Effect of Talc on the Properties of Polypropylene/Ethylene/Propylene/Diene Terpolymer Blends

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ABSTRACT: In the present study, the effect of talc content on the mechanical, thermal, and microstructural properties of the isotactic polypropylene (i-PP) and elastomeric ethylene/propylene/diene terpolymer (EPDM) blends were investigated. In the experimental study, five different talc concentrations, 3, 6, 9, 12, and 15 wt %, were added to i-PP/EPDM (88/12) blends to produce ternary composites. The mechanical properties such as yield and tensile strengths, elongation at break, elasticity modulus, izod impact strength for notch tip radius of 1 mm, and hardness with and without heat treatments and thermal properties, such as melt flow index (MFI), of the ternary composites have been investigated. The annealing heat treatment was carried out at 100°C for holding time of 75 h. From the tensile test results, an increased trend for the yield and tensile strengths and elasticity modulus was seen for lower talc contents, while elongation at break showed a sharp decrease with the addition of talc. In the case of MFI, talc addition decreased the MFI of i-PP/EPDM blends. It was concluded that, taking into consideration, mechanical properties and annealing heat treatment, heat treatment has much more effect on higher yield and tensile strengths, elongation at break, elasticity modulus, impact strength, and hardness. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 3033–3039, 2006

Key words: poly(propylene) (PP); talc; EPDM; mechanical properties; heat treatment

INTRODUCTION

Particulate filled polymers have been a subject of increasing interest in both industry and research. Polypropylene (PP) has a good processability allowing to accept numerous types of natural and synthetic fillers. The incorporation of fillers such as kaolin, mica, talc, and calcium carbonate into thermoplastics is a common practice to reduce production costs and to improve the properties such as strength, stiffness, hardness, flexural modules, dimensional stability, crystallinity, electrical, and thermal conductivity. However, high filler additions may adversely affect the processability, ductility, and strength of composites. The properties of the composites strongly depend on the interfacial adhesion between filler and matrix. In case the adhesion is strong at the interface of filler and matrix, the mechanical properties may increase.[1–10]

Although PP has been used widely in a variety of application because of its advantages, such as low density, low cost, high softening points, and easy processing, it has low impact strength especially at low temperatures and this limits its application as an engineering thermoplastic. To improve the impact strength, various elastomers, such as ethylene–propylene–copolymer, ethylene–propylene–diene terpolymer (EPDM), and ethylene-vinyl acetate, were blended with PP. Among these elastomers, EPDM is considered as one of the most effective impact modifiers for PP. Although toughness strength is improved by blending elastomer, this results in deteriorating tensile strength and modulus. Therefore, numerous studies have been performed to improve the toughness, stiffness, and strength balance. Therefore, PP has been modified by different fillers and elastomers. The mechanical properties of such ternary composites strongly depend on their composition and characteristic of the components and the phase morphology, and in particular relative dispersion of additive components. In the ternary composites containing both elastomer and rigid fillers, two different types in phase structure may occur, where elastomer and filler particles are dispersed separately on the PP matrix or the rubber encapsulates fillers particles, resulting in a low modulus interlayer between matrix and filler.[11–21]

In the literature, there are some published studies relating to PP/elastomer/filler ternary composites.[11–20] However, the effect of talc with heat treat-

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TABLE I
Characteristic Properties of i-PP, EPDM, Talc, and Additives Used to Produce Ternary Composites

Material	Trade name	Supplier	MFI	Money visco.	d (g/cm^3)	T_m (°C)	Shape
i-PP	Petoplen MH 418	Pektim Turkey	4–6[a] (g/10 min)	—	0.925	166	Pellet
EPDM	IP 4770	Nordel	—	70[b]	—	—	Pellet
Talc	Zeta talc EW 20	Eral Turkey	—	—	2.7	—	Powder
Stabilizer	Irganox B 225	Ciba Switzerland	—	—	530–630 (g/lb[c])	—	Powder
PE wax		Eral Turkey	—	—	—	—	Powder

MFI, melt flow index; d, density; T_m, melting temperature.
[a] 230°C, 2.16 kg/10 min.
[b] Mooney typical viscosity, ML 1+4 at 125°C.
[c] Bulk density.

ment on the properties of isotactic polypropylene (i-PP)/EPDM composite have not been, to our knowledge, studied. Therefore, in the present study, i-PP/EPDM/Talc composites, containing 3, 6, 9, 12, and 15 wt % talc, have been produced and the effect of talc content on the mechanical, with heat treatment, thermal, and microstructural properties were investigated.

