

Influence of Some Additives on Rheological Properties, Viscosity, and Dynamic Mechanical Properties in NR Compounds

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ABSTRACT: The effect of some processing aids, Redex-2, Redex-SP-2, and Redex-A, on rheological properties, tensile properties, and dynamic mechanical properties of natural rubber compounds has been studied. The optimum level of these additives was determined and the changes caused by the use of these additives were evaluated by comparison with an additive-free control compound mixed under similar conditions. Redex-2 and Redex-SP-2 improve the processability of the natural rubber compounds, giving a reduction of both Mooney viscosity and shear viscosity, but the concentration of the processing aid necessary to this improvement is different; the optimum concentration is 1.75

phr on Redex-2 compounds, and the optimum concentration is 3 phr on Redex-SP-2 compounds. However, Redex-A behaves as a plasticizer, reducing the Mooney viscosity, the shear viscosity, and the T_g , improving behavior at low temperatures. The optimum proportion is 3 phr. The behavior of these processing aids is related to the temperature, too.
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Key words: processing aids; natural rubber; rheological properties; tensile properties; dynamic mechanical properties

INTRODUCTION

It is known that in rubber compounding some additives are used, in addition to the conventional fillers and curatives, to improve processability.^{1–17} Ideally, this improvement of the processability is achieved without adversely affecting the physical properties of the component produced.

These products cover a wide range of chemicals, from organic chemical peptizers through resinous homogenizers and tackifiers to metal soaps of fatty acids that improve filler dispersion and reduce compound viscosity by lubrication at the molecular level.³

In recent years, a variety of different processing aids have been studied such as rubber seed oil,² Struktol WB 16 and Struktol NS 60,¹ and cashew nut shell liquid and cashew nut shell liquid-formaldehyde resin.⁴

For effective selection and use of a processing aid it is necessary to understand how it functions and to be able to determine the appropriate amount to use.^{5,9–13} The influence of three new process additives, Redex-2, Redex-SP-2, and Redex A, on rheological, dynamical,

and mechanical properties of a carbon black filled natural rubber compound are studied in some detail.

In this article, the optimum level of these additives will be determined and the changes caused by the use of these additives evaluated by comparison with an additive-free control compound mixed under similar conditions.

EXPERIMENTAL

Materials

Table I shows the properties of the three experimental processing aids supplied by Repsol-YPF (Madrid, Spain) used in the present study, and Table II shows the compound formulation.

Compound preparation and vulcanization

Natural rubber gum mix was prepared on a laboratory two-roll mill, with a friction ratio of 1 : 1.20, heated at 80°C.

The compounds were vulcanized in a hydraulic press heated by termofluid to the vulcanization temperature for a specified vulcanization time.

The vulcanization curves were obtained by using a rotorless Monsanto Rheometer model MDR 2000 E at a temperature of 150°C with an oscillation arc of 0.5°.

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TABLE I
Characteristics of the Processing Aids

Trade name	REDEX-2	REDEX-SP-2	REDEX-A
Melt point, °C			63
Ashes, %	2	2	0.01
Refraction index			1.444
Total acidity	42	35	
Band of infrared transmittance, cm ⁻¹	2915	2916	2918
	2848	2848	2848
	1708	1708	
	1537	1537	
	1462	1462	

TABLE II
Formulation of the Compounds in pphr

Mix name	C	A	B
Natural rubber SMR 60	100	100	100
Zinc oxide	4	4	4
Stearic acid	2	2	2
ISAF-N	48	48	48
Aromatic oil Repex E-6	6	6	6
Processing aid	—	1.75	3
Santoflex 13	1.5	1.5	1.5
Flectol H	1	1	1
Paraffin wax ^a	2	2	2
Sulphur	0.5	0.5	0.5
MBS ^b	1.5	1.5	1.5

^a Paraffinic wax Redezone 500
^b 2-(4-morfolinotio) benzotiazol

Viscosity measurement

A Monsanto viscosimeter model MV 2000 E (Alpha technologies Swindon, UK) was used to measure the Mooney viscosity, which is shown as ML (1 + 4) at a temperature of 100°C.

Determination of rheological properties

Rheological properties were measured using a high-pressure capillary rheometer model Göttfert Rheo-

graph 2003. Tests were carried out with a capillary die of diameter 2 mm and L/D ratio of 15. After a warming-up period of three minutes, the melt was extruded through the capillary at the temperatures 80°, 100°, and 120°C. The shear rates were from 0.1 s⁻¹ to 900 s⁻¹.

The barrel pressure and ram rate were converted into the apparent shear stress (τ_a) and shear rate (γ_a), respectively, using the following equations:

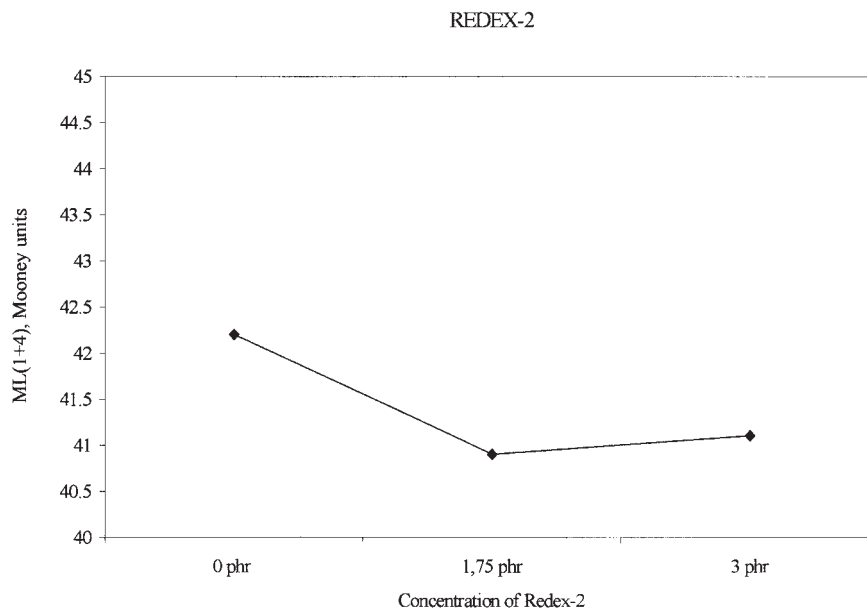


Figure 1 Mooney viscosity ML(1 + 4) at 100°C of Redex-2 compounds.

TABLE III
Flow Behavior Index (n') of Redex-2 at 80, 100, and 120°C for the Weissenberg-Rabinowitsch Correction

Proportion, phr	80°C			100°C			120°C		
	n'	$3n' + 1/4n'$	R	n'	$3n' + 1/4n'$	R	n'	$3n' + 1/4n'$	R
0	0.1871	2.0862	0.9980	0.1266	2.7247	0.9961	0.2633	1.6995	0.9985
1.75	0.2233	1.8696	0.9975	0.1266	2.7247	0.9986	0.4874	1.2629	0.9967
3	0.2283	1.8451	0.9970	0.1267	2.7232	0.9957	0.3489	1.4665	0.9946

$$\tau_a = \frac{B_p}{4 \cdot L/D} \quad (1)$$

where B_p is the barrel pressure and L/D is the capillary length to diameter ratio.

$$\gamma_a = \frac{32Q}{\pi D^3} \quad (2)$$

where Q is the volumetric flow rate and D is the capillary diameter.

