# Phase structure of impact-modified polypropylene blends

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A morphological study of polypropylene/ethylene\_propylene\_diene terpolymer (PP/EPDM) and polypropylene (PP)/polyolefin thermoplastic rubber blends was conducted. Dispersion of the impact modifier in the blend was investigated by transmission and scanning electron microscopy (TEM and SEM). It was established that number-average particle size  $(\bar{D}_n)$  of the EPDM impact modifier increased with its melt viscosity. The differences in melt viscosities of the blended components were characterized by the phase viscosity ratio ( $\mu$ ). The course of  $\bar{D}_n$  vs. log  $\mu$  function was qualitatively in agreement with the Rayleigh–Taylor–Tomotika theory. Accordingly, high degree of dispersion of the impact modifier can be achieved if melt viscosities of the blended components are very closely matched, i.e. if  $\mu \simeq 1$ . It was concluded from SEM results that, below an impact modifier content of 20%, the modifier formed the dispersed phase in the continuous PP matrix. In blends containing 50% of impact modifier, the latter may also form continuous phase depending on its type and  $\mu$  value beside the still continuous PP phase (cocontinuous network structure).

**Keywords** Polymer blends; morphology; ethylene-propylene-diene terpolymers; thermoplastic rubber; high-impact blends; polyolefin, polypropylene

# **INTRODUCTION**

Melt blending of polypropylene (PP) homo- and copolymers with different elastomers and rubbers (such as butyl rubber, ethylene-propylene copolymers (EPM), ethylene-propylene-diene terpolymers (EPDM), polyisobutylene, styrene-butadiene block copolymers, etc.) is widely used for increasing the impact strength of PP especially at low temperatures<sup>1</sup>. In practice, predominantly EPDM and EPM are involved as rubber impact modifiers and styrene-butadiene based and highethylene ethylene-propylene block copolymers as thermoplastic elastomers.

It was revealed by the investigations of amorphous high-impact polymers (such as high-impact polystyrene, ABS, high-impact PVC, modified poly(phenylene oxide), etc.) that a close interrelation existed between their physico-mechanical properties and phase structure. The macroscopic measured physico-mechanical characteristics were regarded to depend on the average particle size, particle size distribution, and some molecular features (such as average molecular mass, degree of crosslinking, etc.) of the rubbery or elastomeric impact modifier forming the dispersed phase in the blends.

Discovery of structure-properties relationships of semicrystalline impact-resistant polymer blends follows essentially two ways nowadays<sup>2</sup>. One of them would find analogies between the behaviour of amorphous and semicrystalline high-impact polymers or blends through knowledge of the regularities for amorphous high-impact systems. On the other hand, the other one deals with the changes in the crystalline phase of modified semicrystalline polymers caused by their melt blending with the impact modifiers.

Studies on the relations between structure and properties of the impact-modified PP have become the centre of interest just recently<sup>3-23</sup> the overwhelming majority of the reported works can be classified as tracking analogies with the high-impact amorphous polymers. Only a few papers have attempted to discover the changes in the crystalline phase of PP by virtue of its impact modification by melt blending with a rubber<sup>3-5,11,15,20</sup>. It is expected that a synthesis of these two approaches will provide a satisfactory interpretation of the experimental observations. Current practical experience surpasses theoretical knowledge by far. According to the present concepts, the direction of searching for analogies with the behaviour of amorphous high-impact polymers holds out more promise.

It is well known from the literature that the degree of dispersity (average particle size) of the impact modifier has a great influence on the physico-mechanical properties, especially on the impact characteristics<sup>6-8,16,18,19,21,23</sup>: principally through the changes in development and interaction possibilities<sup>24</sup> of the energy-absorbing mechanisms/crazing and/or shear yielding<sup>16,19,21,23,25</sup>. It is generally accepted that a higher degree of dispersion of the impact modifier involves more favourable preconditions for generation of a ductile deformation<sup>6,16,19,21,23</sup>. It is widely accepted that different shear fields developed during compounding and moulding can result in a strong variation in rubber particle size