EXPERIMENTAL

Materials

Isotactic polypropylene (i-PP), ethylene-propylene-diene monomer (EPDM), filler (talc), and additives used in this study are all commercial products and their main characteristic properties are listed in Table I. Talc (Zeta talc EW 20) in the particle size of around 2 μm was used as a filler material. The compositions of blends to produce i-PP/EPDM/Talc ternary composites are shown in Table II. Irganox B 225 (0.3 wt %) and 0.4 wt % of PE wax were added as synergistic processing and long-term thermal stabilizer to i-PP/EPDM/Talc composites during melting processing. The materials and blends were dried at 75°C at least 1 h in an oven prior to compounding.

Compounding process and specimen preparation

i-PP/EPDM/Talc blends were prepared by using single-screw extruder (Microsan MTV). The processing parameter used in extrusion of blends is listed in Table III. The extrudate was frozen in-line in a water bath (~15°C) pelletized and dried in oven at 105°C in 2 h. Tensile and impact test specimens (ISO 527.2 and 180) were prepared by injection molding on a 70 ton machine and processing parameters listed in Table IV. Prior to molding, the pelletized were dried at 105°C for 2 h.

Measurement procedures

Mechanical testing

The effect of talc on the i-PP/EPDM blends were evaluated through the mechanical properties such as the yield and tensile strengths, elongation at break, elasticity modulus, Izod impact resistance, and hardness (Shore D). The tensile tests of the blends were carried out on a Zwick Z010 tensile test machine according to ISO 527.2 test procedure with a crosshead speed of 50 mm/min at room temperature. A computer was connected to the Zwick load cell and data acquisition program recorded the force measured by the load cell. At least seven specimens were tested for each blend and average values were calculated. Izod impact strength values of the composites were evaluated with a Zwick impact test instrument according to the ISO 180 test procedure at room temperature. Izod impact tests were performed for notch tip radius of 1 mm. Seven impact test specimens were tested and average values reported for each blends. Prior to the

TABLE II
Compositions of Blends to Produce i-PP/EPDM/Talc Ternary Composites (wt %)

Materials		
i-PP	EPDM	Talc
88.00	12	0
85.36	11.64	3
82.72	11.28	6
80.08	10.92	9
77.44	10.56	12
74.8	10.20	15

TABLE III
Processing Parameters in Extrusion Used to Produce i-PP/EPDM/Talc Composites

Parameters	Values
Screw diameter (D, mm)	30
Length-to-diameter ratio (L/D)	25
Screw speed (rpm)	45
Extrusion pressure (bar)	5
Extrusion temperature of profiles from feed zone to the die (°C)	210–225

TABLE IV
Processing Parameters in Injection Molding Used to Produce i-PP/EPDM/Talc Composites

Parameters	Values
Injection pressure (bar)	500
Injection pressure time (s)	5
Mold temperature (°C)	40
Dwell time in the mold (s)	10
Temperature range (°C)	210–230

mechanical tests, all specimens were kept at room temperature at least 72 h. Hardness of the i-PP/EPDM/Talc composites were determined with a Zwick hardness test machine according to the ISO 868 test method at room temperature. The values of the hardness were evaluated as averages over seven specimens for each composition of the blend.

Microstructural characterization and thermal analysis process

The fracture surfaces of impact test specimens have been evaluated by using scanning electron microscope (SEM) (JSM 5910LV) at 15 kV. The surfaces of the specimens were coated with gold thin film (25 Å) for good conductivity. The melt flow indexes (MFI) of i-PP/EPDM/Talc composites were determined by a Zwick 4100 MFI test instrument according to the procedure ISO 1133 method. Annealing heat treatment of the i-PP/EPDM/Talc was carried out in a hot-air oven (Binder 115) at 100°C for holding time of 75 h. These conditions were chosen to accelerate the aging process without melting the polymers.

