

Effect of Low-Load Reinforcing Nanofillers on Rheological Behavior and Physical Properties of Ionic Elastomers Based on XNBR and MgO

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ABSTRACT: The influence of low load reinforcing fillers in carboxylated nitrile rubber (XNBR), crosslinked with magnesium oxide, has been studied. The nanofillers utilized were carbon black, precipitated silica, natural clay, and synthetic silica. The latter filler was obtained by acid lixiviation of a natural magnesium silicate. The rheological behavior of the samples was studied by means of capillary (consistency index) and rotational (Mooney viscosity) flows. Through these analyses, higher viscosities, and lower flow indexes were observed for the filled samples. The acid character of silica and clay leads to an increase in vulcanization rates, measured by the cure rate index (CRI)

and referring it to the unfilled compound. Moreover, the fillers dilute the ionic microphases formed during the vulcanization process of the XNBR. Since these microphases behave as reinforcing sites, a decrease in tear and abrasion properties takes place. However, the reinforcing effect of the fillers leads to higher values of stress at constant strains. Finally, the variation of glass and ionic transition temperatures with filler addition was analyzed. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 106: 973–980, 2007

Key words: ionic elastomers; nanofiller; rheological and physical properties

INTRODUCTION

The effect of fillers in processing and physical properties of conventional rubbers, e.g. NR, SBR, EPDM. . . , is well known. When these materials are not filled, they cannot be used in most of the applications.

When fillers are added to a rubber matrix, they are able to improve the mechanical properties, tensile strength, tear resistance, hardness, and abrasion resistance.^{1–4} This effect has been widely studied; however, its microscopic origin is still in discussion. Since there are two different phases, filler and rubber, it is obvious that the polymer–solid surface interaction and structure of the aggregates play a major role in the final properties of these compounds.

When high proportions of carbon blacks and silicas are added into elastomeric compounds, an increase in their final properties, e.g., hardness, stress at constant strain, tensile strength, and tear strength takes place. These properties depend on the

chemical nature of the elastomer, the hydrodynamic effect of the fillers, rubber–filler interactions, and interactions between filler particles. Because of the active surfaces of these fillers, organic groups in carbon black⁵ and silanol groups in silica,⁶ these fillers have high superficial activity, which lead to strong interactions between them and the rubber.

The addition of reinforcing fillers in rubber compounds has an additional effect in their rheological behavior.^{7–9} The structural properties of the fillers, as well as the rubber–filler and filler–filler interactions, lead to increases in blends viscosities determining the processing procedures (moulding, injection, extrusion, . . .).

Besides the conventional vulcanization systems, sulphur or peroxides, the XNBR (carboxylated nitrile rubber) can be crosslinked by means of divalent metallic oxides. This kind of crosslinking leads to the formation of ionic bonds through the carboxylic groups from the polymeric chains. As a result of this reaction, an ionic elastomer is formed.¹⁰ These materials present thermoplastic behavior. These associations behave as ionic microphases and act as reinforcing sites leading to high values for tensile properties and abrasion resistance.¹¹ These properties are not achieved when the XNBR is vulcanized with sulphur or peroxides.¹²

Our research group has been working on vulcanization systems for the XNBR. On one hand, when sulphur is employed, only covalent bonds are generated,¹³

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on the other hand, with zinc peroxide,^{14,15} which leads to the formation of ionic and covalent bonds. Furthermore, a mixed system of sulphur and zinc peroxide¹⁶ has been also studied. Alzorritz's works,¹⁷ focused on the crosslinking of XNBR by means of different metallic oxides, optimizing temperature and concentration, have proved that the crosslinking with magnesium oxide (MgO) leads to better final properties than the other metallic oxides. As it can be observed, the vulcanization system of these materials has been deeply studied. However, there are not many works based on the effect of fillers in this kind of elastomer.

In a recent work, Ibarra et al.¹⁸ has studied the effect of low load of layered nanoclays in XNBR. They demonstrate that these fillers have an important effect in the vulcanization characteristics, improving some properties, e.g., stress at constant strain and tear strength, and leading a lower swelling. These effects are the result of the exfoliation of the nanofiller sheets during the processing. However, some properties, like tensile strength and abrasion resistance, are better in the absence of fillers. This phenomenon is due to the dilution effect that these fillers have upon the ionic microphases. Therefore, physical properties of ionic polymers depend on, fundamentally, the ionic microphases formed during the crosslinking process. This formation is strongly affected by the presence of fillers.