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				Melt flow ind	lex (g/10 min)	Mooney	•			viscositγ <sup>£</sup> ο, μ
Trade name	Туре	10 <sup>-5</sup> M	10-4 M <sub>n</sub>	190°C/48.5 N	230°C/21.2N	<ul> <li>viscosity</li> <li>ML (1 + 4)</li> </ul>		proportion (%)	(1)	(2)
Tipplen H523	PP-HC	5.16	3.10		3.80					
Tipplen H791F	РР—Н <sup>с</sup>	4.30	6.60		0.84					
Buna AP 147	EPDM	1.25	0.85			30 <i>e</i>	66.6		1.68	1.15
Buna AP 447	EPDM	1.95	4.88			85 <i>°</i>	63.2		3.17	2.15
Buna AP 251	EPDM	1.57	1.08			42 <i>e</i>	44.9		1.64	1.14
Buna AP 451	EPDM	2.23	0.53			90 <i>e</i>	50.0		2.94	2.01
Buna AP 341	EPDM	1.87	5.33			70 <sup>e</sup>	54.2		2.48	1.70
Buna AP 541	EPDM	2.20	2.02			110 <sup>e</sup>	50.0	,	3.40	2.32
Nordel 2722	EPDM	1.45	1.00			25 <sup>f</sup>	44.2			0.96
Nordel 2522	EPDM	1.27	1.50			25 <sup>f</sup>	48.4			1.20
Nordel 2744	EPDM	1.40	3.15			48 <sup>f</sup>	57.2			1.85
Vestopren TP 2047	POTPR	2.40	0.85	1,1	1 y		72.8	32.2	1.80	
Vestopren TP 2037 POTPR		2.33	0.70	0.8			77.8	20.9	1.98	
Vestopren FB 7239 <sup>d</sup> POTPR		1.86	0.80	0.7			83.0	23.3	1.83	
Vestopren FB 7238d POTPR		1.85	0.90	0.8			85.7	24.0	1.86	

Table 1 Characteristics of the materials used

<sup>a</sup> In terms of ethylene content

b Calculated as the ratio of Brabender torques (190° C, 5 min) of the pure impact modifier to that of the pure PP16,17,19,21,22 in the case of Tipplen H523 (1) and Tipplen H791F matrix (2)

<sup>C</sup> PP homopolymer

d Experimental product

<sup>e</sup> Measured at 100°C, prospectus data

f Measured at 121°C, prospectus data

and size distribution of impact-modified PP blends<sup>26</sup>. On the other hand, several papers<sup>6,9,19,21,23</sup> suggest that these parameters—which influence the blend characteristics dramatically (first of all the impact properties) depend principally on the melt viscosities and melt viscosity ratio of the components during melt mixing.

The present paper aims at a study on the nature of changes in the average particle size and the particle size distribution of the rubber or elastomer impact modifier blended at 10% in PP as functions of melt viscosity ratios measured in the course of mixing the matrix with the impact modifier, i.e. the phase viscosity ratio. Furthermore, it was intended to decide whether the relationship between the average particle size and the phase viscosity ratio followed Rayleigh-Taylor-Tomotika the theory<sup>27-31</sup>. As an additional objective, some information was collected on the questions of: at which concentration of the impact modifier the phase inversion would take place and what it depended on.

# **EXPERIMENTAL**

Tipplen H523 injection-grade and Tipplen H791F extrusion-grade PP homopolymers (Tisza Chemical Works, Leninváros, Hungary) were used in the experiments. The following impact modifiers were applied: Buna AP ethylene-propylene-ethylidenenorbornene, Nordel ethylene-propylene-1,4-hexadiene terpolymers (EPDM, products of Hüls and Du Pont, respectively) and Vestopren TP saturated high-ethylene content ethylenepropylene block copolymer. Since the latter is regarded as a polyolefinic thermoplastic rubber, it is abbreviated as POTPR in the following. The materials and their essential characteristics are collected in *Table 1*.

