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Effect of elastomer interfacial agents on tensile and impact properties of $CaCO_3$ filled HDPE

F. SAHNOUNE, J. M. LOPEZ-CUESTA, A. CRESPY
Laboratoire Matrices, Matériaux Minéraux et Organiques – Ecole des Mines d'Alès-6, avenue de Clavières, 30319 Alès cedex, France

This work focuses on the modification of the tensile yield strength of $CaCO_3$ filled HDPE brought about by the incorporation of SEBS elastomers. Two types of SEBS elastomers were used, grafted and ungrafted with maleic anhydride functions. The grafted elastomer encapsulates the filler particles *in-situ* and creates an adhesive interphase. The tensile yield stress was increased with increasing content of grafted elastomer until a maximum value. The influence of the interfacial area and the volume fraction of filler were studied. It was shown that the relative increase in tensile yield stress when increasing amount of interfacial agent was added, both depends on the volume of filler and the interfacial area.

1. Introduction

The addition of mineral fillers to commercial thermoplastics reduces the overall cost of the composite and offers an important means of achieving new combinations of properties. The stiffness of polymers is generally enhanced by the incorporation of high modulus fillers. The effect of mineral fillers on the elastic modulus of polymers has been widely studied and there are many theoretical models available predicting the behavior of composites in the elastic zone.

The tensile behavior of filled polymer at higher strain level is more complex and depends on non-elastic deformation mechanisms. The effect of fillers on the tensile strength of polymers has been studied by many authors. Some theories and models have been proposed [1–8]. The elaboration of theoretical models is a very difficult task because of the great number of parameters affecting the tensile strength of particulate composites. The main parameters are: the filler weight or volume fraction, the particle shape, the particle size, the nature of the matrix, and its adhesion to the filler particles. Among these factors, the interfacial adhesion is of cardinal importance and markedly influences the tensile strength of filled polymers [2–4, 9].

In the case of poor interfacial adhesion, the tensile strength generally decreases with increasing filler loading. During tensile deformation, the dewetting phenomena i.e. loss of contact between the matrix and the filler particles [10–12] contributes together with shear yielding to the non-elastic deformation. It produces a situation in which the filler cannot sustain much load and the matrix may be considered as filled with voids. Most of the models regard the matrix as the only stress-bearing component in the composite when adhesion is poor. They assume that the tensile strength of the composite is proportional to the cross-sectional area of the load-bearing polymer matrix. The incorporation of filler leads to an effective decrease in the cross-sectional area of polymer and thus a decrease in tensile yield strength. Moreover, when there is poor interfacial adhesion, the filler acts as stress concentrator in a more effective manner. This effect also contributes to the reduction in tensile strength of the material [9].

Tensile strength is generally enhanced when interfacial adhesion is improved. This can be ascribed to better stress transfer at the interface between the matrix and the filler [2, 3]. The improvement of interfacial adhesion can prevent dewetting (i.e. loss of contact) at the matrix-filler interface during tensile deformation [11]. Therefore, well adhering filler particles can bear on part of the load applied to the matrix and contribute to the tensile strength of the composite. The addition of filler combined with an efficient coupling agent has proved to be a way to enhance the tensile strength of polymers.

However, the increase in adhesion may produce local changes in the micromorphology and the mechanical properties of the polymer chains bonded to the filler particles. That may result in the formation of a rigid polymer layer surrounding the filler particles. This rigid interphase may have mechanical properties rather closer to the filler than to the matrix [13, 14]. It increases the apparent volume fraction of filler and leads to a stiffer but more brittle composite.

Previous works [15] have shown that the encapsulation of kaolin particles by latex improves impact properties of PP/kaolin. The embrittlement of the material can be reduced if mineral fillers are coated with a soft interphase. Impact properties were also improved in chalk filled PP [13] and HDPE [16] by coating chalk particles with a liquid oligomer. However, the lack of interfacial adhesion did not allow a good stress transfer from the matrix to the filler particles and no increase in tensile strength was obtained.

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In this paper, we consider the possibility of enhancing the tensile strength of $CaCO_3$ filled HDPE without embrittlement of the material by creating a soft, adhesive interphase. For this purpose, SEBS (Styrene-Ethylene/Butylene-Styrene) tribloc copolymers were used. These elastomer copolymers have already been used as interfacial agents in polymer blends. Previous works [17, 18] have shown their efficient compatibilizing effect in PE/PS blends. In order to create a soft interphase in $CaCO_3$ filled HDPE, two SEBS copolymers were selected. They have the same structure but one is grafted with maleic anhydride functions and the other is not. The grafted one is expected to give rise to better interactions with the calcium carbonate particles. Some authors [19, 20] have already reported the possibility of enhancing interfacial adhesion by the addition of an elastomer grafted with appropriate reactive functions.

The aim of this paper is to contribute to the understanding of the effect of elastomers on the tensile and impact properties of $CaCO_3$ filled polymers according to their location in the system (at the interface or dispersed in the matrix). Particular attention was paid to the evolution of tensile strength when the elastomer acts at the interface and improves interfacial adhesion.

2. Experimental

2.1. Materials

The matrix used was a high density polyethylene HDPE 2003SN53 supplied by Elf Atochem. Four calcium carbonate mineral fillers were incorporated in the HDPE matrix. The first one was an ultrafine $CaCO_3$ supplied by ECC with the commercial designation Polcarb. The other three, having different average diameters, were supplied by Omya with the commercial designations Hydrocarb, Millicarb and BL24. Details about the fillers characteristics are given in Table I. The two elastomers used were triblock copolymers supplied by Shell. They have the same structure, with a poly(ethylene-butylene) midblock ($M_w = 37500$) and two polystyrene endblocks ($M_w = 7000$). The former, called SEBS, commercialized as Kraton G1652 is not functionalized. The latter, called MAgSEBS, commercialized as Kraton FG1901X is grafted with 2% by weight of maleic anhydride functions on the central block.

2.2. Processing

All the compounds were prepared in a Clextral BC21 co-rotating twin-screw extruder. HDPE and SEBS elastomer pellets were mixed and then introduced into the first feed section. The mineral filler was introduced separately into the second feed section at the melting zone. Mixing conditions were carefully controlled and kept constant for all compounds. The mixing temperature and the screw rotation speed were 200 °C and 300 rpm respectively. These compounds were injection molded in a Sandretto injection molding machine at 220 °C in the form of dog-bone specimens for tensile tests. The injection conditions were also kept constant for all compounds. All the specimens have the same preparation history (thermal and mechanical).