The true shear rate was calculated from the apparent shear rate by using the Weissenberg–Rabinowitsch correction:^{18–21}

$$\gamma = \frac{3n' + 1}{4n'} \gamma_a \quad (3)$$

The term n' is the flow behavior index, which is given by:

$$n' = \frac{d \log(\tau_a)}{d \log(\gamma_a)} \quad (4)$$

The flow behavior index was determined by the regression analysis of the values of τ_a and γ_a obtained from the experimental data. The shear viscosity (η) was calculated from the ratio between apparent shear stress and the true shear rate obtained by the Weissenberg–Rabinowitsch correction:^{19–21}

$$\eta = \frac{\tau_a}{\gamma} \quad (5)$$

To understand the influence of temperature on viscosity, Arrhenius^{21,22} plots at a constant shear rate were drawn. In the Arrhenius plots, $\log \eta$ is plotted versus $1/T$ by the following equation:

$$\eta = A \cdot e^{-(E/RT)} \quad (6)$$

where A is a constant characteristic of the polymer, E is the activation energy, and R is the universal gas constant.

Measurement of dynamic mechanical properties

The dynamic properties were determined in a Metra-vib Viscoanalyser RAC 815, based on a principle of nonresonant forced vibrations. Tests were performed over a wide frequency range (5, 15, 50, 100 Hz) and the temperature programs were run from -80 to 130°C under a controlled sinusoidal strain, under a flow of liquid nitrogen. An oscillating dynamic strain of 0.012% was used. The viscoelastic properties such as the storage modulus (E'), the loss modulus (E''), and

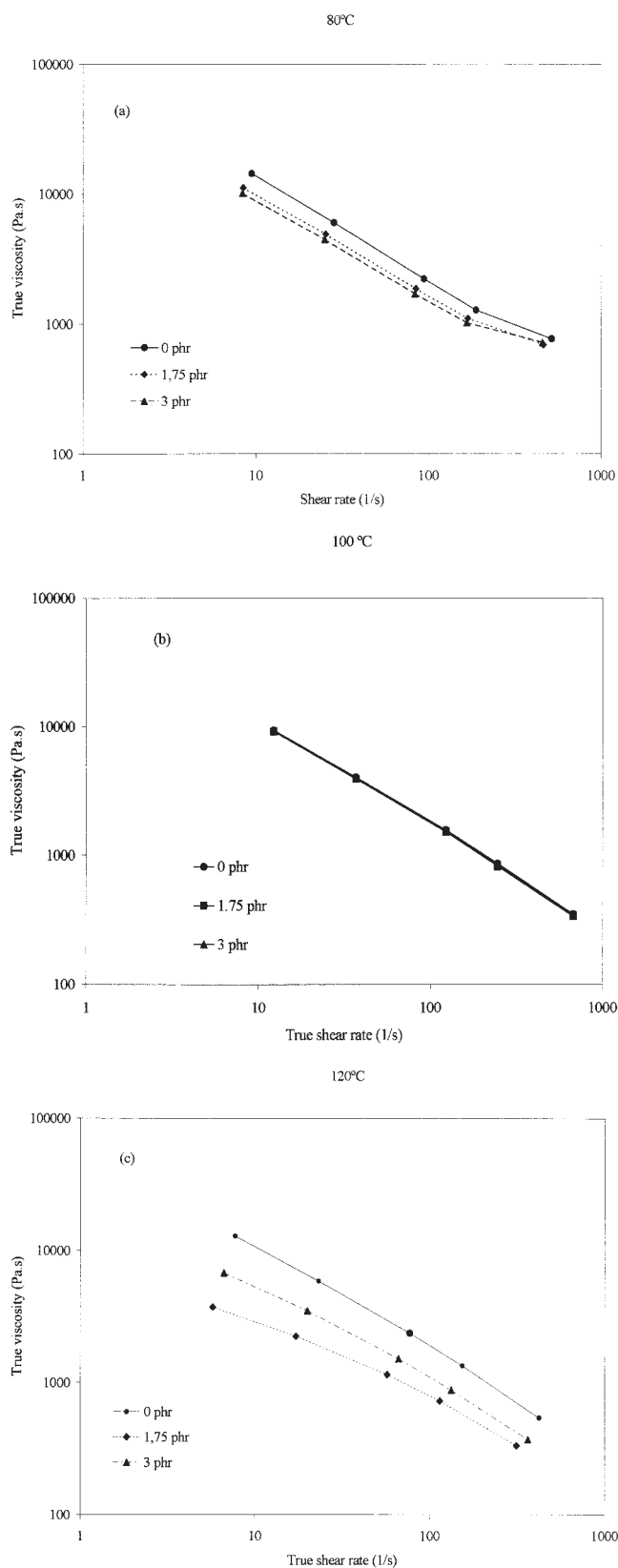


Figure 2 Log-log true viscosity versus shear rate plots of Redex-2 compounds, $L/D = 15$ die: (a) 80°C, (b) 100°C, and (c) 120°C.

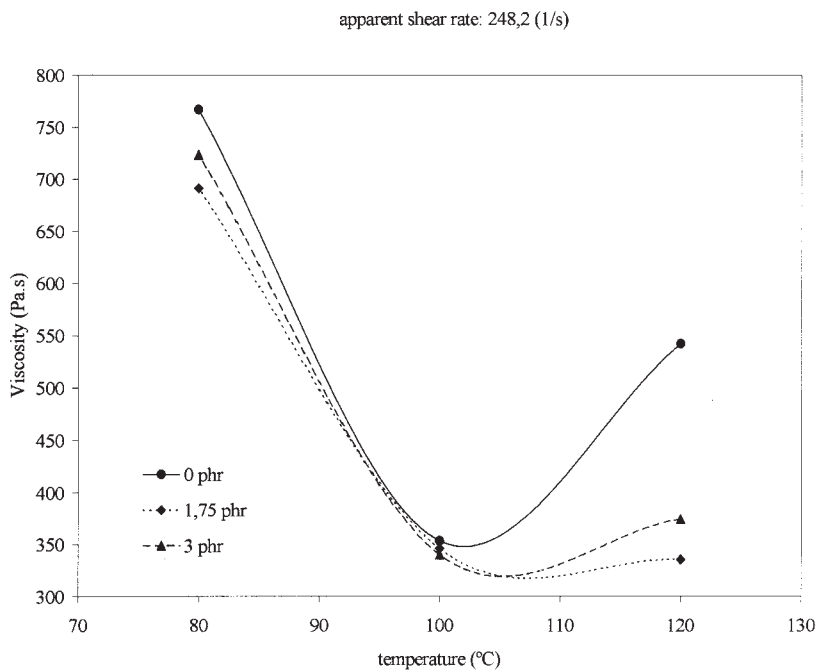


Figure 3 Effect of temperature on shear viscosity of different Redex-2 compounds at a constant shear rate (248,2 s⁻¹).

the mechanical loss factor ($\tan \delta = E''/E'$) were recorded as a function of temperature and frequency.