 $\bar{M}_m$  and  $\bar{M}_n$  values of the polymers were determined by gel permeation chromatography (g.p.c.) in 1,2,4-trichlorobenzene at 130°C (Waters instrument). For the impact

modifiers, regarding their high ethylene content, calibration for polyethylene was used. The ethylene proportions of the impact modifiers were determined by infra-red spectroscopy using the calibration curve of Corish and Tunnicliffe<sup>32</sup> for  $A_{1380\,\text{cm}^{-1}}/A_{1460\,\text{cm}^{-1}}$  ratio. The crystalline PE proportion of POTPR was determined by differential scanning calorimetry (Du Pont 910 DSC instrument), supposing the heat of fusion of 100%-crystallinity PE as<sup>33</sup> 287 J g<sup>-1</sup>.

Techniques of blending and determination of the phase viscosity ratio ( $\mu$ ) were described in previous reports<sup>16, 17, 19, 21, 22</sup>.

The ultrathin sections for transmission electron microscopy (TEM) were prepared by means of a Reichert Om U2 ultramicrotome using an FC-2 deep-freezer at  $-50^{\circ}$ C. Samples containing EPDM were soaked in a 1% aqueous solution of OsO<sub>4</sub> for 3 days by the Kato's method<sup>34</sup> before sectioning. The specimens sectioned by the glass knife were floated onto the surface of an equivolumetric mixture of dimethyl sulphoxide (DMSO) and water. They were fished out before placing into the sample holder. TEM records were taken by a Zeiss Elmi D-2 electron microscope.

Fracture surfaces of small notched Charpy specimens were studied by scanning electron microscopy (SEM) at different temperatures by a Cambridge Stereoscan S 4 10 electron microscope. Conductivity of the fracture surface was brought about by a Au layer applied by cathodic metallization. Since the information content of the records is different depending on the fracture temperature of the specimen<sup>17, 22</sup>, i.e. a fracture below  $T_g$  of the impact modifier may lead to falsified results as the fracture line can cross a particle of the additive<sup>7</sup>, a selective dissolution of the impact modifier was also attempted. In order to determine the particle size and particle size distribution of the dispersed impact modifier, chemically etched fracture surfaces were recorded.

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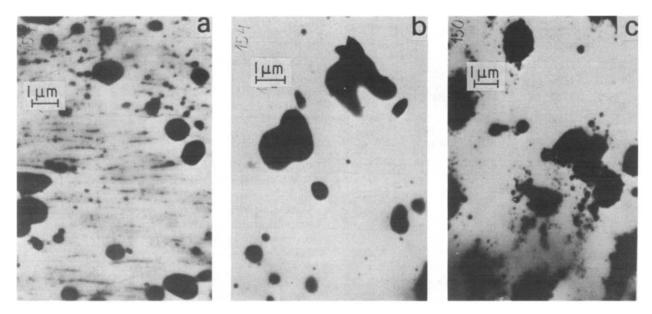


Figure 1 TEM micrographs on blends of Tipplen H523 with 10% of Buna AP 447 EPDM. Mixing by a Brabender Plastograph for (a) 5 min, (b) 15 min and (c) 40 min

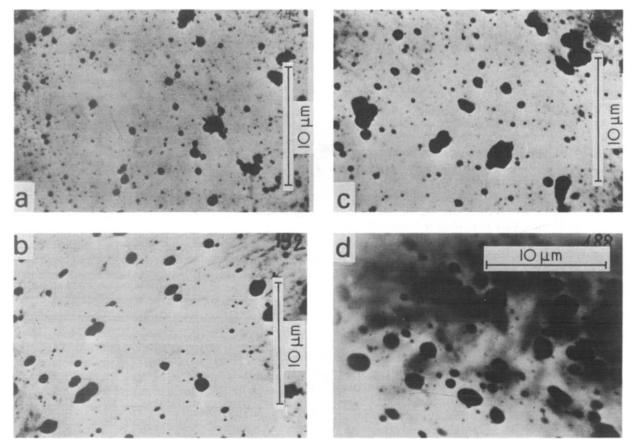


Figure 2 TEM micrographs on blends of Tipplen H523 with 10% of (a) Buna AP 341 ( $\mu$ =2.48), (b) Buna AP 451 ( $\mu$ =2.94), (c) Buna AP 447 ( $\mu$ =3.17) and (d) Buna AP 541 ( $\mu$ =3.40)

# **RESULTS AND DISCUSSION**

TEM records on ultrathin sections of blends of Tipplen H523 with 10% of Buna AP 447 are shown in *Figure 1* after different mixing times. Black areas refer to EPDM contrasted by  $OsO_4$ .