2.3. Characterization

Tensile tests were performed at a cross-head speed of 100 mm/min by means of an Adamel Lhomargy apparatus. Tensile yield stress was determined from the recorded force vs elongation curve. For each material, an average of 10 measurements was taken. All measurements were made at room temperature. Scanning electron microscopic (SEM) observations were carried out on fracture surfaces. Before fracturing, samples were frozen for 5 min in liquid nitrogen. The fracture surfaces were coated with gold under vacuum and then examined using a JEOL JSM-35-CF apparatus. The measurements of the dynamic rheological properties of HDPE and SEBS elastomers were carried out by means of a Metravib Viscoanalyser. The dynamic viscosity was measured by the annular shearing technique on molten polymer within a range of shear rates from 5 to 250 s^{-1}. The temperature was stabilized at 200 °C corresponding to the extrusion temperature.

3. Results and discussion

3.1. Effect of the elastomers on tensile yield stress

3.1.1. In HDPE matrix

It was necessary to study the effect of each SEBS elastomer in the unfilled HDPE matrix before investigating their effects on the more complex filled HDPE system. The incorporation of both grafted and ungrafted SEBS lowers the tensile yield stress of HDPE as shown in Fig. 1. This is in accordance with the results obtained by Pukanszky *et al.* [21] which also noticed a decrease in tensile yield stress with increasing EPDM elastomer content in PP. Gupta *et al.* [22] reported the same effect on PP tensile yield stress with the addition of SEBS elastomer. According to Gupta *et al.*, the porosity model describes reasonably well the lowering in tensile yield stress with increasing content of SEBS elastomer in PP matrix. This model, proposed by Nielsen [2] is:

$$\sigma_y = \sigma_{ym} \cdot \exp(-a \cdot P)$$

TABLE I Characteristics of calcium carbonate fillers

Commercial designation of filler	Polcarb	Hydrocarb	Millicarb	BL24
Producer (carrier)	ECC (Salisbury, UK)	Omya (Orgon, F)	Omya (Orgon, F)	Omya (Orgon, F)
Average particle diameter (μm)	1.2	1.5	3.6	32
Specific surface area (m^2/g)	7	5	2.2	<1

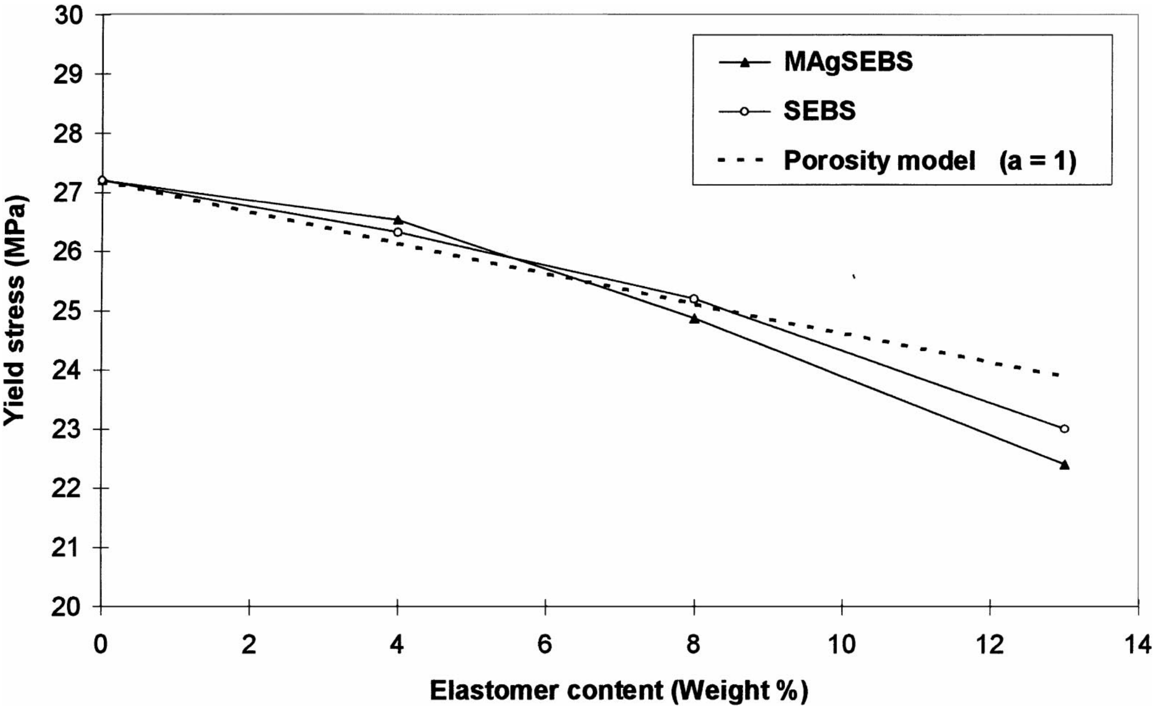


Figure 1 Effect of SEBS and MAgSEBS on HDPE tensile yield stress.

It considers the dispersed phase as voids which do not carry any load and lower the tensile strength. The porosity P of the material is equal to the volume fraction of the dispersed phase. σ_{ym} is the tensile yield stress of the matrix. Kunori *et al.* [23] suggested that the value of a is an indicator of stress concentration around the dispersed phase. It increases with stress concentration.

Fig. 1 shows that for $a = 1$, the theoretical curve fits the experimental plot rather well up to 8% by weight of SEBS and MAgSEBS elastomers. Considering these results, we can assume that HDPE containing dispersed elastomer has mechanical behavior similar to that of a porous material. When dispersed in the matrix, the effect of elastomer on tensile yield stress can be compared to that of holes or voids.

In polycarbonate blended with an increasing amount of dispersed HDPE, Kunori *et al.* [23] found a good correlation with the porosity model for $a = 4$. Gupta *et al.* [22] obtained a value of parameter a equal to 1.7 when SEBS was dispersed in PP. Therefore, the low value of a (equal to 1) indicates that both SEBS and MAgSEBS create low stress concentrations in HDPE.