In the present work, the influence of organic and inorganic fillers in the formation of the ionic crosslinks is studied for elastomeric systems based on XNBR crosslinked with MgO. The fillers studied are carbon black, commercial silica, commercial clay, and silica obtained in our laboratory (silsep). This silica comes from the acid lixiviation of natural magnesium silicate (sepiolite). Their influence in the rheological behavior and physical properties is also analyzed.

EXPERIMENTAL

Materials

The carboxylated nitrilic rubber is a Krynac 7.40, supplied by Lanxess Elastomers SAS (Germany). It contains 7% by weight carboxylic groups ($-\text{COOH}$) and a Mooney viscosity of 40 UM and a content of 27% in acrylonitrile groups.

The stearic acid used is rubber grade. The magnesium oxide is an Elastomag 170 Special, supplied by Akrochem (Ohio, USA).

The carbon black filler N-330 (Vulcan 3), with specific area BET of 83 m²/g and pH = 8.50, was supplied by Cabot (USA). Two silicas filler were used. The precipitate silica filler, supplied by Iqesil (Spain) named Ebrosil PD with specific area BET of 186 m²/g

TABLE I
Recipes of Compounds at phr
(Parts Per Hundred of Rubber)

| | C-1 | C-2 | C-3 | C-4 | C-5 |
|----------------------|-----|-----|-----|-----|-----|
| XNBR | 100 | 100 | 100 | 100 | 100 |
| Stearic acid | 3.0 | 3.0 | 3.0 | 3.0 | 3.0 |
| Magnesium oxide | 4.3 | 4.3 | 4.3 | 4.3 | 4.3 |
| Carbon black | – | 7.0 | – | – | – |
| Clay | – | – | 7.0 | – | – |
| Silica | – | – | – | 7.0 | – |
| Sil-sep ^a | – | – | – | – | 7.0 |

^a silica from sepiolite.

and pH = 6.12, and silica obtained in our laboratory¹⁹ by acid lixiviation of socialite. Sepiolite was supplied by Tolsa S.A. (Spain). The specific area of the silica obtained from sepiolite was 395 m²/g and pH = 5.30. The clay filler (pH = 5.12) was supplied by Caobar (Spain), and it is named as Caosil SP-SE. Its specific area BET was 15.5 m²/g.

Compounding

The compounds were prepared on a Gumix laboratory cylinder mixer, with a cylinder diameter of 15 cm and a length of 30 cm. The cylinders were kept cold by circulating cold water through them. The friction relation between them was 1 : 1.15. Table I shows the used recipes of this work.

Testing of raw materials

Curing of the different blends was followed on an MDR 2000E oscillating disc rheometer, from Alpha Technologies (Wilshire, UK), with an oscillation rate of 1.63 Hz and strain amplitude of 6.98%. The reaction temperature was set at 150°C. The rheometer used allows torque decomposition into its two components, the elastic S' , and the viscous component, S'' . The plot of S' variation, i.e. the elastic torque component, as a function of time is taken in the crosslinking graphs.

Rheological measurements were performed in a Rheograph 2003 capillary rheometer from Göttert-Werkstoff-Promaschinen (Karlsruhe, Germany) at 90°C with a shear rate range between 2 and 500 s⁻¹. The used die had a 30/2 length/diameter ratio and an entrance angle of 180°. Mooney viscosities (ML₂₊₄) were determined at 90°C and 2 rpm (shear rate = 1.58 s⁻¹) using a viscosimeter model MV-2000E from Alpha Technologies (Swidon, UK).

Testing of crosslinked materials

The vulcanization reaction was carried out in a hydraulic press heated by electric resistances at 150°C,

and the curing time was the t_{90} determined from rheometer curves.

Shore A hardness was determined by using an automatic Bareiss Durometer on cylindrical samples, according to UNE EN ISO 868, after 15 s of indentation.

Tensile tests were performed with an Instron model 4301 dynamometer equipped with a video camera to measure deformations, at a deformation rate of 500 mm/min on Type 2 dumbbell samples according to UNE 53510.