It is easily seen in *Figure 1* that no considerable improvement in the degree of dispersion can be achieved by increasing the mixing time beyond a reasonable  $f(x) = \frac{1}{2} \int_{-\infty}^{\infty} \frac{1}{2} \int_{-\infty}^{$ 

period; the average particle size of EPDM is about  $1 \,\mu m$  throughout.

In Figure 2, effect of  $\mu$  value of EPDM on the degree of dispersity of the rubber is illustrated by the example of systems containing 10% of EPDM.

It is well demonstrated in *Figure 2* that the average particle size of EPDM increases with  $\mu$ . The average particle size is especially high when Buna AP 447 and Buna AP 541 are involved. TEM records are, however,

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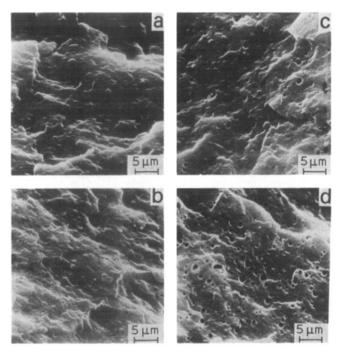


Figure 3 SEM records on surfaces of Tipplen H523 blend with 10% of Buna AP 251 fractured (a) at  $-196^{\circ}$ C, (b) at  $-60^{\circ}$ C, (c) at  $-40^{\circ}$ C and (d) at  $-40^{\circ}$ C followed by etching with n-heptane vapour for 20 s

unsuited to determination of the average particle size and the particle size distribution of the additive since they refer only to very small microsurfaces where only few particles are found. The risk of non-representativity of the studied surface part to the phase structure was also pointed out by Thamm<sup>7</sup>. In spite of this fact, this method was used by Speri and Patrick<sup>6</sup> in their often cited work for determination of the average particle size and the particle size distribution of EPDM through the  $D_m/D_n$  ratio which is characteristic to the broadening of distribution ( $D_m$  and  $D_n$  are the mass- and number-averages of the particle diameter, respectively).

The average particle size and the particle size distribution of the impact modifiers were determined by SEM records (*Figure 3*) of fracture surfaces broken at different temperatures or etched chemically prepared from Charpy specimens of the blends<sup>17,23</sup>.

It can be seen in Figure 3 that the information content of a surface broken at the temperature of liquid N<sub>2</sub> is quite low since the fracture line or surface passes through the EPDM particles occurring in the glassy state. Surfaces broken at  $-40^{\circ}$ C are much more structured, presumably due to the fact that this temperature is above or very close to  $T_g$  of EPDM<sup>2, 19, 21</sup>. Hemispherical embeddings and hollows representing removed particles are clearly visible on the fracture surface. Determination possibilities of the particle size and its distribution can be improved by a selective removal of the impact modifier by chemical etching.

It is accepted that the atactic proportion of PP is determined by its heptane dissolubility (cf. DIN 53738 standard). If PP particles higher than  $100 \,\mu\text{m}$  (prescribed in the standard) are extracted by n-heptane, the extractable proportion will be much lower; in fact, from a granulated polymer, usually not more than 2% can be extracted. On the other hand, EPDM pieces, even of bigger size than the granules, dissolve practically up to 100% in the course of a 24 h extraction in n-heptane. This means that n-heptane is suitable for the selective chemical etching of EPDM as was reported previously<sup>17,22</sup>. This etching technique was successfully applied by other workers<sup>10,35</sup> as well for studies on phase structure. In *Figure 4*, SEM records of surfaces broken and etched at  $-40^{\circ}$ C are presented for Tipplen H791F based blends containing 10% of EPDM with different  $\mu$  and  $\bar{D}_n$  values.

Micrographs of chemically etched fracture surfaces of Tipplen H523 matrices containing 10% of EPDM with different  $\mu$  values are shown in *Figure 5*.

It is clearly seen in Figure 4 and particularly in Figure 5 that the average particle size of the dispersed EPDM is increasing with increasing phase viscosity ratio. At SEM magnifications suitable for determination of particle size distribution—where 100 separate particles are visible at least—a visual reading is possible. Covering curves of the histograms for relative frequencies of given particle diameters are demonstrated in Figure 6 on the basis of visual evaluation of blends on extrusion-grade PP matrix containing 10% of Buna AP EPDM types.