RESULTS AND DISCUSSION

Mechanical properties

The effect of talc content on the yield and tensile strengths, elasticity modulus, and elongation at break are shown in Figure 1(a–d), respectively. From the Figure 1(a–b), in general, it can be seen that the talc addition for lower contents showed a slight increase on the yield and tensile strengths of the i-PP/EPDM composite. From the figure, the effect of talc on the yield and tensile strengths can be assessed in two

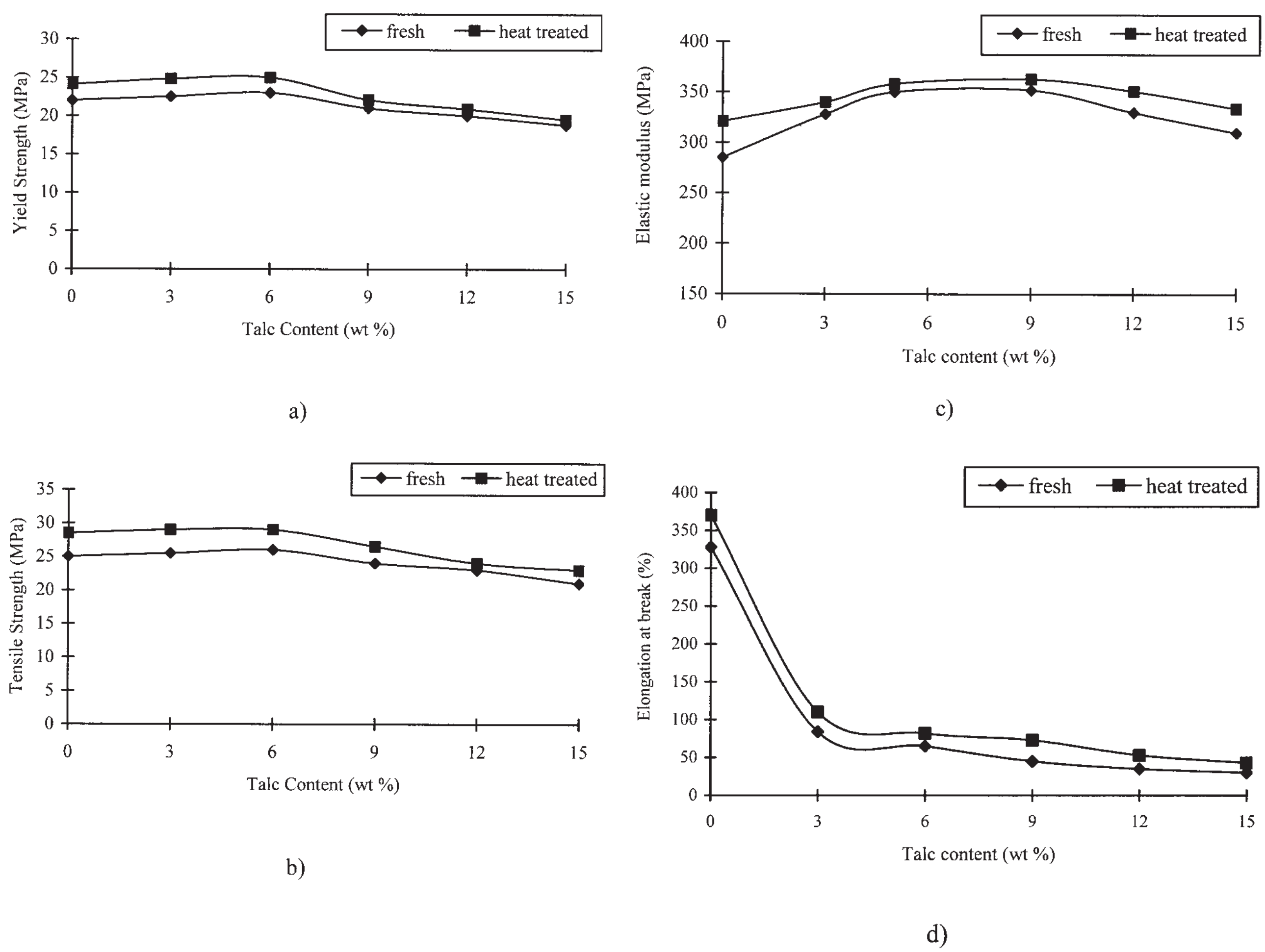

Figure 1 Effect of talc content on (a) yield, (b) tensile strengths, (c) elasticity modulus, and (d) elongation at break of i-PP/EPDM/Talc ternary composite.