Measurement of mechanical properties

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 Delft-type samples according to UNE 53516 part 2 at the same deformation rate used for the tensile test. Five samples were tested in each use.

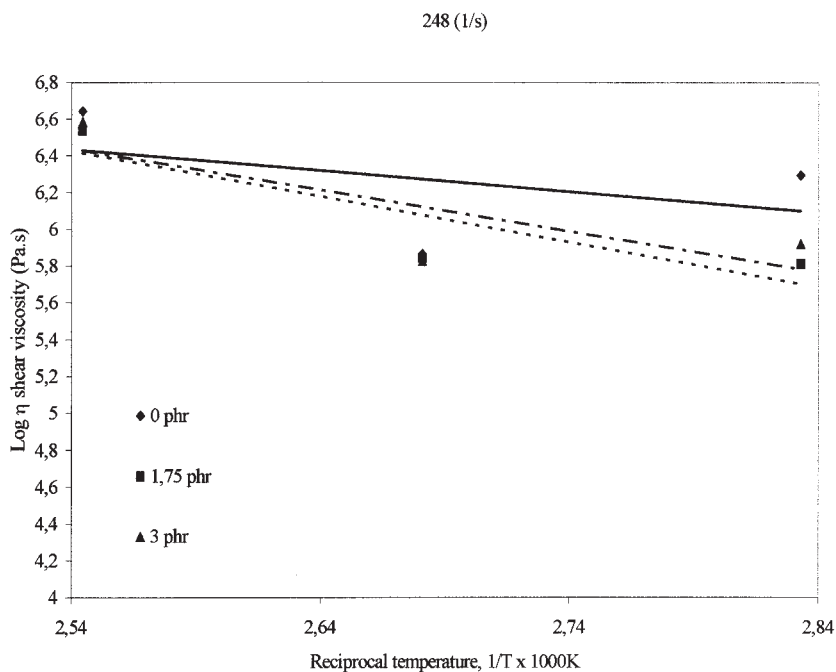


Figure 4 Arrhenius plots for different Redex-2 compounds.

TABLE IV
Activation Energies Calculated from Arrhenius Equation

Concentration of Redex-2, phr	ΔE , cal/mol
0	2537.7
1.75	4902.4
3	4445.4

TABLE V
Glass Transition Temperatures of Redex-2 by Using $\tan \delta$ and Loss Modules $\max(E'')$

Concentration of REDEX-2, phr	$T_g(\tan \delta)$, °C	$T_g(E'')$, °C
0	-51.9	-57.9
1.75	-53.6	-59.8
3	-52.1	-58.0

RESULTS AND DISCUSSION

Influence of Redex-2

Mooney viscosity

It can be seen from Figure 1 that Mooney viscosity decreases when the processing aid is added. Redex-2 acts as a viscosity modifier and the optimum concentration of this processing aid seems to be 1.75 phr.⁵ The proportion of the processing additives is critical; each compound needs an optimum proportion. The typical proportions are from 0.5 to 3 phr, and in most compounds the optimum concentration is 2 phr.^{5,6} Properties of Redex-2 are showed in Table I. The main bands of IR transmittances are 2915, 2848, 1708, 1537, and 1462 cm^{-1} . The bands at 2915 and 2848 cm^{-1} are very strong, which indicates that Redex-2 has a long hydrocarbon chain. The band at 1708 cm^{-1} indicates that Redex-2 has a carboxylic acid group. The bands at 1537 and 1462 cm^{-1} show that Redex-2 has a salt fatty acid group in its composition. The presence of the carboxylic group is confirmed by the total acidity value of this

chemical product. The presence of the carboxylic acid group causes a physical wetting between the hydrophilic molecules of rubber and hydrophilic molecules of the filler.⁶ These structures form micelles³ in either a polar or a nonpolar medium. These micelles are formed in mineral oil with soap fatty acids like Redex-2. Although there is no evidence that these micelles are formed in polymer systems, it seems reasonable to argue that rubber, as a high molecular weight hydrocarbon, is not significantly different from mineral oils and these micelles would be formed in rubber compounds. The viscosity reduction would result from effective interchain lubrication between the polymer molecules and between the polymer molecules and filler.³ Above 2 phr of processing aid, such as 3 phr, the increase in viscosity could be attributed to a weak interface between adjacent layers of Redex-2 when the concentration of the processing aid is higher than that necessary to form only a monolayer of micelles around the filler and the polymer.⁵

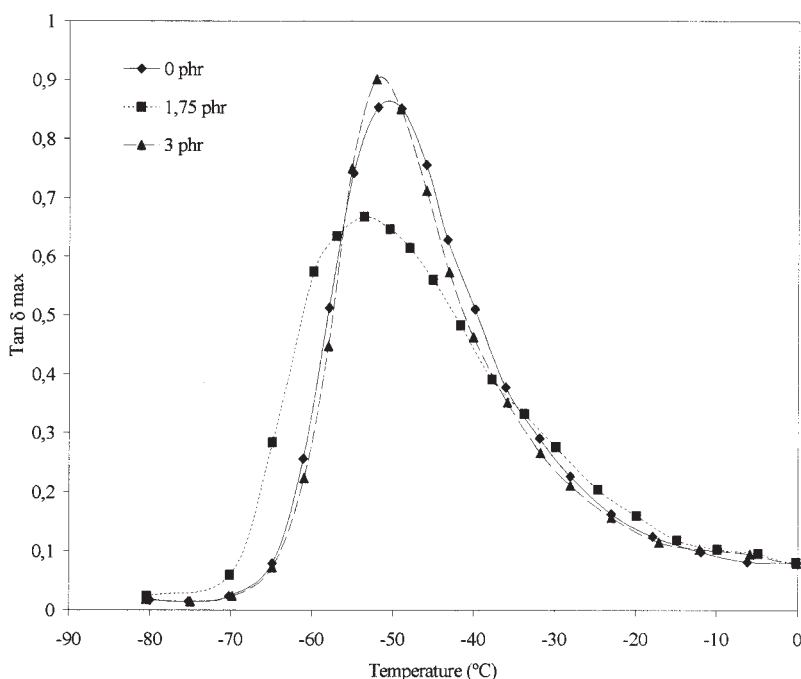


Figure 5 Loss tangent ($\tan \delta$) as a function of temperature at 5 Hz for Redex-2 compounds.

TABLE VI
Mechanical Properties of Redex-2

Mix	Stress at 100% elongation, MPa	Stress at 300% elongation, MPa	Elongation at break, %	Tensile strength, MPa	Tear strength, N
C	1.2	4.2	706	29.1	14.1
A	1.4	6.4	700	26.5	12.8
B	1.5	6.3	706	27.2	12.4

Rheological properties

The extent of pseudoplasticity of the materials can be understood from n' values. Pseudoplastic materials are characterized by n' below 1.^{19,21-23} Flow behavior index values of natural rubber compounds at three temperatures are given in Table III. All the mixes are non-Newtonian pseudoplastic fluids characterized by n' below 1.