The same covering curves for the blends of injectiongrade PP matrices with 10% of EPDM of different  $\mu$ values and the corresponding average particle sizes were presented in a previous paper<sup>17</sup>.  $\bar{D}_n$  values obtained by visual evaluation of blends containing 10% of EPDM are plotted against the logarithm of phase viscosity ratio in *Figure 7*.

 $\overline{D}_n$  tends to increase with increasing  $\mu$  value and, as a rough estimation, it may be linearized at least in this range (*Figure 8*). This means that essentially identical relations are obtained by plotting the physico-mechanical characteristics against  $\overline{D}_n$  instead of  $\mu$ .

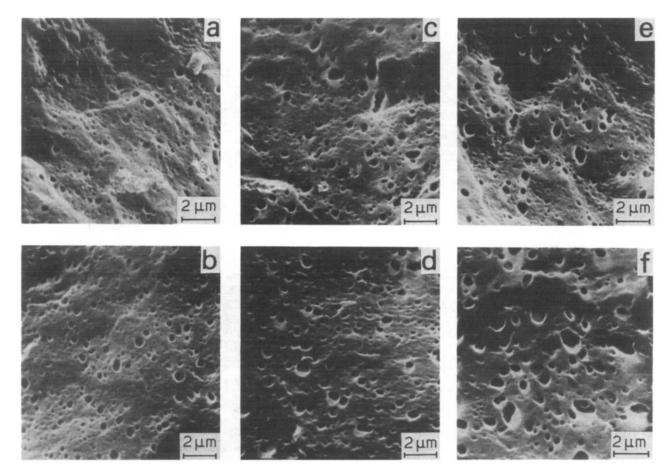
The course of the  $\overline{D}_n$  versus log  $\mu$  function corresponds to expectations on the basis of the Rayleigh-Taylor-Tomotika theory $2^{7-31}$ , plotting the changes in the radius of the fluid drop formed by disintegration of a cylindrical thread in another viscous liquid (lubricating oil thread in a syrup) it will break up into a series of small droplets spaced at nearly regular intervals. Treating this phenomenon mathematically, Rayleigh<sup>28</sup> established that a cylindrical column of a viscous liquid in vacuo, i.e. neglecting the effect of the liquid surrounding the viscous thread, showed maximum instability when the wavelength of an assumed initial varicosity was infinitely great. Based on this theory-on the contrary to the experiments-very large drops would have been expected. The effect of the liquid surrounding the thread was also taken into account by Tomotika<sup>30,31</sup> who gave the following function for the instability coefficient (q):

$$q = \frac{\gamma}{2\eta_0 R} (1 - x^2) F(x, \mu) = \frac{\gamma}{2\eta_0 R} \Omega(x, \mu)$$

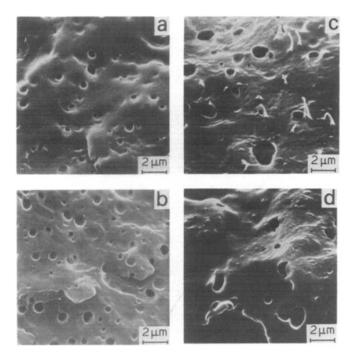
where  $\gamma$  is the interfacial surface tension,  $x = 2\pi R/\lambda$ , R is the diameter of the thread,  $\lambda$  is the wavelength of the varicosity on the thread,  $\mu$  is the phase viscosity ratio defined as  $\mu = \eta_1/\eta_0$ , where  $\eta_1$  is the viscosity of the thread and  $\eta_0$  is that of its surrounding liquid.

