Society of Testing Materials. Standard Test Method for Flexural Properties of Non-reinforced and Reinforced Plastics and Electrical Insulating Materials, D790–90, Philadelphia, USA 1991, 8(1), 272.
- Legge, N.; Holden, G.; Schroeder, H. In *Thermoplastic Elastomer*; Riess, G., Hanser Publishers: New York, 1987; Chapter 12.
- Fortelný, I.; Cerná, Z.; Binko, J.; Kovár, J. *J Appl Polym Sci* 1993, 48, 1731.