At 80°C as the concentration of Redex-2 increases the n' values increase. This suggests that the system becomes less pseudoplastic as processing aid concentration increases, improving the processability at this temperature. The flow behavior indexes are very sim-

ilar at 100°C, although the effect of the processing aid seems less pronounced than at 80°C. When the concentration of Redex-2 is 1.75 phr, there is a maximum at 120°C, and this mix shows the lowest pseudoplastic behavior.

Figures 2(a), 2(b), and 2(c) show the log-log plots of viscosity versus true shear rate at the three temperatures: 80, 100, and 120°C, calculated applying the correction of Weissenberg-Rabinowitsch for the three compounds with the L/D = 15 die.

The viscosity of the two compounds with the processing aid is lower than the viscosity of the control compound, indicating that this processing aid acts to

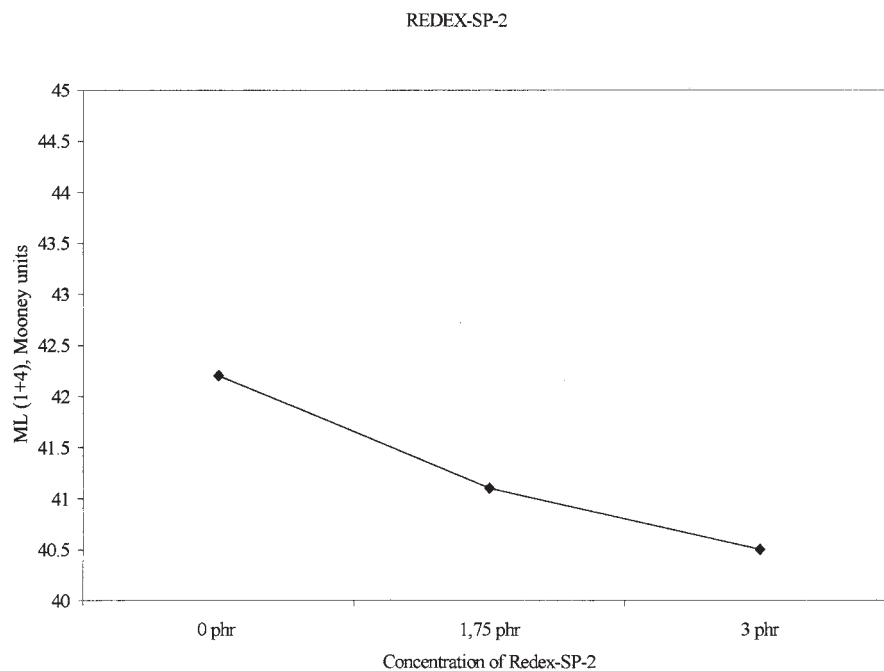


Figure 6 Mooney viscosity ML(1 + 4) at 100°C of Redex-SP-2 compounds.

TABLE VII
Flow Behavior Index (n') of Redex-SP-2 at 80, 100, and 120°C for the Weissenberg-Rabinowitsch Correction

Proportion, phr	80°C			100°C			120°C		
	n'	$3n' + 1/4n'$	R	n'	$3n' + 1/4n'$	R	n'	$3n' + 1/4n'$	R
0	0.1871	2.0862	0.9980	0.1266	2.7247	0.9961	0.2633	1.6995	0.9985
1.75	0.2979	1.5892	0.9916	0.1311	2.6569	0.9998	0.3316	1.5039	0.9987
3	0.3246	1.5202	0.9987	0.1304	2.6672	0.9985	0.3387	1.4881	0.9930

improve the processability of the mix, reducing the viscosity at all shear rates and at the three temperatures. As the concentration of the processing aid increases the viscosity decreases at 80°C, while the viscosity increases when increasing the concentration of the processing aid at 120°C. These results confirm the Mooney viscosity results. The highest reduction in both Mooney viscosity and shear viscosity is obtained in mix A at 120°C.

The variation of the shear viscosity with the temperature of the three mixes at a constant shear rate (248.2 s⁻¹) is given in Figure 3 as an example of the temperature effect on the viscosity of the mixes. Shear viscosity decreases with temperature first, and then increases at 120°C temperature. As the temperature increases the viscosity^{19–21,24} must decrease due to the physical and chemical degradation of the material, but the system begins to undergo crosslinking at 120°C so the viscosity of the three mixes slightly increases at this temperature. Here also, the pseudoplasticity is maintained in all the samples.

The Arrhenius plots of the samples at 248.2 s⁻¹ apparent shear rate are given in Figure 4. The activation energies of the mixes calculated from the slopes of this plot are given in Table IV. Among some other characteristics that are not referred to in this article, the activation energy of a material provides valuable information on the sensitivity of the material towards the change in temperature. The higher the activation energy, the more sensitive the material will be.¹⁹ The compound with 1.75 phr concentration of processing aid shows the highest activation energy. This compound is the most sensitive towards the change in temperature. Such information is highly useful in selecting the temperature for processing during manufacture¹⁹ of the product.

Dynamic mechanical properties

The loss factor-temperature curves have a maximum that coincides with the glass transition temperature T_g of the compounds. Figure 5 shows the peaks of the $\tan \delta$ curves of the compounds as a function of temperature. Table V shows the T_g temperatures of all the compounds determined from $\tan \delta$ and E'' curves, respectively. When the concentration of Redex-2 is 1.75 phr, the maximum loss factor peak shifts to low temperature and the peak decreases and expands. This is because Redex-2 acts as internal lubrication, improving the mobility of the polymer chains and improving the filler dispersion.

Mechanical properties

Results on the mechanical properties of the vulcanizates are given in Table VI. With addition of Redex-2, tensile strength and tear strength are slightly smaller