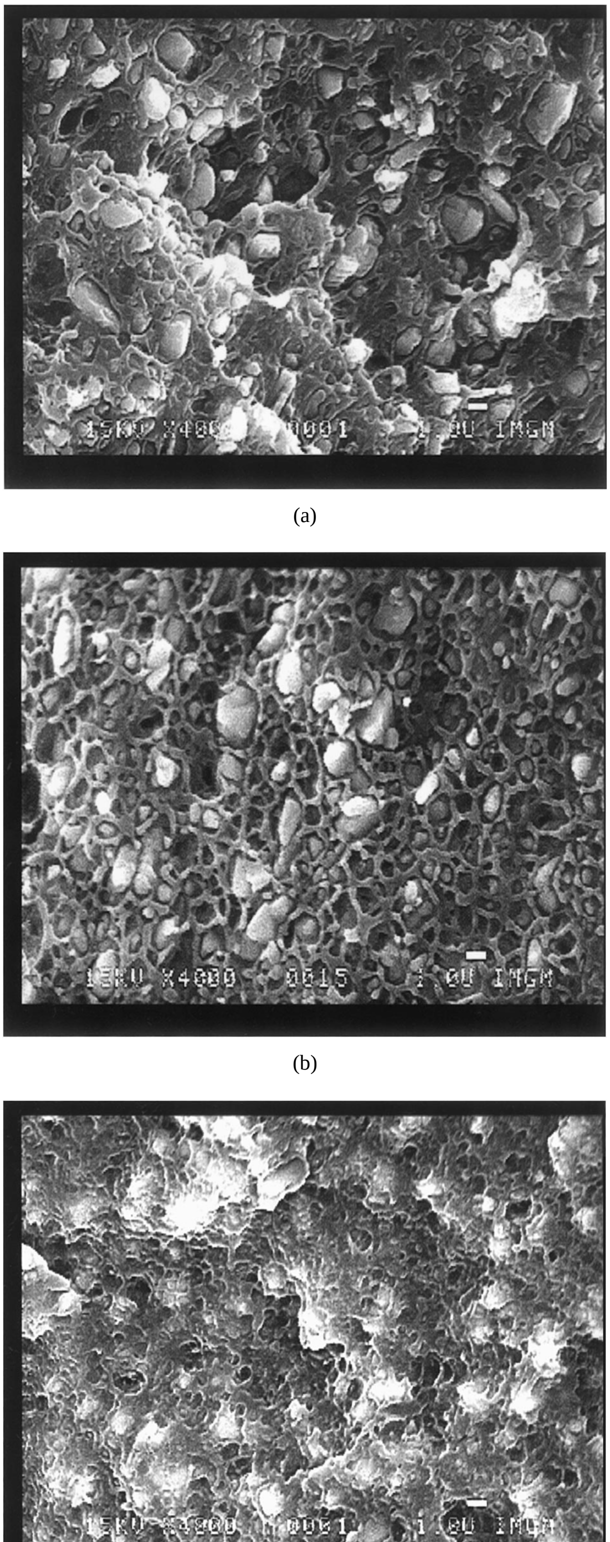
3.1.2. In $CaCO_3$ filled HDPE

3.1.2.1. Structure. Because of the non-polar nature of the matrix, no strong polar interactions take place between the matrix and the filler particles surface in $CaCO_3$ (Polcarb) filled HDPE. Effectively, S.E.M. observations of fractured samples show poor adhesion between the matrix and the dispersed filler particles, as shown in Fig. 2a.

The effect of the elastomers in $CaCO_3$ filled HDPE may be more complex than that observed in the unfilled HDPE. As described by some authors [24, 25], matrix/filler/elastomer ternary composites may exhibit two different phase structures, as shown in Fig. 3: (1) filler particles and elastomer inclusions are dispersed separately, (2) filler particles are encapsulated by the elastomer. According to Marosi *et al.* [26], the formation of an elastomer layer around the filler particles during melt-processing is controlled by kinetic and thermodynamic parameters. The kinetic control is governed by the viscosity ratio between the elastomer and the matrix. It favours the wetting of the filler particles by the polymer with the lowest viscosity. Thermodynamic control is governed by the elastomer-filler and matrix-filler interactions. It favours the wetting of the filler particles by the polymer with the strongest interactions with the filler surface.

The viscosity of HDPE and SEBS elastomers was measured using separate rheological equipment. However, the viscosity of a polymer during compounding operations depends strongly on the shear conditions and especially on the shear rate encountered in the machine. Thus, the viscosity ratio also depends on the shear conditions. Based on the equation developed by Burkhardt and coworkers [27] taking into account the screw characteristics, Wu [28] considered that the value of the effective shear rate in a co-rotating twin screw extruder is in the order of magnitude of the screw rotation speed. This latter is generally within the range from fifty to several hundred rpm. Considering the crude nature of these estimates, Wu chose the value of $100\,\mathrm{s}^{-1}$ (corresponding to 100 rad/s) as roughly corresponding to the shear rate encountered in a twin screw extruder. Based on these considerations, we have given in Table II, three values of the viscosity ratio in a range of shear rates around $100\,\mathrm{s}^{-1}$ from 80 to $250\,\mathrm{s}^{-1}$. The plots of the dynamic viscosity η' vs shear frequency ω at 200 °C for the HDPE and the two SEBS elastomers are given in Fig. 4.

The rheological results reported in Table II shows that the viscosities of the ungrafted SEBS and the HDPE matrix are quite close in the range of shear rates



(a)

(b)

(c)

Figure 2 Micrographs of HDPE/$CaCO_3$ (2a), HDPE/SEBS/$CaCO_3$ (2b), HDPE/MAgSEBS/$CaCO_3$ (2c).

around 100 s^{-1}. Moreover, both have a non polar nature. It is therefore difficult to predict the structure of the ternary system HDPE/SEBS/$CaCO_3$. As observed using S.E.M. micrographs in Fig. 2b, the incorporation of ungrafted SEBS apparently causes no modification at the HDPE-$CaCO_3$ interface. The $CaCO_3$ particles appears denuded with clean surface. It may be assumed that most of the ungrafted SEBS is separately dispersed in the matrix. However, no conclusions can be made about the final structure of this ternary system because it is also possible that one part of the SEBS is located at the interface without any adhesion to the filler particles.

TABLE II Values of the dynamic viscosity and viscosity ratio at various shear rates

	Dynamic viscosity η' (Pa·s)			Viscosity ratio $\eta'_{\text{elastomer}}/\eta'_{\text{HDPE}}$		
Shear rate (s^{-1})	80	120	250	80	120	250
HDPE	318	240	130	—	—	—
SEBS elastomer	314	222	103	0.99	0.92	0.79
MAgSEBS elastomer	222	164	85	0.70	0.68	0.65

One the other hand, the addition of maleic anhydride grafted SEBS significantly improves the interfacial adhesion. The filler particles are completely covered by polymer as shown in Fig. 2c. The improvement in adhesion by MAgSEBS requires firstly the wetting of $CaCO_3$ particles by the elastomer and secondly the establishment of strong interactions between the elastomer and both the matrix and the surface of the $CaCO_3$ particles.