Tearing tests were carried out on angular-type samples in agreement with UNE 53516. Part 1 at the same deformation rate used for the tensile test. Five samples were tested in each use.

Abrasion resistance, and compression set were determined according to national standards.

The dynamic properties were determined with a Rheometric Scientific DMTA V dynamic mechanical thermal analyzer working under norm ISO 6721 in the tensile mode at frequencies of 5, 10, 15, and 20 Hz. The temperature was varied from -80°C to 200°C at a rate of 1.5 K/min. Each specimen for dynamic mechanical experiments was cut out into a rectangular strip with approximate dimensions $30 \times 6 \times 2 \text{ mm}^3$.

Scanning electron microscopy (SEM) analysis has been carried in an ESEM Philips XL30 microscope, operating at high vacuum mode and 30 kV. The SEM micrographs ($5000\times$) were taken from the newly exposed surface of the rubber specimens fractured immediately after embrittling them in liquid nitrogen. The specimens were coated with Au-Pd to prevent charging on the surface.

RESULTS AND DISCUSSION

Vulcanization characteristics of the compounds

The values for the rheometric parameters obtained from vulcanization curves, at 150°C , are shown in Table II. The presence of the filler leads to an increase in the values of the maximum torque (S'_{max}), with regard to the unfilled compound. The maximum values are obtained for the compounds filled with carbon black and silicas. The influence in the

TABLE II
Cured Parameters at 150°C

| | C-1 | C-2 | C-3 | C-4 | C-5 |
|------------------------------------|-------|-------|-------|-------|-------|
| S'_{max} , dNm | 6.68 | 8.20 | 7.09 | 7.29 | 7.82 |
| S'_{min} , dNm | 0.75 | 0.73 | 0.70 | 0.79 | 1.03 |
| $\Delta S'$, dNm | 6.15 | 7.47 | 6.39 | 6.49 | 6.79 |
| $t_{\Delta 2}$, min | 7.03 | 10.76 | 11.15 | 5.47 | 6.47 |
| t_{90} , min | 42.09 | 53.58 | 44.00 | 35.68 | 33.06 |
| CRI^a , min^{-1} | 2.85 | 2.31 | 3.03 | 3.30 | 3.76 |

^a $\text{CRI} = 100/t_{90} - t_{\Delta 2}$.

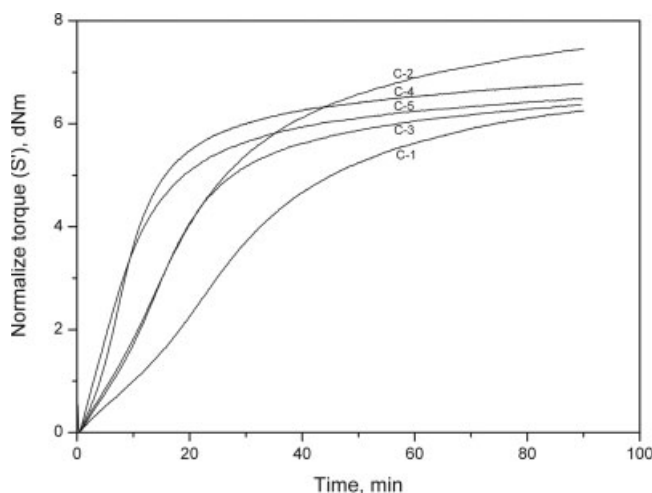


Figure 1 Normalized rheograms curves of compounds at 150°C .

minimum torque values (S'_{min}) of the low-load of the filler is not significant. This parameter can be related to the viscosity of the sample, and only in the compound C-5 (filled with silica from sepiolite, with high specific surface area), higher S'_{min} values are obtained, leading to a compound with higher viscosity.

The curing rate index (CRI) of the compounds is affected by the pH of the fillers. On one hand, the basic character of carbon black ($\text{pH} > 7$) has many different consequences in the vulcanization reaction: a decrease in the formation rate of the ionic crosslinks, an increase in the induction time, $t_{\Delta 2}$, and an increase in the vulcanization optimum time, t_{90} . All these variations are referred to the unfilled compound.

On the other hand, the fillers with acid pH, especially the silica obtained from sepiolite, affect the cure process increasing vulcanization rate and decreasing induction and optimum time.