categories: for the talc contents up to 6 wt % and for the contents higher than 6 wt % talc. As can be seen from the figure, yield and tensile strengths increased slightly for up to 6 wt % talc and tended to decrease for the higher contents. This behavior may be explained by the effects of both talc with EPDM and the distribution of the talc in matrix. The filler type, distribution, and interaction with matrix are the most important factors affecting the mechanical strength of both binary (PP/filler)[2,4–6] and ternary (PP/filler/elastomer)[11–16,20] composites. Homogenous distribution of the filler and strong adhesion between matrix and filler may cause mechanical strength to increase.[1,2,4,8,20] It is well documented that talc addition to the i-PP increases the yield and tensile strengths,[2,4,6,7] while addition of elastomer, EPDM, decreases.[21–23] In the present study, for lower talc contents up to 6 wt %, the effect of talc on the slightly higher yield and tensile strengths can be related to the good distribution of talc in the matrix. For the talc contents higher than 6 wt %, the decrease in yield and tensile strengths can be related to the nonhomogenous distribution of talc. As clearly evidenced by SEM studies (Fig. 4), talc agglomerated as its content increased. Therefore, it can be said that EPDM is dominant on the lower yield and tensile strengths for the talc contents higher than 6 wt %. From the Figure 1(a,b), it can also be seen that heat treatment increased the yield and tensile strengths. This increase with heat treatment can be related to the good adhesion between i-PP and talc. It is thought that wetability of the matrix is improved by the heat treatment performed after producing ternary composites.

Figure 1(c) shows the effect of talc content on the elasticity modulus of i-PP/EPDM blends. It can be seen from the figure that elasticity modulus of the ternary composites increased as the talc content increased up to 9 wt % talc. The increase in elasticity modulus can be related to both the rigid filler particles and the fillers that restrict the mobility and deformability of the matrix.[2,4] However, for the talc contents higher than 9 wt %, elasticity modulus started to decrease. This decrease in modulus can be related to the nonhomogenous distribution of talc as the content increased. As the heat treatment applied to the composites, the elasticity modulus increased. This increase in the elasticity modulus by the heat treatment can be related to the good adhesion between matrix and talc. In general, considering the effect of talc on the elasticity modulus, the elasticity modulus obtained with and without heat treatment is not as high as obtained for i-PP/Talc binary composite by other researchers.[2,4] It is well documented that elastomer, EPDM, decreases the elasticity modulus,[21,22] while talc increases.[2,4] Therefore, these obtained elasticity modulus values that are lower than expected can be directly related to the presence of EPDM.

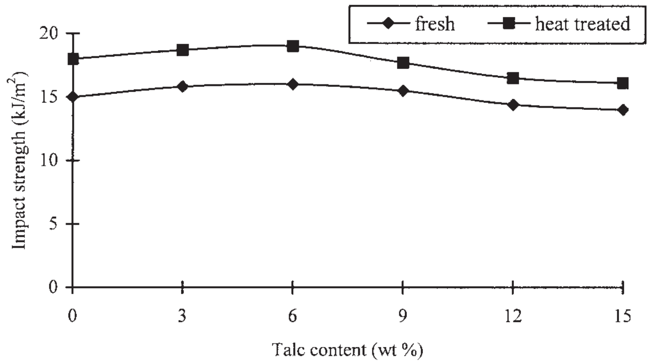

Figure 2 Effect of talc content on impact strength of i-PP/EPDM/Talc ternary composite.

Figure 1(d) shows the effect of talc on the elongation at break. As can bee seen from the figure, elongation at break decreased as the talc content increased. Incorporation of low talc (3 wt %) significantly decreases elongation at break; however, higher talc contents caused elongation at break to decrease more slowly. As reported in the literature, this decrease can be related to the fillers that restrict the mobility of the matrix[2,4] and the result of the matrix reinforcement.[6] In addition, the reduction in elongation at break is not as much as the reported values for the i-PP/Talc composite.[2,6] This can also be directly related to the more flexible EPDM.

Impact test results

Figure 2 shows the notch impact strength values. As can be seen from the figure, the impact strength changed depending on the talc concentration and heat treatment. The impact strength of i-PP/EPDM/Talc composites not subjected to the heat treatment showed a slight increase up to 6 wt % talc and a further decrease at higher filler contents. This may be related to the nonhomogenous distribution of the talc in the matrix for higher contents. In general, from the result of the impact tests without heat treatment, the impact strength of i-PP/EPDM/Talc ternary composite did not show a significant change. This is also related to the effects of both EPDM and talc. It is well known that EPDM increases the impact strength,[21–24] while talc decreases, especially for higher talc contents.[2,4] Therefore, it is accepted that the effect of EPDM on higher impact strength was lowered by the talc. Figure 2 also shows that heat treatment is effective on the higher impact strength values. This increase can be directly related to the high level of adhesion between the matrix and talc. As the crack is generated because of an impact, it propagates towards a poor interfacial region.[4] However, heat treatment provides higher load transfer due to the sufficient interaction or bond strength. For the heat treated com-