The rheological results in Table II shows that the viscosity of MAgSEBS is slightly lower than that of the HDPE whatever the shear rate. Moreover, MAgSEBS has a polar nature while HDPE is non-polar. Thus, both thermodynamic and kinetic controls predict the encapsulation of the $CaCO_3$ particles by MAgSEBS.

The adhesive action of the MAgSEBS may be ascribed to the following phenomena: (a) the alkaline nature of the $CaCO_3$ and the acidic nature of the maleic anhydride function leads to strong acid-base interactions [24] and to the formation of ionic bonds between the elastomer and the filler surface, (b) the interdiffusion and entanglement of the poly(ethylene-butylene) segments with the polyethylene macromolecules may lead to a good physical anchoring between the MAgSEBS and the HDPE matrix. Both effects may contribute to the efficient action of MAgSEBS as adhesion promotor in $CaCO_3$ filled HDPE.

3.1.2.2. Effect of the elastomers on tensile yield stress.

Depending on the type of SEBS used (grafted or ungrafted), the differences in elastomer-filler interactions lead to differences in the final structure of the ternary composite. This also results in different effects on the mechanical properties of $CaCO_3$ filled HDPE. Tensile tests were carried out on $CaCO_3$ (Polcarb) filled HDPE containing increasing amount of grafted and ungrafted SEBS (0, 1, 4, and 8% of the total weight) at constant filler loading (40% of the total weight). The results are given in Fig. 5.

When ungrafted SEBS is added to $CaCO_3$ filled HDPE, a small increase in yield stress is measured up to 1% content. However, the yield stress value remains lower than that of pure HDPE (27.2 MPa). Above 1%, there is a continuous decrease in yield stress with increasing content of ungrafted SEBS. This continuous decrease is attributed to the dispersion of the elastomer in the matrix which entails a lowering of the tensile strength, as observed in pure HDPE.

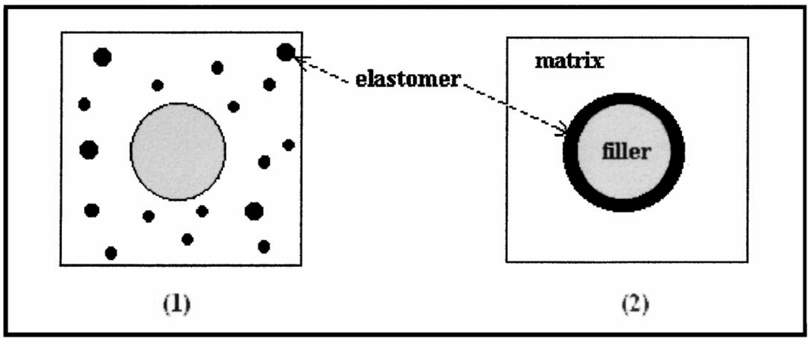


Figure 3 Extreme phase structures of a matrix/elastomer/filler system.

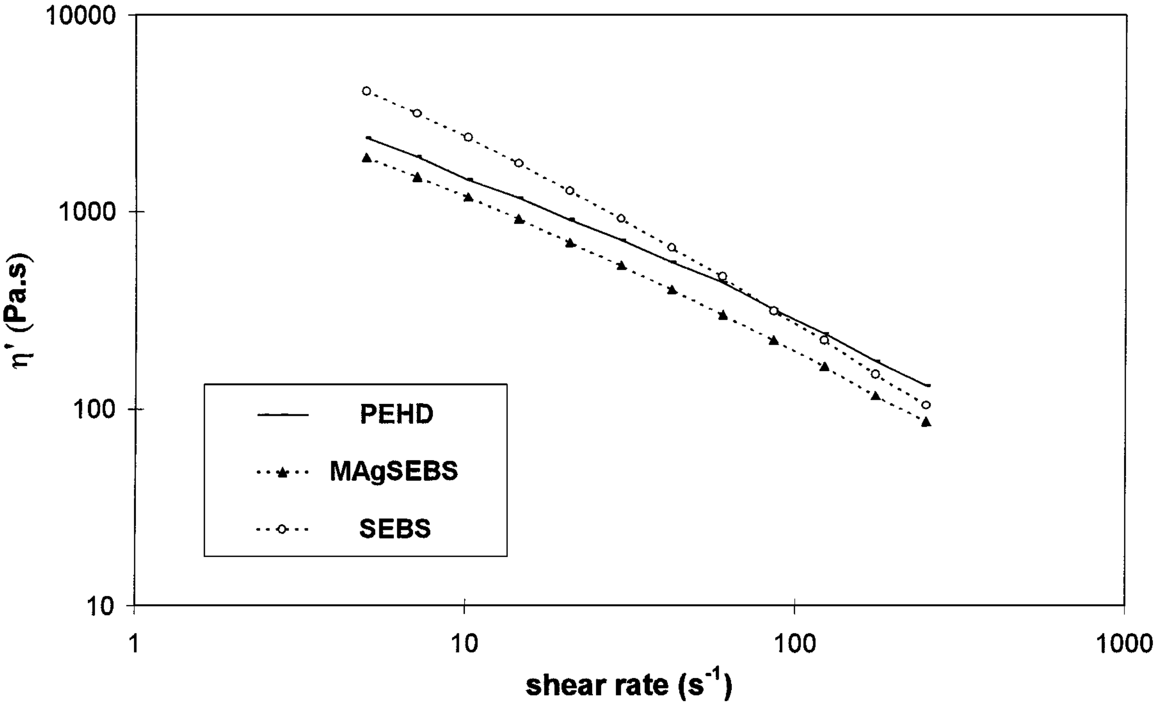


Figure 4 Dynamic viscosity vs ω for HDPE and SEBS elastomers at 200 °C.