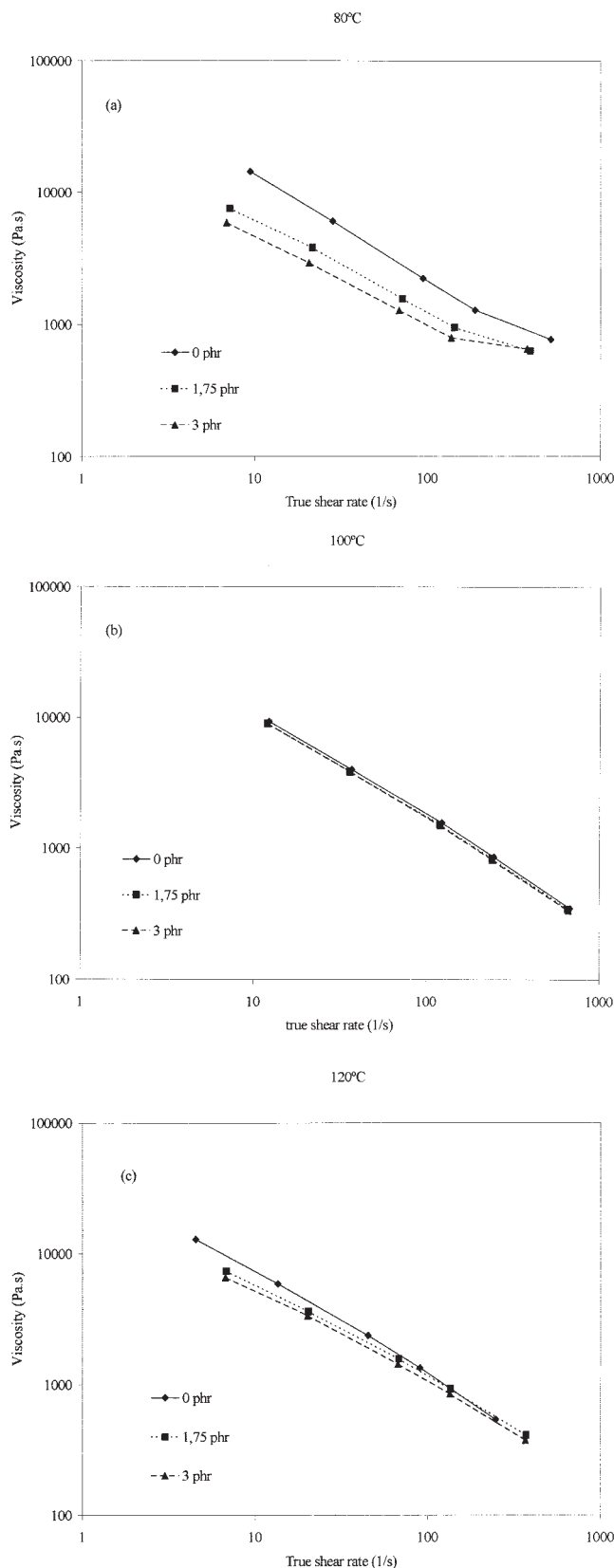


Figure 7 Log-log true viscosity versus shear rate plots of Redex-SP-2 compounds, L/D = 15 die: (a) 80°C, (b) 100°C, and (c) 120°C.

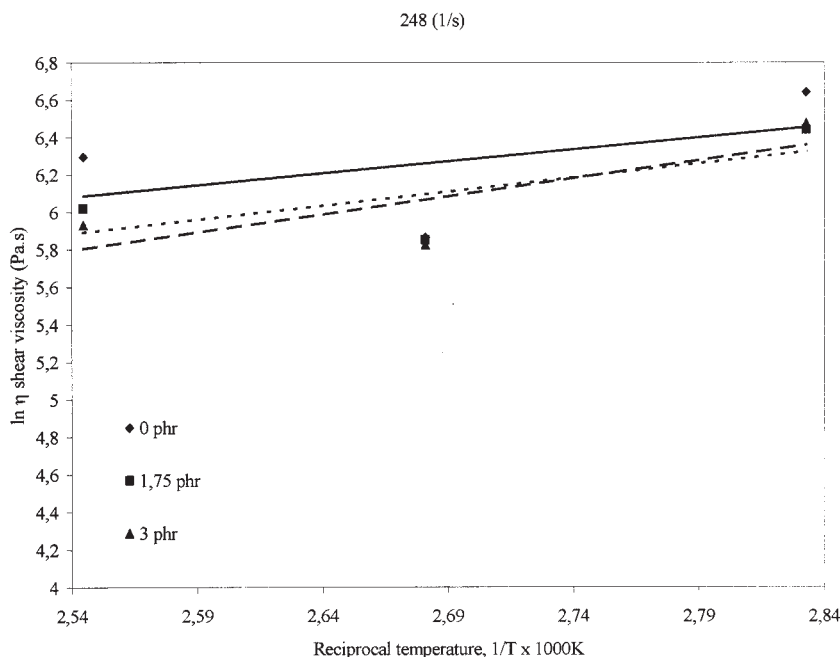


Figure 8 Arrhenius plots for different Redex-SP-2 compounds.

TABLE VIII
Activation Energies Calculated from Arrhenius Equation

Concentration of Redex-SP-2, phr	ΔE , cal/mol
0	2537.7
1.75	2994.1
3	3841.8

than the control. Modulus at 100 and at 300 strains of mix A is slightly higher than mix C. Elongation at break appears to decrease slightly with the incorporation of the additive. Tensile modulus, tensile strength, and elongation at break show constant values when the concentration of Redex-2 increases until 3 phr. The incorporation of the processing aid worsens slightly the mechanical properties of the vulcanizates. Al-

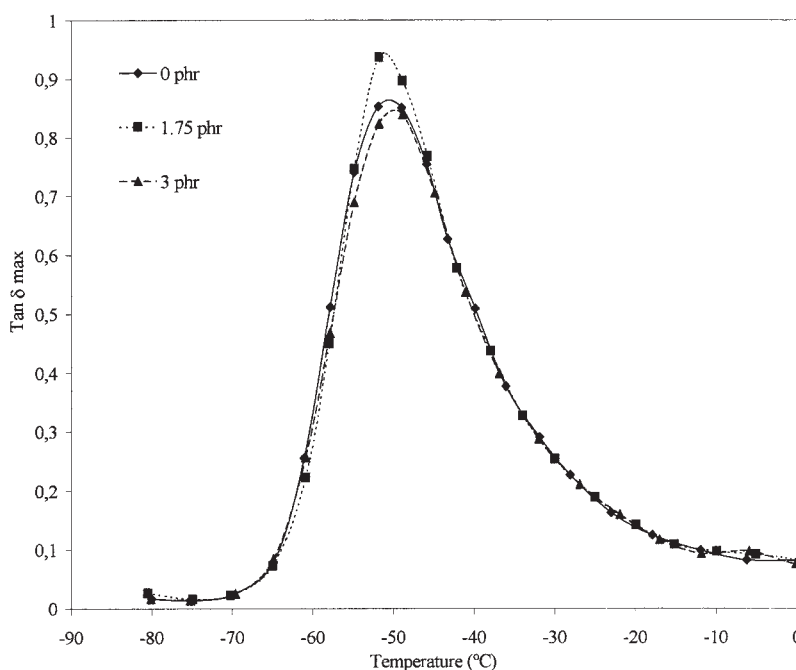


Figure 9 Loss tangent ($\tan \delta$) as a function of temperature at 5 Hz for Redex-SP-2 compounds.

TABLE IX
Glass Transition Temperatures of Redex-SP-2 by Using $\tan \delta$ and Loss Modulus $\max(E'')$

Concentration of REDEX-SP-2, phr	$T_g(\tan \delta)$, °C	$T_g(E'')$, °C
0	-51.9	-57.9
1.75	-51.8	-58.0
3	-51.8	-57.9

though the processing aid improves the dispersion of the filler, promoting the improvement of the mechanical properties, there is an adverse effect of dilution that counteracts this. There is a balance between the

improvement of the processability achieved and the physical properties of the component produced. Redex-2 promotes an improvement of the processability of the compound, but the physical properties of the compounds are a little lower than the physical properties of the control-compound.