Figure 1 shows the normalized vulcanization curves of the compounds. Considering that the maximum normalized torque, S'_N ($S'_N = S'_{\text{max}} - S'_{\text{min}}$) is a measure of the crosslinking density, it can be observed that, in the presence of the filler the crosslinking increases. The higher increase is obtained when carbon black is used due to its active sites of organic nature, present in the surface of filler particles.⁵ These filler characteristics improve the compatibility with the elastomer, leading to stronger filler-rubber interactions during the vulcanization process, and, consequently, achieving higher values for the normalized torque S'_N .

Viscoelastic properties of the unvulcanized compounds

Rheological parameters are shown in Table III. They were obtained by means of rotational and capillary

TABLE III
Rotational and Capillary Flow Rheological Parameters

| | C-1 | C-2 | C-3 | C-4 | C-5 |
|-----------------------------------|-------|-------|-------|-------|-------|
| Rotational parameters | | | | | |
| Initial viscosity, Nm | 4.25 | 4.48 | 4.43 | 4.83 | 5.99 |
| Mooney viscosity, Nm | 2.69 | 2.95 | 2.78 | 3.28 | 3.76 |
| Δ viscosity, Nm | 1.56 | 1.53 | 1.65 | 1.55 | 2.23 |
| Capillary flow parameters | | | | | |
| Flor index, (n) | 0.354 | 0.314 | 0.311 | 0.301 | 0.342 |
| Consistency index, (k) kPa | 96.8 | 118.4 | 123.7 | 143.1 | 189.9 |
| Correlation coefficient (R^2) | 0.993 | 0.994 | 0.995 | 0.996 | 0.996 |

flow. Both methods are usually employed in the evaluation of processing properties of elastomers compounds.

The rotational flow has been carried out in a Mooney viscosimeter, at low deformation rate (1.58 s^{-1}). When filler is added to a polymer matrix, an increase in the initial viscosity takes place. This increase is due to, fundamentally, three factors: (i) filler–filler interactions, (ii) filler–polymer interactions, which reduce chain mobility, and (iii) hydrodynamics effect of a solid particle (the filler) in a fluid media. The presence of fillers in XNBR compounds produces an increase in both, initial viscosity and Mooney viscosity. This effect is even more significant in the case of the sample filled with the silica obtained from sepiolite. Its high specific surface ($\approx 400 \text{ m}^2/\text{g}$) favors the creation of many interactions between filler particles, even though when the proportion of the silica in the rubber matrix is very low. The viscosity drop observed in the compound C-5 is 40% superior if it is compared with the other components. This fact can be explained if the breakage of the interactions between filler particles takes place during the rotational flow process is assumed.

In the capillary flow, the material is subjected to high shear rates, or deformation rates, if we compare it with the rotational flow. Figure 2 shows the viscosity curves of the compounds, in a wide range of shear rates. The behavior for all of them is similar, and it corresponds with the behavior expected for pseudoplastics materials, i.e., the viscosity decreases when the shear rate increases. Filler addition produces an increase in the viscosity for the entire shear rates studied, regarding to the unfilled sample. This effect is less significant at high shear rates. The compound filled with silsep is the one showing higher viscosity differences in the entire shear rate sweep. In this case, the amount of “free rubber” (rubber that is not directly interacting with filler particles) is lower due to the high surface area of the filler produce stronger interaction with matrix rubber.

By applying potential equation $\eta = K\dot{\gamma}^{n-1}$ to the experimental values of the viscosity and shear rates, η and $\dot{\gamma}$, we can obtain the consistency index, K , related to the viscosity of the compound, and the exponent of the potential equation, n , as a measure of the flow index. These values are shown in Table III. Again, the presence of the filler in the elastomeric matrix gives lower flow index values and higher elastic component, as a result of filler–rubber interactions. On the contrary, the compound C-5 gives a value similar to the one obtained for the unfilled compound. This can be explained if we consider the fibrillar morphology of this filler, which remains unaffected during the lixiviation process of the sepiolite.²⁰ These fibers are orientated in the flow direction, improving the processing properties of the material. Also, consistency indexes of the samples increase with filler addition.