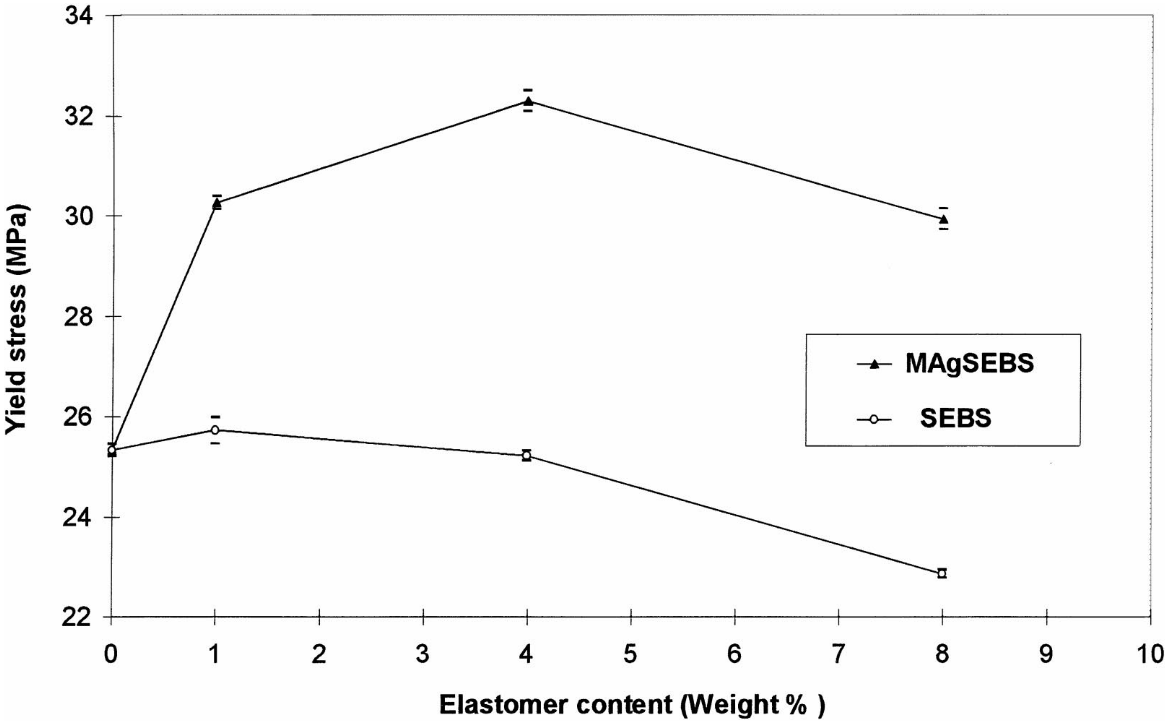


Figure 5 Effect of SEBS and MAgSEBS on HDPE/Carb1 (60/40) tensile yield stress.

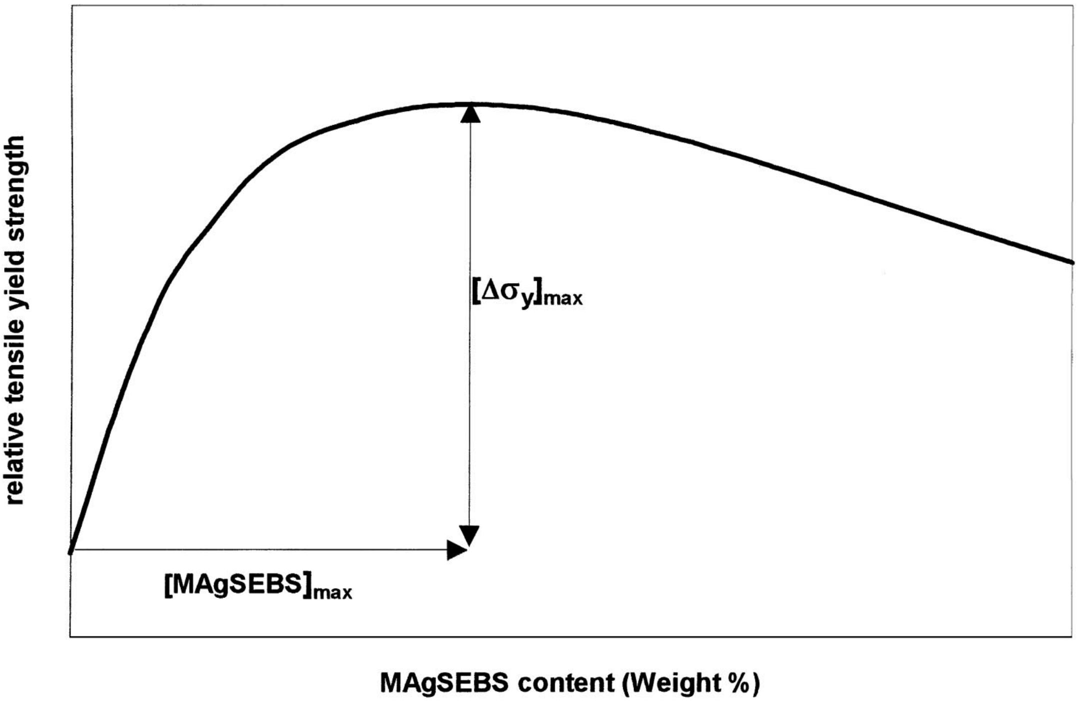


Figure 6 Schematic representation of $[MAgSEBS]_{max}$ and $[\Delta\sigma_y]_{max}$.

The small increase in yield stress at low SEBS content may be explained either by a better stress transfer at the interface or by a reduction in stress concentration around filler particles. It can not be attributed to the former because no improvement in interfacial adhesion was observed. It may therefore be explained by the latter because: a) stress concentration around the particles is partially responsible for the reduction of tensile strength [9] and b) stress concentration can be reduced by an elastomeric interlayer [29, 30]. This assumes that one part of the elastomer locates at the interface when SEBS is added up to 1%.

On the other hand, the effect of the grafted elastomer in HDPE/$CaCO_3$ (60/40) is very different. There is a considerable increase in yield stress with increasing content of MAgSEBS up to 4%. The yield stress of the ternary composite HDPE/$CaCO_3$/MAgSEBS is higher than that of pure HDPE (27.2 MPa). The load seems to be effectively transmitted from the matrix to the filler particles thanks to the adhesive effect of MAgSEBS. This leads to an increasing contribution of filler particles up to 4% of MAgSEBS. It is assumed that the thickness of the interphase increases until an optimum value at 4% MAgSEBS which correspond to the maximum stress transmission between the matrix and the filler and then to the maximum contribution of filler particles to the tensile strength. This supposes that the grafted SEBS locates primarily at the interface up to 4% weight fraction. Even if some of the grafted elastomer may be dispersed in the matrix, it is assumed that the most is at the interface because of its strong interactions with the filler surface. The decrease in tensile yield stress above 4% loading may be interpreted in two different ways. The MAgSEBS in excess may be dispersed in the matrix, thus lowering its tensile strength, as observed when MAgSEBS is added to the unfilled HDPE. It is also possible that the MAgSEBS in excess locates at the interface and increases its thickness. This thicker elastomer shell would become softer and less able to transfer the load to the embedded core particle.