It was found by Tomotika<sup>30</sup> that, at finite values of  $\mu$ , the maximum instability occurred at a certain definite value of  $\lambda$  indicating that drops of definite size would be formed and that  $\lambda$  varied with  $\mu$ . The analysed x versus log  $\mu$  function gave maximum at log  $\mu \simeq -0.5$  (it is the extremum of the q function). It means that the instability

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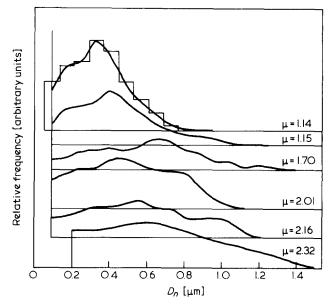


*Figure 4* SEM micrographs on chemically etched fracture surfaces of Tipplen H791F based blends containing 10% of (a) Nordel 2722  $(\bar{D}_n=0.28 \ \mu\text{m})$ , (b) Buna AP 251  $(\bar{D}_n=0.33 \ \mu\text{m})$ , (c) Buna AP 447  $(\bar{D}_n=0.54 \ \mu\text{m})$ , (d) Nordel 2522  $(\bar{D}_n=0.54 \ \mu\text{m})$ , (e) Nordel 2744  $(\bar{D}_n=0.58 \ \mu\text{m})$  and (f) Buna AP 341  $(\bar{D}_n=0.60 \ \mu\text{m})$ 



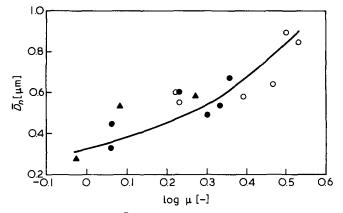
*Figure 5* SEM micrographs on chemically etched fracture surfaces of Tipplen H523 based blends containing 10% of (a) Buna AP 147 ( $\mu$ =1.68), (b) Buna AP 251 ( $\mu$ =1.64), (c) Buna AP 447 ( $\mu$ =3.17) and (d) Buna AP 541 ( $\mu$ =3.40)

of a viscous thread is maximum if the  $\eta_1$  and  $\eta_0$  values are very closely matched, i.e. if  $\mu$  is near to unity. Destabilization of such cylindrical threads plays a very great



*Figure 6* Covering curves of histograms from the evaluation of SEM records on surfaces of Tipplen H791F blends with 10% of Buna AP EPDM types fractured at  $-40^{\circ}$ C followed by chemical etching as plotted against  $\mu$  values of the various impact modifiers

part in melt mixing as well<sup>27</sup>. Validity of the above theory was shown not only for liquid elastomers<sup>36</sup> but also for melts of polymeric blends<sup>37</sup> and it was established that the theory covered not only Newtonian liquid pairs. Accordingly, the  $\bar{D}_n$  versus log  $\mu$  function shows a minimum in the



*Figure* 7 Function of  $\overline{D}_n$  vs. log  $\mu$  following the example of blends containing 10% of EPDM:  $\bigcirc$ , Buna EPDM types in Tipplen H523 matrix;  $\bigcirc$ , Buna EPDM types in Tipplen H791F matrix;  $\blacktriangle$ , Nordel EPDM types in Tipplen H791F matrix

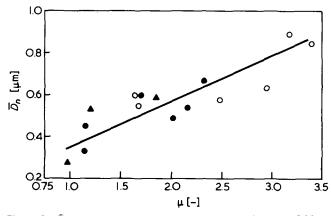


Figure 8  $\bar{D}_n$  vs.  $\mu$  function of blends containing 10% of EPDM: for symbols see Figure 7

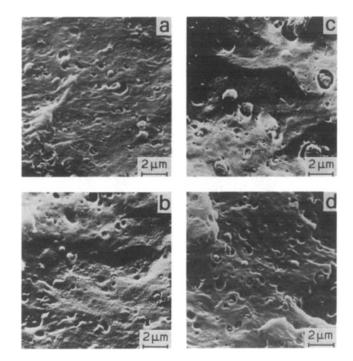
vicinity of  $\mu = 1^{27,30,36,37}$ . EPDM and PP grades used in these experiments did not permit us to reach  $\mu$  values far below 1, thus they made the determination of the minimum in the  $\overline{D}_n$  versus log  $\mu$  function impossible.

In Figure 9, SEM micrographs of surfaces broken at  $-40^{\circ}$ C and etched by n-heptane are presented for Tipplen H523 based blends containing 10% of POTPR.