Figure 3 shows the photographs of the compounds extruded by capillary flow, at different shear rates, 10 and 100 s^{-1} . As it can be observed, at low shear rate, the extruded samples show similar swelling, with a smooth texture without superficial defects. At high shear rates, the extruded compounds flow distorted with defects that can break the melted sample. However, the compound C-5 shows a smooth surface even at shear rates higher than 100 s^{-1} . This improvement in the processing behavior is due to the orientation of the fibrillar particles of the silsep in the flow direction.²¹

Physical properties of vulcanized compounds

Mechanical properties

Table IV shows the values obtained for the physical properties of the compounds. As it was expected, the presence of the filler produces an increase in Shore A hardness and in the stress values at low

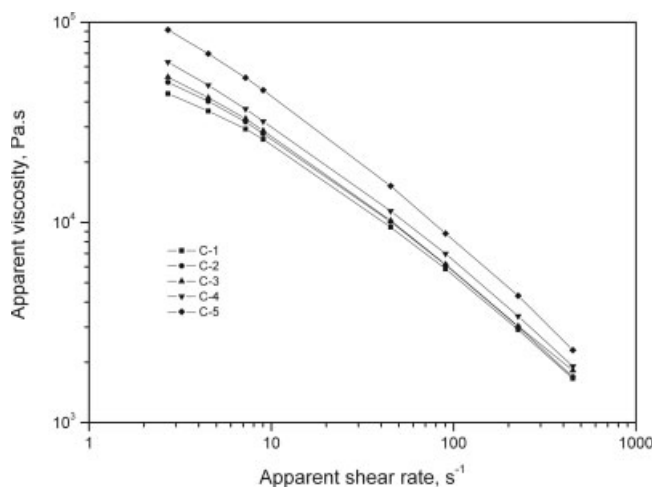


Figure 2 Variation of apparent viscosity with shear rate on capillary flow.

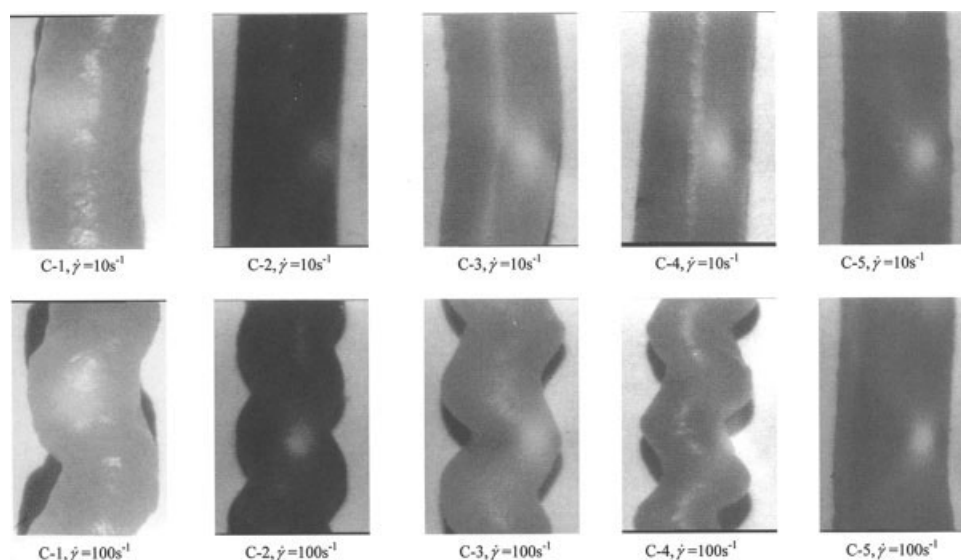


Figure 3 Optical photograph of extrudates of compounds at different shear rates.

deformations. The effect is more significant in the sample with active fillers, i.e., C-2, C-4, and C-5. The compound filled with carbon black, C-2, shows the highest values as a consequence of the higher compatibility between carbon black and rubber, which leads to higher rubber–filler interactions and, therefore, higher reinforcing effect on the vulcanized samples. Compounds C-4 and C-5, which are filled with commercial silica and silsep, respectively, also lead to high interactions with the XNBR, due to their high surface area. However, the compound C-3, with the clay filler, shows values similar to the unfilled compound due to its low specific and surface activity.