Whatever the interpretation, we note that there is an optimum content of MAgSEBS corresponding to the maximum contribution of the filler particles. As indicated in Fig. 6, the optimum content of MAgSEBS will be given as $[MAgSEBS]_{max}$ and the maximum increase in yield strength will be given as $[\Delta\sigma_y]_{max}$.

We propose to study the dependence of $[MAgSEBS]_{max}$ and $[\Delta\sigma_y]_{max}$ on the weight fraction and the specific surface area of the mineral filler.

3.1.2.3. Influence of filler loading. Tensile tests were performed on HDPE filled with various weight fractions of $CaCO_3$ (10, 20 and 40% of the total weight). For each system, an increasing amount of MAgSEBS (0.5, 1, 2, and 4% of the total weight) were added.

The plots in Fig. 7 give the evolution of the relative tensile yield stress (modified/unmodified system) versus MAgSEBS content at different filler loadings. It clearly shows that $[MAgSEBS]_{max}$ increases with the filler loading. In HDPE filled with 10, 20, and 40% of $CaCO_3$ filler, the maximum yield strength is reached with $[MAgSEBS]_{max}$ values approximately equal to 1.1, 2.1, and 3.4% respectively, as shown in Table III.

TABLE III Influence of the weight fraction of calcium carbonate (Polcarb) on $[SEBSgAM]_{max}$ and $[\Delta\sigma_y]_{max}$ values – Estimates from the results given in Fig. 7.

Weight fraction of $CaCO_3$ (%)	10	20	40
$[SEBSgAM]_{max}$ (weight %)	1.1	2.1	3.4
$[\Delta\sigma_y]_{max}$ (relative increase %)	7	14	28

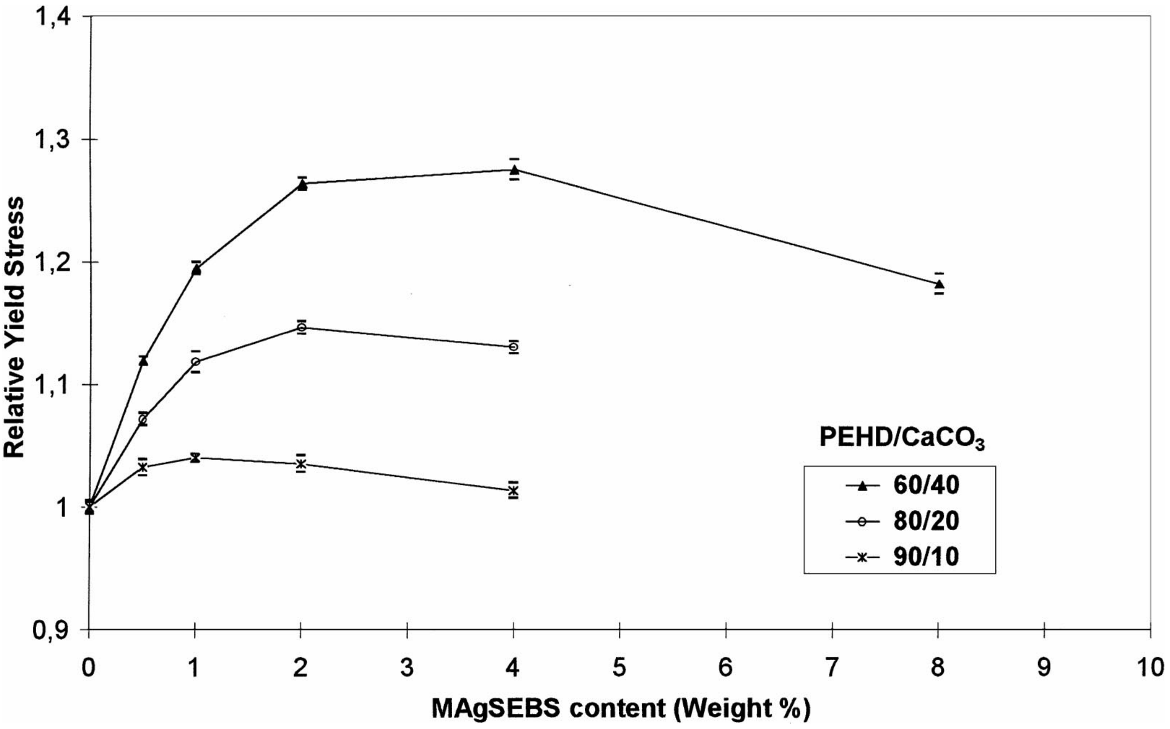


Figure 7 Effect of MAgSEBS on the relative tensile yield stress of HDPE/Carb1 with different filler loading.

The weight fraction of SEBSgAM needed to reach the maximum contribution of the filler ($[\text{MAgSEBS}]_{\text{max}}$) increases almost linearly with the filler loading. The maximum contribution of the filler particles is obtained for an approximately constant weight ratio MAgSEBS/$CaCO_3$ close to 1/10.

The results given in Table III also show a linear increase in $[\Delta\sigma_y]_{\text{max}}$ with the filler loading. Therefore, the potential contribution of the filler particles to the tensile yield strength of the composite material is proportional to the filler loading. This shows that the increase in tensile yield strength entailed by an improvement in interfacial adhesion depends strongly on the filler loading.

Note that any increase in the weight fraction of filler entails a proportional increase in both the surface and the volume of filler in the system. Then the increase in $[\text{MAgSEBS}]_{\text{max}}$ and $[\Delta\sigma_y]_{\text{max}}$ with the filler loading may be attributed to a surface or volume effect. For example, it may be supposed that $[\text{MAgSEBS}]_{\text{max}}$ is proportional to the interfacial area to be covered by MAgSEBS in the system, and thus proportional to the filler weight fraction. However, before coming to any conclusions about the influence of the surface or volume effect of filler, we propose studying, in the following part, the influence of the interfacial area without variation in the volume of filler.

3.1.2.4. Influence of filler specific surface area. In order to study only the influence of the interfacial area without variation in the volume of filler, we studied the effect of MAgSEBS on systems filled with $CaCO_3$ fillers of different specific surface at constant weight fraction.

For this purpose, three $CaCO_3$ fillers (Hydrocarb, Millicarb and BL24) with the same origin (same producer and the same source) but different size distribution were selected. The tensile mechanical properties of each HDPE/$CaCO_3$ (60/40) system without interfacial agent is given in Fig. 8. The results obtained with Polcarb are also given.