Influence of Redex-SP-2

Mooney viscosity

Figure 6 shows the Mooney viscosity of the three compounds. As the concentration of Redex-SP-2 increases, the Mooney viscosity decreases. Redex-SP-2 would act as an internal lubricant between the poly-

TABLE X
Mechanical Properties of Different Concentration of Redex-SP-2

Mix	Stress at 100% elongation, MPa	Stress at 300% elongation, MPa	Elongation at break, %	Tensile strength, MPa	Tear strength, N
C	1.2	4.2	706	29.1	14.1
A	1.5	6.4	668	26.8	12.3
B	1.5	6.3	668	27.2	14.7

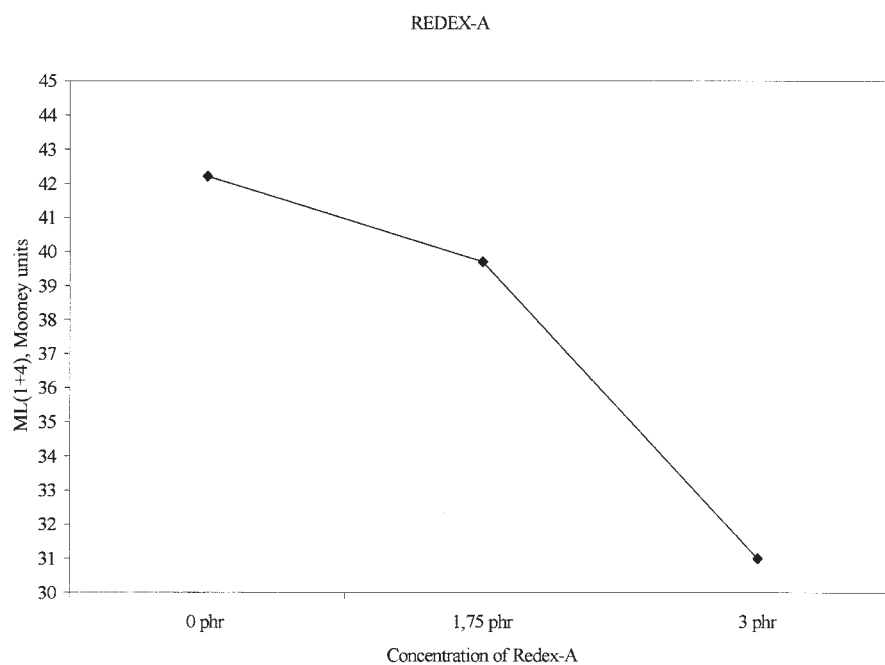


Figure 10 Mooney viscosity ML (1 + 4) at 100°C of Redex-A compounds.

TABLE XI
Flow Behavior Index (n') of Redex-A at 80, 100, and 120°C for the Weissenberg-Rabinowitsch Correction

Proportion, phr	80°C			100°C			120°C		
	n'	$3n' + 1/4n'$	R	n'	$3n' + 1/4n'$	R	n'	$3n' + 1/4n'$	R
0	0.1871	2.0862	0.9980	0.1266	2.7247	0.9961	0.2633	1.6995	0.9985
1.75	0.1921	2.0514	0.9887	0.1405	2.5294	0.9936	0.3209	1.5291	0.9934
3	0.1947	2.0340	0.9902	0.1443	2.4825	0.9902	0.3675	1.4303	0.9974

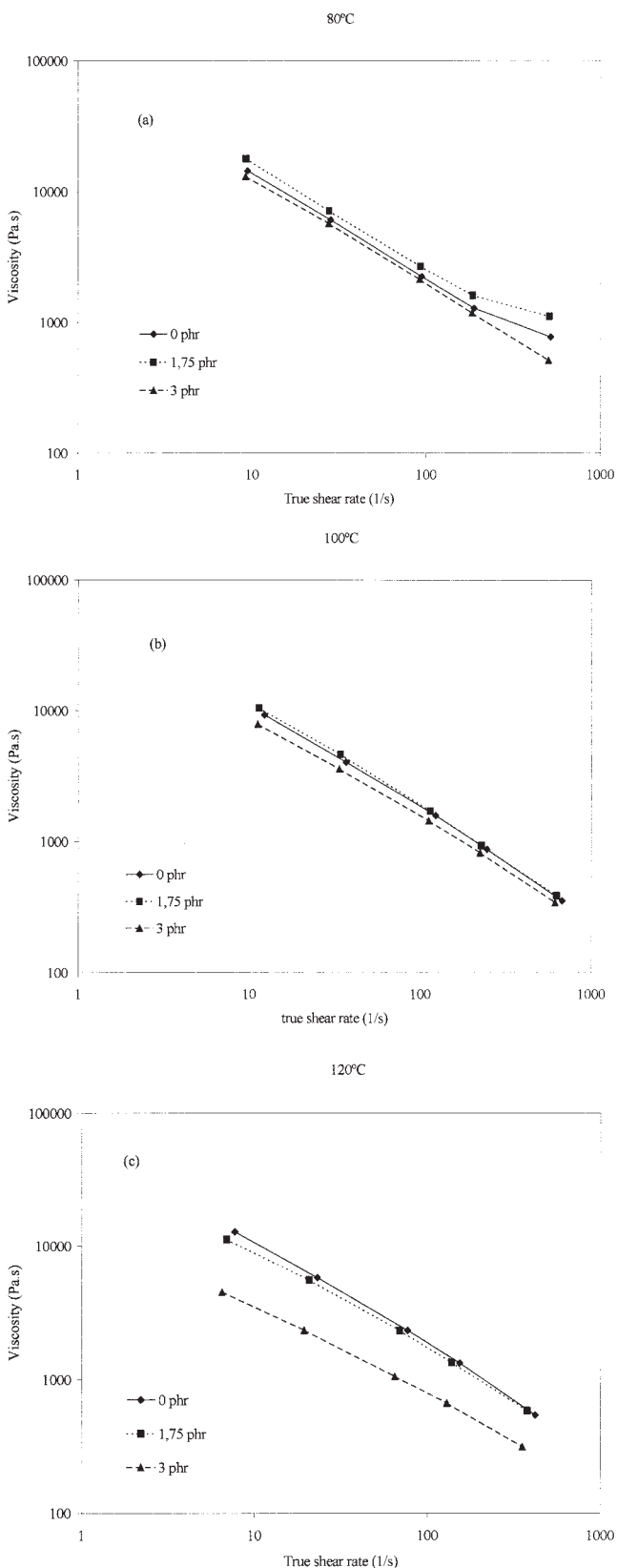


Figure 11 Log-log true viscosity versus shear rate plots of Redex-A compounds, L/D = 15 die: (a) 80°C, (b) 100°C, and (c) 120°C.

mer chains, promoting the viscosity reduction. As noted above, the proportion of the processing aids is critical. The typical proportion is from 0.5 to 3 phr. For this additive, the optimum proportion seems to be 3 phr. Properties of Redex-SP-2 are shown in Table I. The main IR transmittances are the same bands that described Redex-2. Redex-SP-2 has a similar structure to Redex-2. However, the total acidity value of this additive is lower than that of Redex-2, because the proportion of the carboxylic acid group is lower than Redex-2; therefore, it would be necessary to use more proportion of this additive than Redex-2, for wetting the hydrophilic molecules of rubber and the filler. Mix B has the optimum proportion of this additive. In Redex-SP-2 only a monolayer of micelles would be formed around the filler and the polymer when the concentration of Redex-SP-2 is 3 phr.