It must be kept in mind that ionic elastomers possess, even without the addition of fillers, some exceptional mechanical properties, due to their ionic microphases formed during crosslinking, which behave as reinforcement sites. This increase in mechanical resistance is reflected, especially, in tensile strength and in abrasion resistance.

Tear strength is also improved when fillers are added. As the superficial activity of the fillers increase, the values for tear strength improve. Tensile or deformation at break achieves maximum values in the unfilled compound (C-1), as it can be observed in Table IV. The decrease with filler addition is probably due to the dilute effect of the fillers upon the ionic microphases.¹⁸ The inorganic nature of the fillers in the compounds C-3, C-4, and C-5, would lead to a higher dilution effect, giving higher values for abrasion losses.

Dynamics properties

In the measure of dynamic properties of cured materials, two types of experiments were carried out:

(a) deformation sweep at constant frequency; and (b) temperature sweep at different frequencies, maintaining the strain constant.

Figure 4 shows the variation in the complex modulus, E^* , with double strain amplitude (DSA) and 12 Hz of frequency. For all compounds, a decrease in E^* with deformation is observed, however, the values of the modulus is always higher than the ones obtained for the unfilled compound.

In the same figure, the values of an unfilled sample of XNBR vulcanized with sulfur are plotted. For this sample, the value of E^* remains constant during the strain sweep. This would be the expected behavior for an unfilled compound. However, if we regard to the compound unfilled C-1, a decrease in the modulus is observed when deformations higher than 1% are achieved. This response can be explained by the breakage of the ionic associations, which behave as reinforcing sites. This behavior is also observed in filled compounds (but with higher modulus values) and it is due to the breakage of the filler network.

It is well known that the modulus is dependent on (i) the crosslinking density of the rubber matrix,

TABLE IV
Physical Properties

| | C-1 | C-2 | C-3 | C-4 | C-5 |
|--------------------------------|-------|-------|-------|-------|-------|
| Hardness, Shore A | 70 | 74 | 71 | 73 | 74 |
| 100% Modulus, MPa | 4.11 | 5.85 | 4.03 | 5.08 | 5.54 |
| 300% Modulus, MPa | 8.86 | 12.53 | 8.52 | 9.56 | 10.95 |
| 500% Modulus, MPa | 15.99 | 22.12 | 14.75 | 16.64 | 17.73 |
| Tensile strength, MPa | 56.22 | 47.84 | 37.30 | 39.13 | 36.26 |
| Elongation at break, % | 810 | 705 | 700 | 750 | 700 |
| Tear strength, kN/m | 44.4 | 64.8 | 49.2 | 60.0 | 55.6 |
| Abrasion loss, mm ³ | 62.8 | 74.0 | 92.0 | 81.8 | 96.1 |

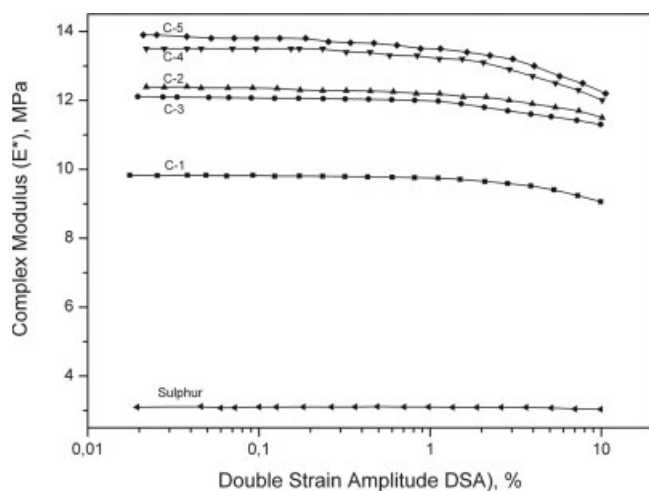


Figure 4 Complex modulus E^* as a function of double strain amplitude (DSA) at 12 Hz of frequency at room temperature.