The tensile yield stress decreases slowly when particle diameter (d) increases for fillers from the same source. This is in agreement with the results reported by Alter [1] and later by Leidner *et al.* [3]. Alter proposed an empirical equation in which the tensile stength varies with d^{-1}. According to Leidner *et al.*, tensile strength varied with $d^{-1/2}$. However no attempt was made to propose a relation or to compare our results with the existing empirical equations. It seems difficult to find an appropriate mathematical relation between tensile strength and average particle diameter without taking into account the particle size distribution and the specific surface of the filler.

The plots in Fig. 9 show the evolution of the relative tensile yield stress versus MAgSEBS in HDPE/$CaCO_3$ (60/40) systems containing fillers of different specific surface area. The results given in Table IV clearly shows that $[\text{MAgSEBS}]_{\text{max}}$ and $[\Delta\sigma_y]_{\text{max}}$ increases with the specific surface area of filler particles. However, a high increase in the specific surface area of the filler causes only a small increase in $[\text{MAgSEBS}]_{\text{max}}$ and $[\Delta\sigma_y]_{\text{max}}$. Therefore, the proportional relation which was observed between both $[\text{MAgSEBS}]_{\text{max}}$ and $[\Delta\sigma_y]_{\text{max}}$ and the weight fraction of the filler can not be attributed only to a surface effect.

TABLE IV Influence of the specific surface area of calcium carbonate (Polcarb) on $[\text{SEBSgAM}]_{\text{max}}$ and $[\Delta\sigma_y]_{\text{max}}$ values at constant weight fraction of filler (40%) – Estimates from the results given in Fig. 9

Specific surface area of $CaCO_3$ (m^2/g)	7	5	2.2	<1
$[\text{SEBSgAM}]_{\text{max}}$ (weight %)	3.4	2.9	2.4	2.1
$[\Delta\sigma_y]_{\text{max}}$ (relative increase %)	28	25	23	17

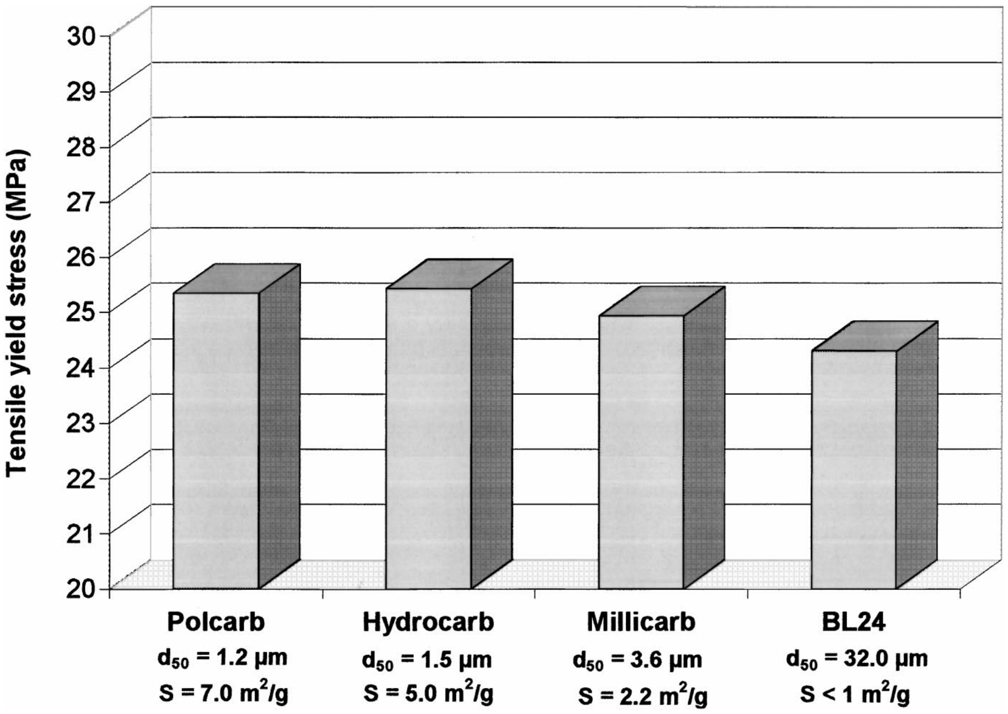


Figure 8 Tensile yield stress of HDPE/$CaCO_3$ (60/40) systems – Influence of calcium carbonate average size and specific surface area.

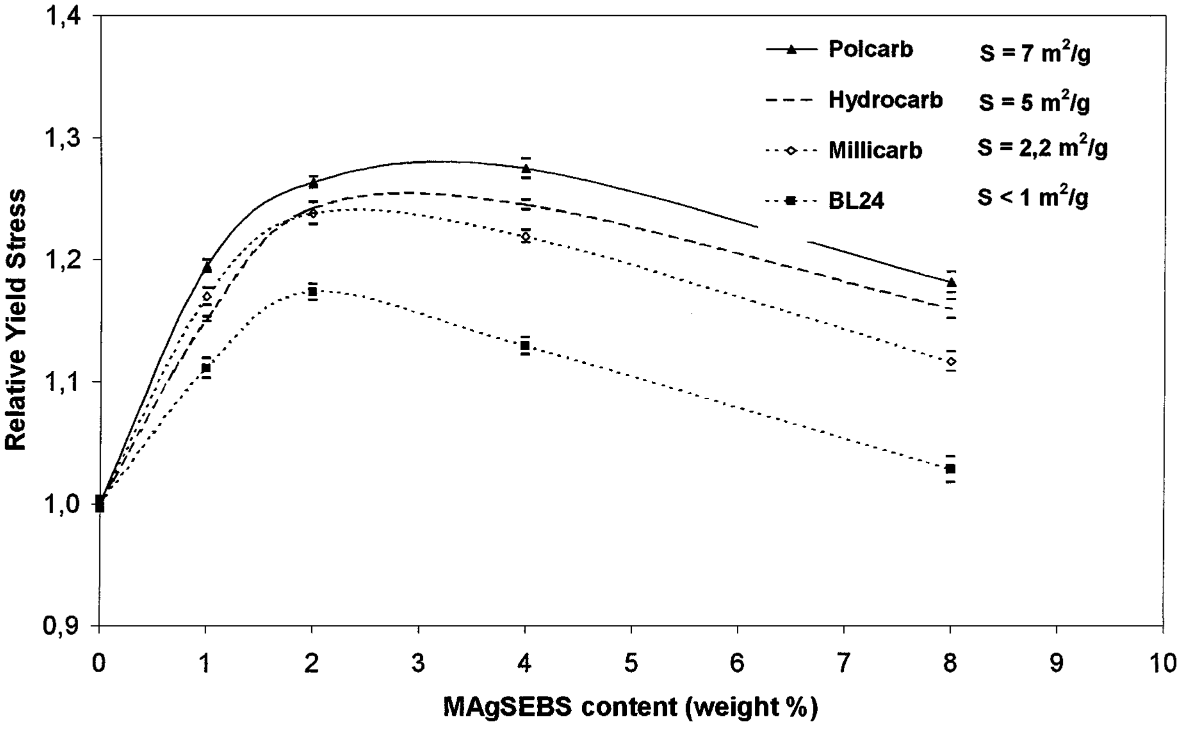


Figure 9 Effect of MAgSEBS on the relative tensile yield stress of HDPE/$CaCO_3$ (60/40) – Influence of the specific surface of filler.