In view of the records in Figure 9, dispersion and average particle size of POTPR can easily be established though solubility of POTPR grades in n-heptane is only 60-64%-depending on their PE proportion. This means that not all the additive particles are represented by hollows and craters after etching. PE domains undissolved by heptane and embedded into the particles are visible in Figure 9 representing the physical crosslinking domains of POTPR. Such a composite structure is in accord with phase contrast microscopic and dynamic mechanical spectroscopic results<sup>38</sup>; in fact, it is identical to those obtained for PP/EPDM/PE ternary systems<sup>14,18</sup>. It is very interesting that Kojima<sup>15</sup> found the elastomer phase biphasic consisting of an EPM and a PE phase in his SEM investigations on the structure of PP block copolymers. It would mean that the elastomeric proportion of PP block copolymers has an identical structure to POTPR.

In the present work, quantitation of size distribution of the dispersed particles was based on the  $\bar{D}_m/\bar{D}_n$  ratio according to Speri and Patrick<sup>6</sup>. The mass-average particle diameter  $(\bar{D}_m)$  could be calculated through the determination of relative frequencies of the particle diameters. Dispersity of EPDM or POTPR in the blends containing 10% of the additive is indicated in *Table 2* by  $\bar{D}_n$ ,  $\bar{D}_m$ , and  $\bar{D}_m/\bar{D}_n$  values as determined from the SEM records.

The results collected in *Table 2* are in accord with those reported in the literature<sup>6,7,18</sup>. It can also be established from these data that technique and conditions of blending were efficient. No information has been obtained, however, from the experimental results about the reason for the formation of the observed particle size distribution  $(\bar{D}_m/\bar{D}_n)$  and about some deviating values in the course of



*Figure 9* SEM records on chemically etched fracture surfaces of Tipplen H523 based blends containing 10% of (a) Vestopren TP 2047 ( $\mu$ =1.80), (b) Vestopren TP 2037 ( $\mu$ =1.98), (c) Vestopren FB 7239 ( $\mu$ =1.83) and (d) Vestopren FB 7238 ( $\mu$ =1.86) POTPR grades

Table 2 Particle size data of blends containing 10% of various impact modifiers (for designations of Table 1)

		μ	<i>D̄<sub>n</sub></i> (μm)	<i>D̄m</i> (μm)	$\overline{D}_m/\overline{D}_n$
Impact modifier	(1)	(2)			
EPDM					·
Buna AP 251	1.64		0.60	1.17	1.95
		1.14	0.33	0.51	1.55
Buna AP 147	1.68		0.55	0.70	1.27
		1.15	0.45	0.81	1.80
Buna AP 341	2.48		0.58	1.03	1.78
		1.70	0.60	0.91	1.52
Buna AP 451	2.94		0.64	1.13	1.77
		2.01	0.49	0.73	1.49
Buna AP 447	3.17		0.90	1.65	1.83
		2.16	0.54	0.83	1.54
Buna AP 541	3.40		0.85	1.40	1.65
		2.32	0.67	1.08	1.61
Nordel 2722		0.96	0.28	0.46	1.64
Nordel 2522		1.20	0.54	1.03	1.91
Nordel 2744		1.85	0.58	1.10	1.90
POTPR					
Vestopren TP 20	47 1.80		0.65	1.32	2.03
Vestopren TP 20	37 1.98		0.75	1.20	1.60
Vestopren FB 72	39 1.83		0.70	0.90	1.29
Vestopren FB 72	238 1.86		0.64	0.79	1.23

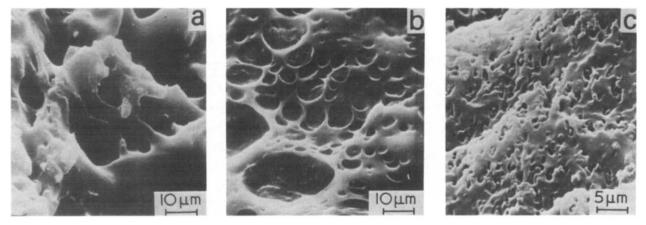


Figure 10 SEM micrographs on chemically etched fracture surfaces of Tipplen H523 based blends containing 50% of (a) Buna AP 147, (b) Buna AP 447 and (c) Vestopren TP 2047 impact modifiers