Rheological properties

Table VII shows the behavior index values (n') of the three compounds. All the mixes are non-Newtonian pseudoplastic fluids characterized by n' below 1. As the concentration of Redex-SP-2 increases, the n' values increase at the three temperatures. This suggests that the system becomes less pseudoplastic as processing aid concentration increases, improving the processability.

Figures 7(a), 7(b), and 7(c) show the log-log plots of viscosity versus true shear rate, calculated applying the correction of Weissenberg–Rabinowitsch for the three compounds, with the L/D = 15 die at the three temperatures 80, 100, and 120°C. The viscosity of the two compounds with Redex-SP-2 are lower than the viscosity of the control compound, which suggests that this processing aid acts to improve the processability of the mix, reducing the viscosity at all shear rates and at the three temperatures. As the concentration of the processing aid increases, the viscosity decreases. These changes are more pronounced at 80°C and at 120°C, while the viscosity at 100°C of the three mixes are more similar. These results confirm the Mooney viscosity results, with the greatest reduction in either Mooney viscosity and shear viscosity obtained in mix B.

The variation of shear viscosity with the temperature of the three mixes at a constant shear rate is similar to that of Redex-2. The viscosity at 120°C increases because the system begins to undergo crosslinking at 120°C.

The Arrhenius plots of the samples at 248.2 s⁻¹ apparent shear rates are given in Figure 8. The activation energies of the mixes calculated from the slopes of these plots are given in Table VIII. As the concentration of the processing aid increases, the values of the activation energies of the mixes increase. Therefore, the compound with 3 phr concentration of Redex-SP-2

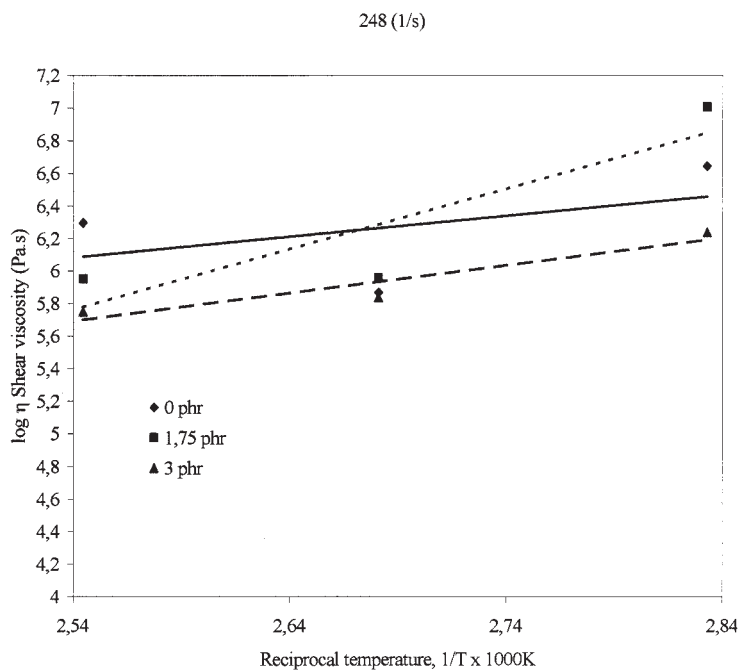


Figure 12 Arrhenius plots for different Redex-A compounds.

TABLE XII
Activation Energies Calculated from Arrhenius Equation

Concentration of Redex-A, phr	ΔE , cal/mol
0	2537.7
1.75	7382.3
3	3388.9

is the most sensitive material towards the change in temperature.

Dynamic mechanical properties

Figure 9 shows the peaks of the $\tan \delta$ curves of the compounds as a function of the temperature, and

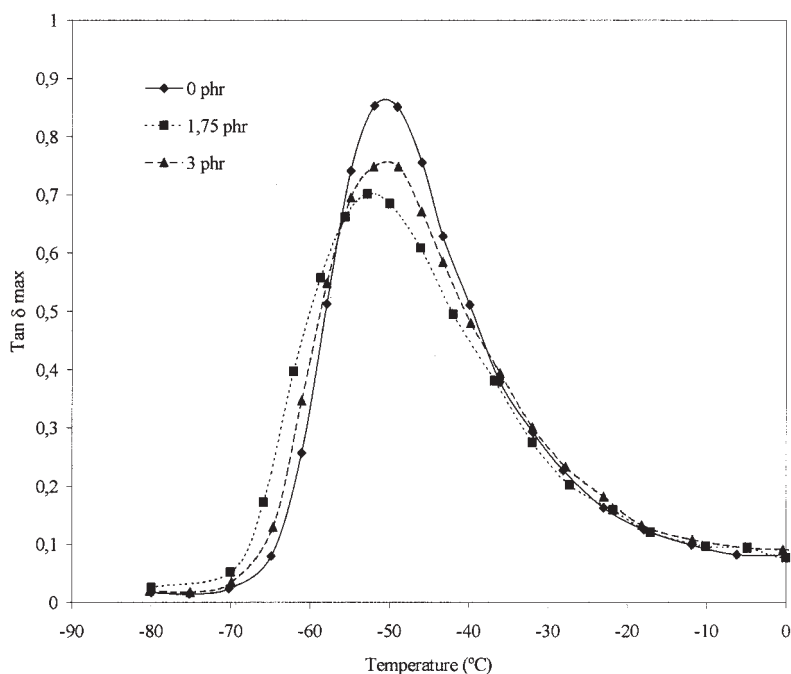


Figure 13 Loss tangent ($\tan \delta$) as a function of temperature at 5 Hz for Redex-A compounds.

TABLE XIII
Glass Transition Temperatures of Redex-A by Using tan δ and Loss Modules max(E'')

Concentration of REDEX-A, phr	T _g (tan δ), °C	T _g (E''), °C
0	-51.9	-57.9
1.75	-52.8	-62.1
3	-50.4	-61.1

Table IX shows the T_g temperatures of all the compounds determined from tan δ and E'' curves, respectively. With this processing aid, large differences in the values of tan δ are not observed. The maximum of the three curves is achieved at the same temperature.

Mechanical properties

Results on the mechanical properties of the vulcanizates are given in Table X. Modulus at 100 and at 300 strains of mix A are slightly higher than for mix C. Elongation at break appears to decrease slightly with the incorporation of the additive. Tensile strength of mix A is slightly lower than for mix C. As described for Redex-2, the mechanical properties of mix A and mix B are slightly lower than in the control compound because of the effect of the dilution of Redex-SP-2.