(ii) the hydrodynamic effect caused by the filler, (iii) the filler–rubber interactions, and (iv) the filler–filler interactions.^{3,22} Filler–filler interactions depend on the deformation degree, whereas the other parameters remain constant, i.e., as the strain applied increases, the modulus drop due to the breakage of the filler network. Therefore, the variation of modulus, ($E_0^* - E_\infty^*$), can be understood as a measure of filler–filler interactions in rubber compounds.^{22,23}

Table V shows the values obtained for the complex modulus at low strain (E_0^*) and at 10% strain (E_∞^*). If the difference between these values is considered as a measure of the interactions between filler particles, the relative drops of the modulus are higher in the compounds filled with silica. This is due to the presence of hydroxyl groups in their surface, which leads to interactions between them, that are broke down when a dynamic deformation is applied.

The compound C-3 shows modulus drops similar to the unfilled compound, due to the low surface activity of the clay employed as filler.

When a temperature sweep is done, at constant strain and frequency, the compounds of XNBR cross-linked with MgO, show two different transitions: the glass transition of the polymer, T_g , at low temperatures, and the ionic transition, T_i , at high tempera-

TABLE V
Values of Complex Modulus, E^*

| | C-1 | C-2 | C-3 | C-4 | C-5 |
|----------------------------|-------|--------|--------|--------|--------|
| E_0^* , kPa | 9,830 | 12,392 | 12,108 | 13,525 | 13,905 |
| E_∞^* , kPa | 9,065 | 11,505 | 11,319 | 12,017 | 12,229 |
| $E_0^* - E_\infty^*$, kPa | 765 | 887 | 789 | 1,508 | 1,676 |
| ΔE^* relative, % | – | 16 | 3 | 97 | 119 |

TABLE VI
Values of the Transitions Temperatures of the Compounds Obtained from the Maximum in the Viscous Modulus

| Frequency (Hz) | C-1 | C-2 | C-3 | C-4 | C-5 |
|---------------------------------------|-------|-------|-------|-------|-------|
| Glass transition temperature, °C | | | | | |
| 5 | –8.9 | –9.2 | –8.6 | –13.8 | –11.7 |
| 10 | –8.3 | –8.0 | –7.6 | –12.2 | –10.1 |
| 15 | –7.7 | –7.4 | –6.9 | –11.3 | –9.4 |
| 20 | –6.8 | –6.8 | –6.4 | –10.5 | –8.8 |
| E_a , kJ/mol | 375.8 | 354.7 | 372.1 | 238.9 | 276.5 |
| Ionic transition temperature, °C | | | | | |
| 5 | 75.7 | 80.4 | 73.8 | 66.6 | 69.0 |
| 10 | 79.9 | 88.0 | 78.7 | 71.3 | 73.9 |
| 15 | 82.7 | 90.9 | 82.2 | 72.0 | 76.7 |
| 20 | 84.0 | 93.2 | 84.2 | 74.4 | 78.8 |
| E_a , kJ/mol | 169.1 | 115.1 | 135.8 | 146.9 | 141.5 |
| ΔT between peaks, °C, at 5 Hz | 84.6 | 89.6 | 82.4 | 80.4 | 80.7 |

tures, due to the presence of the ionic microphases independent from the polymer matrix.²⁴

Table VI shows the temperatures for both transitions at different vibration frequencies, obtained by means of mechano-dynamical analysis (DMA). These values were obtained from the maximum in E'' curves. For all the analyzed frequencies, the values for the glass transition temperature decrease in the presence of fillers. This effect is more noticeable in the compounds filled with silica and it is explained due to the dilution effect previously explained. This effect generates higher mobility in the chains as the ionic microphases are diluted. The ionic transition, T_i , is also affected by the presence of the filler. This temperature decreases when fillers are added, however, the value for the compound filled with carbon black (C-2) is higher than the one obtained for the unfilled compound.

Applying the Arrhenius equation, apparent activation energies were calculated for both transitions. The obtained values are shown in Table VI. As it can be observed, the presence of the fillers decreases the activation energy of both transitions.

In short, it can be concluded that the presence of inorganic fillers in XNBR compounds vulcanized with MgO leads to a compatibilization between the polymer and the ionic phase. This hypothesis can be deduced from the lower difference found between both transitions temperatures.

SEM analysis

Figure 5 shows the SEM micrographs of the fracture morphology for the filled vulcanized compounds.