the  $\bar{D}_n$  versus log  $\mu$  and  $\bar{D}_n$  versus  $\mu$  curves. The scattering of points on these functions is probably due to different compatibilities or segmental solubilities<sup>39</sup> of the various EPDM types with PP owing to their various molecularlevel parameters such as ethylene contents,  $\bar{M}_n$  and  $\bar{M}_m$ values, termonomer quality and content, block of stereoblock lengths of the ethylene and/or propylene segments, etc. It seems that fine dispersion can be achieved if the applied EPDM has a low ethylene content, a low  $\bar{M}_n$ , a low Mooney viscosity and regarding the latter in connection with the PP matrix if  $\mu \simeq 1$ .

The concepts that appear in the literature are very contradictory concerning the impact modifier concentration of the phase inversion, i.e. where the impact modifier transforms into the continuous phase. Onogi *et al.*<sup>3</sup> concluded from their dynamic mechanical and stress-strain studies that it occurred at an additive content of 60–70%. Similar results were reported by Danesi and Porter<sup>9</sup> for PP-EP(D)M systems on the basis of phase contrast microscopic investigations. According to Duvdevani *et al.*<sup>10</sup>, both phases are continuous (designated as 'co-continuous' system or qualified sometimes as interpenetrating network) in the range of impact modifier concentration of 20–75%. The concept of Kresge<sup>35</sup> is essentially similar as PP forms a continuous phase even at an additive content of 85%.

In the present instances of PP blends containing 20% of EPDM or POTPR, the impact modifier is the dispersed phase in the continuous PP matrix. In addition of EPDM grades having high  $\mu$  values (Buna AP 447 and 541) at 20%, however, a co-continuous network was actually observed in some samples. It was concluded on the SEM micrographs of etched fracture surfaces at 50% of additive content (*Figure 10*) that the EPDM elastomers formed continuous phases (PP was in a continuous phase by all means) while POTPR was present as a dispersed phase even at this concentration of 50%.

The different concepts and conclusions about the phase inversion may lead to a common stand by taking the role of  $\mu$  value and chemical-physical structure of the additive into consideration. Incorporation of an EPDM having higher  $\mu$  value may result in a co-continuous network in some samples at a level as low as 20%. This can be explained by the much smaller chance of destabilization or even destruction of the cylindrical threads during the blending process than for additives of lower  $\mu$  value. This phenomenon is favourable to the formation of cocontinuous phases. In the system containing 50% of POTPR (*Figure 10c*), possibly the effect of the physical network structure formed due to the presence of PE domains is responsible for holding the additive in the dispersed phase.

## CONCLUSIONS

It was unequivocally demonstrated by TEM records that PP/EPDM blends were two-phase systems consisting of the impact modifier in the form of spherical particles at a size of about 1  $\mu$ m. TEM records suggested the qualitative conclusion that, with increasing difference in the melt viscosity of PP and EPDM—as characterized by the phase viscosity ratio ( $\mu$ )—the dispersion of the impact modifier decreased, i.e. the average particle size increased.

SEM micrographs on chemically etched fracture surfaces of blends were suitable for determination of the average particle size and the particle size distribution. According to the experimental results, higher  $\mu$  values involved higher number-average particle diameter  $(\bar{D}_n)$  of the impact modifier. The  $\bar{D}_n$  versus log  $\mu$  function was qualitatively in agreement with the Rayleigh-Taylor-Tomotika theory as was expected. It showed again that this theory for immiscible pairs of Newtonian liquids was valid for polymers as well. According to the results, very fine dispersions can be achieved if the ethylene content and the  $\bar{M}_n$  of the EPDM impact modifier is low, and the  $\mu$ value is near to unity.

It was revealed by the investigations on phase structure of PP blends with polyolefin thermoplastic rubbers (POTPR) that the latter was biphasic itself; in fact, after an extraction by n-heptane, PE domains were clearly perceptible in its texture.

SEM records showed that the impact modifier formed the dispersed phase up to a concentration of 20% in the continuous PP matrix. At an additive content of 50%, PP still existed as a continuous phase but the impact modifier might also form a continuous phase depending on its type and  $\mu$  value (co-continuous network structure).

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