Influence of Redex-A

Mooney viscosity

Figure 10 shows the Mooney viscosity of the three compounds. As the concentration of Redex-A increases, the Mooney viscosity decreases. Properties of Redex-A are shown in Table I. The main IR transmittance bands are 2915 and 2848 cm⁻¹, which indicates that Redex-A has only a long hydrocarbon chain. The viscosity reduction is caused by the plasticizing effect of Redex-A.¹ Redex-A cannot form micelles because it does not have a polar functional group in its composition.

Rheological properties

Table XI shows the behavior index values (n') of the three compounds. All the mixes are non-Newtonian

pseudoplastic fluids characterized by n' below 1. As the concentration of Redex-A increases, the n' values increase at all three temperatures. This suggests that the system becomes less pseudoplastic as processing aid concentration increases, improving the processability.

Figures 11(a), 11(b), and 11(c) show the log-log plots of viscosity versus true shear rate, calculated applying the correction of Weissenberg–Rabinowitsch for the three compounds, with the L/D = 15 die at the three temperatures 80, 100, and 120°C. The viscosity of mix B is lower than the viscosity of the control compound, which seems to indicate that the optimum concentration of this processing aid is 3 phr, reducing the viscosity at all shear rates and at the three temperatures. However, the viscosity of mix A is lower than mix C only at 120°C.

The variation of shear viscosity with temperature of the three mixes at a constant shear rate is similar to Redex-2 and Redex-SP-2. The viscosity at 120°C increases because the system begins to undergo crosslinking at 120°C.

The Arrhenius plots of the samples at 248.2 s⁻¹ apparent shear rates are given in Figure 12. The activation energies of the mixes calculated from the slopes of these plots are given in Table XII. Mix A shows the highest activation energy. This compound is the most sensitive towards the change in temperature.

Dynamic mechanical properties

Figure 13 shows the peaks of the tan δ curves of the compounds as a function of the temperature. Table XIII shows the T_g temperatures of all the compounds determined from tan δ and E'' curves, respectively. When the concentration of Redex A is 1.75 phr, the maximum loss factor peak shifts to low temperature and the peak decreases and expands, indicating that Redex A has a small plasticizer effect.

Mechanical properties

Results on the mechanical properties of the vulcanizates are given in Table XIV. Modulus at 100 and at 300 strains of mix A and mix B are slightly higher than for mix C. Elongation at break appears to decrease slightly with the incorporation of the additive. Tensile

TABLE XIV
Mechanical Properties of Redex-A

Mix	Stress at 100% elongation, MPa	Stress at 300% elongation, MPa	Elongation at break, %	Tensile strength, MPa	Tear strength, N
C	1.2	4.2	706	29.1	14.1
A	1.4	6.6	656	27.3	13.0
B	1.3	5.8	660	25.0	13.3

strength of mix A and mix B are slightly lower than for mix C. The mechanical properties of mix A and mix B are slightly lower than for the control compound because of the plasticizing effect of Redex-A. When the concentration of Redex-A increases, the mechanical properties decrease slightly.

CONCLUSION

Both Redex-2 and Redex-SP-2 improve the processability of the natural rubber compounds giving a reduction of both Mooney viscosity and shear viscosity, but the concentration of the processing aid necessary to achieve this improvement is different. The optimum concentration is 1.75 phr for Redex-2 compounds, and the optimum concentration is 3 phr for Redex-SP-2 compounds. In both systems, the processing aid would form micelles around the polymer and the filler due to the presence of the carboxylic acid group in their structures. Redex SP-2 has a lower proportion of the acid group than Redex-2 meaning that it is necessary to increase the proportion of this additive to reduce the viscosity. The mechanical properties of these mixes are slightly lower than the control compound due to the effect of the dilution.

The behavior of these processing aids is related to temperature, too. Mix A is the most sensitive towards changes in the temperature in Redex-2 compounds, and mix B is the most sensitive towards changes in the temperature in Redex-SP-2 compounds.

Redex-A behaves as a plasticizer, reducing the Mooney viscosity, the shear viscosity, and the T_g , improving behavior at low temperatures. The optimum proportion is 3 phr, but this mix shows the lowest mechanical properties.

REFERENCES

1. Sirisinha, C.; Sittichokchuchai, W. *J Appl Polym Sci* 2001, 80, 2474.
2. Aigbodion, A. I.; Menon, A. R. R.; Pillai, C. K. S. *J Appl Polym Sci* 2000, 77, 1413.
3. Dick, J. S. *Rubber Technology: Compounding and Testing for Performance*; Hanser Gardner Publications, Inc.: Cincinnati OH, 2001; Chap 14, pp. 363–377.
4. Menon, A. R. R.; Aigbodion, A. I.; Pillai, C. K. S.; Mathew, N. M.; Bhagawan, S. S. *European Polymer Journal* 2002, 38, 163.
5. Abidin, M. Z.; Ismail, H.; Murray, G. A. W.; Freakley, P. K. *Rubber World* 1994, 211, 30.
6. Hepburn, C. *Revista del Caucho* 1999, 484, 24.
7. Lloyd, D. G. *Caoutchoucs et plastiques* 1990, 699, 90.
8. Lau, W.W. Y.; Swee-hin, T.; Suat-Hong, G. *Polymer International* 1991, 24, 95.
9. Ismail, H.; Ishiaku, U. S.; Din, R. H. *Polymer International* 1996, 39, 309.
10. Ismail, H.; Chia, H. H. *Polymer Testing* 1998, 17, 199.
11. Ismail, H.; Freakley, P. K.; Sheng, E. *J Eur Polym* 1995, 31, 1049.
12. Ismail, H.; Freakley, P. K.; Sutherland, I.; Sheng, E. *J Eur Polym* 1995, 31, 1109.
13. Ismail, H.; Freakley, P. K. *J Eur Polym* 1996, 32, 411.
14. Ismail, H.; Ishiaku, U. S.; Ishak, Z. A. M.; Freakley, P. K. *J Eur Polym* 1997, 33, 1.
15. Chattaraj, P. P.; Mukhopadhyay, R. *Rubber Chem Technol* 1996, 70, 90.
16. Okieimen, F. E.; Akinlabi, A. K. *J Appl Polym Sci* 2002, 85, 1070.
17. Nandan, V.; Joseph, R.; George, K. E. *J Appl Polym Sci* 1999, 72, 487.
18. Sirisinha, C.; Sittichokchuchai, W. *J Appl Polym Sci* 2000, 76, 1542.
19. Asaletha, R.; Groeninckx, G.; Kumaran, M. G.; Thomas, S. *J Appl Polym Sci* 1998, 69, 2673.
20. Liang, J.-Z. *J Appl Polym Sci* 2002, 85, 606.
21. George, J.; Ramamurthy, K.; Varughese, K. T.; Thomas, S. *J Polym Sci: Part B: Polym Phys* 2000, 38, 1104.
22. Antony, P.; Bhattacharya, A. K.; De, S. K. *De J Appl Polym Sci* 1999, 71, 1257.
23. Jansen, P.; Soares, B. G. *J Appl Polym Sci* 2002, 84, 2335.
24. Fujiyama, M.; Kondou, M.; Ayawa, K.; Inata, H. *J Appl Polym Sci* 2002, 85, 762.