All filled compounds show a homogeneous distribution of the filler. The microphotography of the

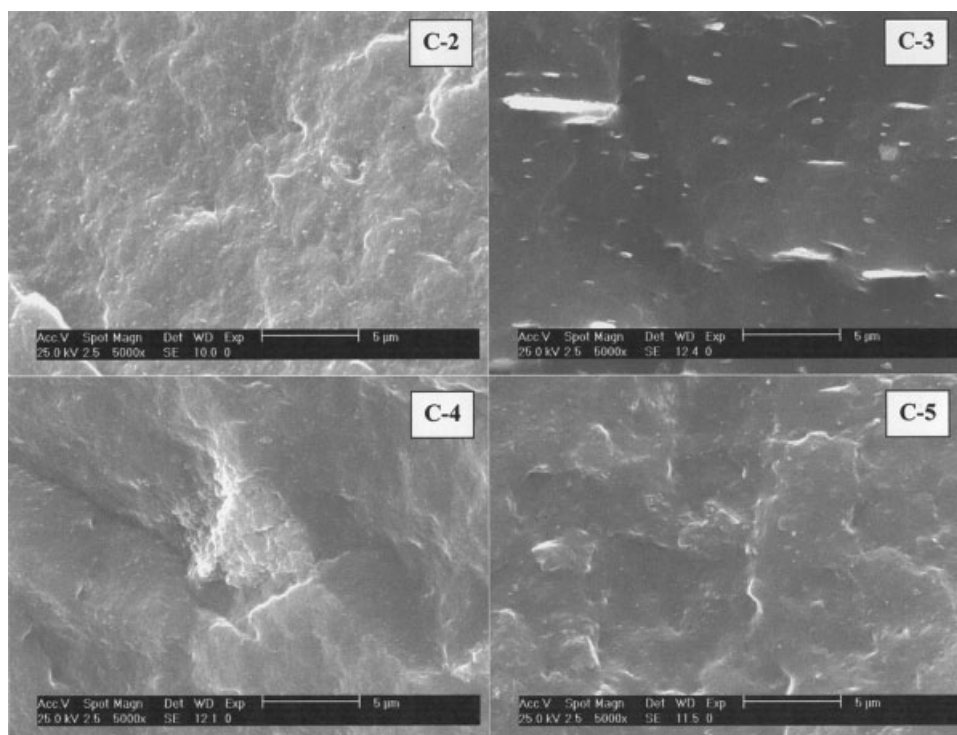


Figure 5 SEM micrograph of the crosslinking compounds.

sample filled with the clay, C-3, shows particles sizes bigger than a micron. However, for the compounds C-2, C-4, and C-5, a homogeneous distribution of nanoparticles is obtained. In the microphotography of the sample C-5, silsep particles with their fibrillar morphology are observed. They present high length-diameter ratio. This morphology improves the processing by extrusion, at high shear rates.

CONCLUSIONS

The influence of low-load reinforcing fillers in XNBR rubbers crosslinked with magnesium oxide has been studied. According with the data obtained, it can be conclude that:

The filler addition affects the vulcanization kinetics. When the fillers added have acid character, the vulcanization reaction is accelerated. However, when the fillers have a basic nature, the opposite behavior is observed. The values for the maximum and minimum torques are also affected by the presence of the fillers. The minimum torque, which can be related to the viscosity of the sample, keeps constant, except for the compound filled with the silica obtained from acid lixiviation of sepiolite (silsep). In this case, an increase in viscosity is observed.

When the rheology of the sample is analyzed, by means of rotational and capillary flows, the

optimum behavior is observed for the sample filled with silsep. The flow index of this sample is similar to the unfilled compound. The excellent processing properties of this sample can be explained by the fibrillar morphology of the sample. These fibres are oriented during the extrusion process, improving the material processing.

From the mechanical analysis, it can be concluded that a small amount of filler improves some properties, e.g., stress at constant strains, while others (tear or abrasion properties) got worse. This fact is due to the dilution effect of the fillers over the ionic microphases, formed during the vulcanization reaction.

To summarize, ionic elastomers (XNBR) show significant variations in properties when small amounts of fillers are added. This behavior does not happen in rubbers crosslinked by conventional vulcanization systems. In this case, higher filler proportions are required in order to achieve an optimum reinforcing level. From all the fillers analyzed, the silica obtained by acid lixiviation of sepiolite, silsep, shows a better response concerning to the rheological behavior and processing properties of the material.

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