When the weight fraction of the filler increases, the interfacial agent has to cover a higher interfacial area but must also involve a higher volume of filler. It is thus assumed that the pronounced dependance of $[\text{MAgSEBS}]_{\text{max}}$ on the weight fraction of filler is due to both a surface and a volume effect of the filler particles.

Therefore, the amount of interfacial agent necessary to reach the maximum yield stress $[\text{MAgSEBS}]_{\text{max}}$ and the extent of the increase in yield stress $[\Delta\sigma_y]_{\text{max}}$ depend on both the interfacial area and on the volume of filler which may potentially contribute to the final yield stress.

TABLE V Effect of MAgSEBS on mechanical properties of 40% $CaCO_3$ (Polcarb) filled HDPE

HDPE/$CaCO_3$/MAgSEBS (wt %)	60/40/0	56/40/4	52/40/8
Tensile yield strength (MPa)	25.3	32.3	29.9
Unnotched Charpy Impact strength (kJ/m^2)	95	123	164
Notched Charpy impact strength (kJ/m^2)	25	43	50

3.2. Effect of the elastomers on impact strength

It has been shown that the grafted and ungrafted SEBS have different effects on the tensile strength of

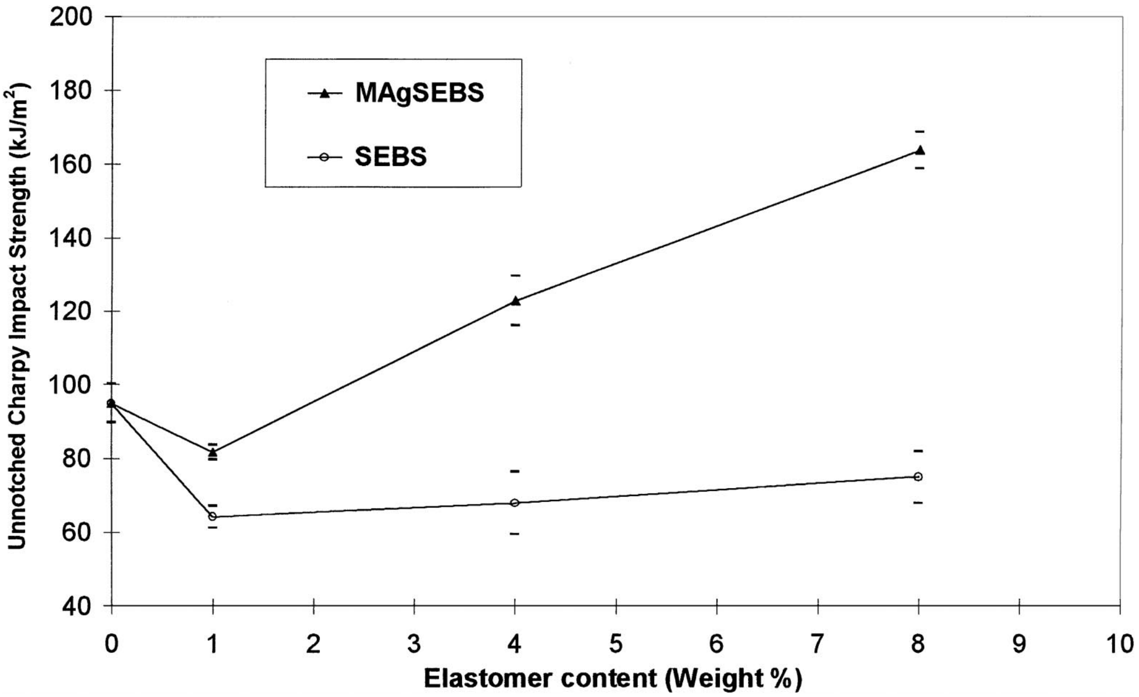


Figure 10 Effect of SEBS and MAgSEBS on HDPE/CaCO$_3$ (60/40) Charpy impact strength.

HDPE/CaCO$_3$. The results given in Fig. 10 show that these two elastomers have also different effects on impact strength. The grafted elastomer MAgSEBS, which encapsulates mineral particles, entails an increase in impact strength, while the ungrafted SEBS dispersed in the matrix gives no positive effect. These results show that the location of the elastomer has a pronounced influence on the tensile and impact properties of filled polymers. The formation of an elastomer interphase with high molecular mobility may act as a "bumper" interlayer around filler particles, which absorbs the impact energy and prevents the initiation of cracks. Moreover the adhesion created by the elastomer interphase may also prevent the propagation of cracks at the interface.

Conclusion

It is shown that interfacial adhesion may be improved by the addition of an elastomer which creates entanglements with the matrix and interacts with the filler surface. In order to increase both impact properties and the tensile strength of filled polymers, it appears interesting to create an adhesive elastomer interphase. When the amount of elastomer interfacial agent increases, the tensile yield stress is gradually enhanced until a maximum value corresponding to the maximum contribution of the filler. The tensile yield stress seems to be an interesting macroscopic property to follow the adsorption of the interfacial agent on the filler particles surface and the gradual modifications of the interface (i.e. surface coverage, degree of adhesion). It was observed that the extent of the increase in yield stress (i.e. filler contribution) with increasing amount of interfacial agent depends on both the interfacial area and the volume of filler. To obtain the highest potential contribution of fillers to the tensile strength, it is preferable to use fine particles and high filler loading. In this case, the amount of elastomer interfacial agent necessary to reach the maximum contribution of filler will also be high.

Moreover, these results show the necessity for modeling to take into account both the interfacial area and the volume of filler. It seems also important to take into account the degree of adhesion rather than the too extreme cases of poor and perfect adhesion.

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