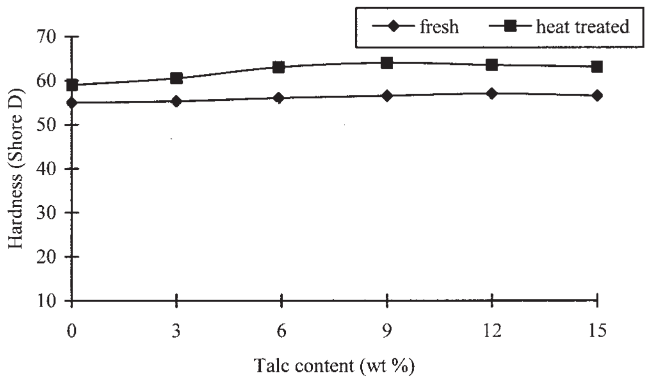

Figure 3 Effect of talc content on the hardness values of i-PP/EPDM/Talc ternary composite.

posites, higher level of impact strength values were also seen for lower talc contents. This is also related to the homogenous distribution of the talc for lower contents.

From the results of the tensile and impact tests, in general, the obtained results are in good agreement with the literature when the effects of EPDM[21–24] and talc[2,4,6,7] on the i-PP are considered, respectively. However, the production of such a ternary composite, i-PP/EPDM/Talc, and the effect of heat treatment on the tensile and impact properties have been reported by the present study for the first time.

Hardness measurement

The hardness values of i-PP/EPDM/Talc ternary composites are shown in Figure 3. As can be seen from the figure, no significant changes depending upon talc increase were observed in the hardness values of the specimens that were not subjected to heat treatment. However, the hardness values showed a slight increase with heat treatment. This increase in hardness with heat treatment can be related to good interaction between matrix and talc and this restricts the mobility and deformability of the matrix.[2]

Melt flow index

Table V shows the MFI of i-PP/EPDM/Talc composites. As can be seen from the table, the MFI values change depending on talc concentrations. When the 0 wt % talc point is considered to be the reference point, the MFI values showed a slight decrease with the increase in the talc content. This is due to the undeformability of the filler and its lack of contribution to the flow. This result is consistent with the previous work that was performed on the i-PP/Talc binary composite.[2] However, the effect of the talc on the MFI of such a ternary composite, i-PP/EPDM/Talc, has been reported in the present study for the first time. Increasing the filler concentration in the i-PP/EPDM blends increases the viscosity. Polymer flow into the mold decreases depending on the filler concentration.

Scanning electron microscopy

SEM micrographs of Izod impact fracture surfaces of specimens subjected to heat treatment are shown in Figure 4 for the contents of 3, 9, and 15 wt % talc. As can be seen from the micrographs, talc and EPDM have quite a fine distribution in the i-PP matrix and there is a quite strong adhesion between matrix and filler. However, it was also seen that tendency towards formation of agglomerates as the talc content increased. This result is in good agreement with the literature.[1] As apparent from the tensile strength properties [Fig. 1(a–c)], the tendency to decrease in yield and tensile strengths, and elasticity modulus for the higher talc contents, can be related to the talc distribution. The increase in the amount of talc makes the distribution within the matrix difficult.

CONCLUSIONS

The effect of talc on the mechanical properties such as yield and tensile strengths, elongation at break, elasticity modulus, hardness, and impact strength with and without heat treatment, thermal, such as MFI, and microstructural properties of i-PP/EPDM blends was investigated. The following results can be drawn from the experimental results.

1. Yield strength with heat treatment increased for the talc contents up to 6 wt %.
2. Tensile strength with heat treatment increased for the talc contents up to 6 wt %.
3. Elongation at break decreased as talc content increased.
4. Elastic modulus increased as the talc content increased up to 9 wt %. Heat treatment increased the elastic modulus.
5. Impact strength with heat treatment increased up to 6 wt % talc content.
6. Heat treatment caused the hardness to increase.
7. MFI decreased as talc content increased.

TABLE V
Effect of Talc Content on MFI Values of i-PP/EPDM/Talc Ternary Composite

Talc (wt %)	MFI (g/10 min)
0	4.92
3	4.85
6	4.76
9	4.65
12	4.61
15	4.54

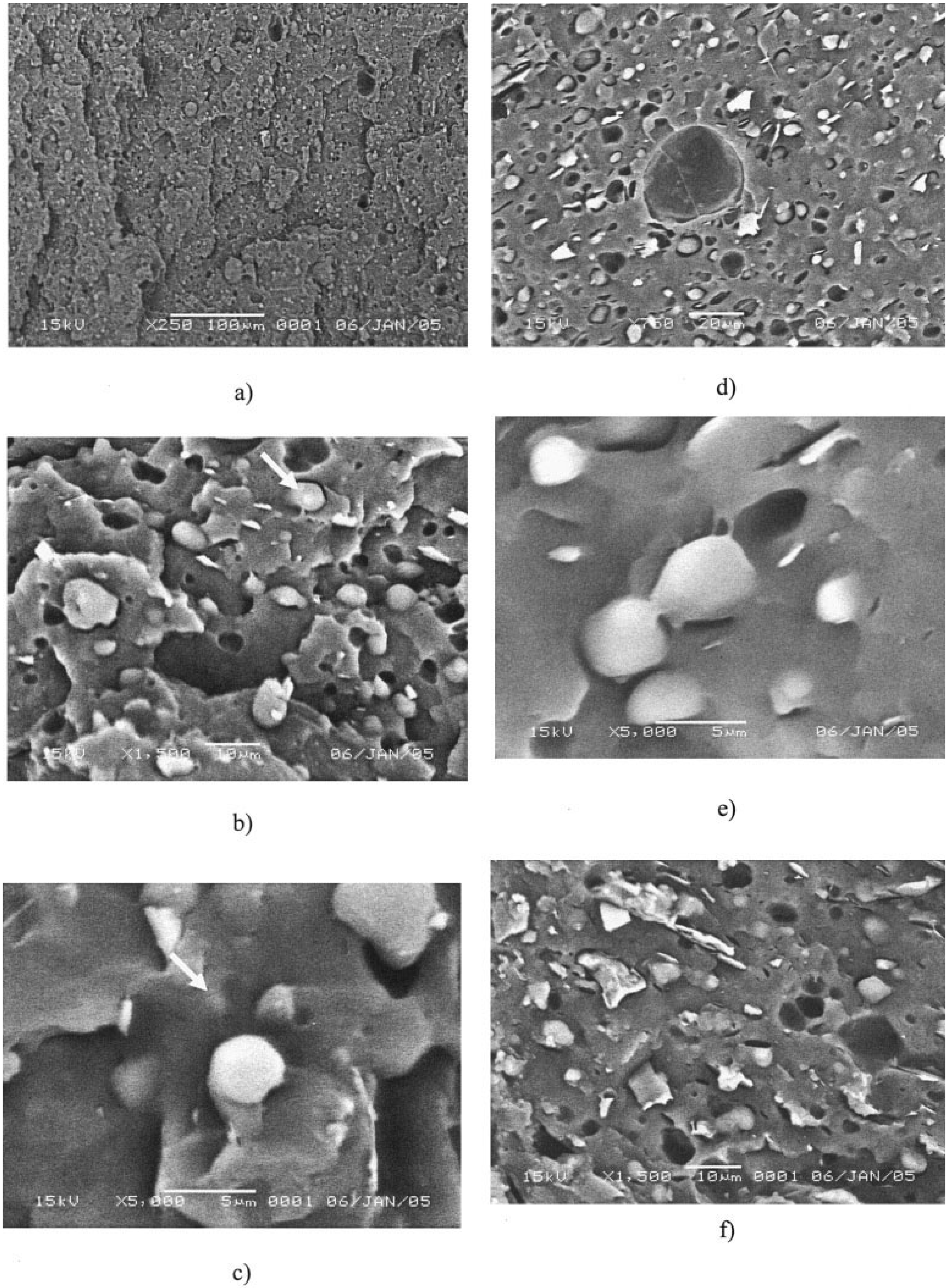

Figure 4 SEM micrographs of i-PP/EPDM/Talc blends: (a), (b) (arrow shows EPDM), and (c) (arrow shows talc) i-PP/EPD/3 wt %Talc; (d) and (e) i-PP/EPDM/9 wt %Talc; and (f) i-PP/EPDM/15 wt %Talc.

8. Talc showed a fine distribution in the i-PP matrix for contents up to 6 wt %.

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