

# Rheological Behavior of Blends of Natural Rubber and Styrene-Butadiene Rubber Latices

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## SYNOPSIS

The rheological behavior of blends of natural rubber (NR) and styrene-butadiene rubber (SBR) latices has been studied with reference to the effects of blend ratio, shear rate, surface-active agents (casein and sodium carboxymethyl cellulose), and temperature. When the SBR content was less than 50%, the viscosities of the blends appeared to be a nonadditive function of the viscosities of the constituent homopolymers; i.e., a positive deviation was observed. This was due to the structural buildup of the SBR domains. The SBR domains underwent agglomeration and consequently so-called microfloculation took place. The viscosities of all the blends were found to decrease with increase of temperature and shear rate. The increase in temperature and shear rate marginally weakened the structural buildup as evidenced by the lowering of viscosity. As the SBR content in the system increased, the pseudoplasticity of the blend increased. Even in the presence of surface-active agents the blends showed composition-dependent positive deviation. However, surface-active agents marginally reduced the extent of structural buildup by reducing the microfloculation behavior of SBR domains. © 1995 John Wiley & Sons, Inc.

## INTRODUCTION

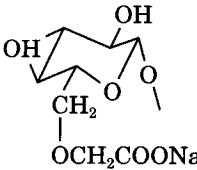
The technique of blending of two or more polymers to produce end products having a combination of properties has gained significant importance recently. Many studies have been reported on blending of rubbery polymers.<sup>1-7</sup> The blending of polymer latices is more easier compared to dry rubber blending. Latex stage blending<sup>8,9</sup> offers the possibility of finer scale dispersion of components.<sup>10</sup> Shundo et al.<sup>11</sup> reported the composition-dependent properties of natural rubber and styrene butadiene rubber blends. They compared solution blending, latex blending, roll blending, and banbury mixer blending and found that the properties of natural rubber/styrene-butadiene rubber (NR-SBR) blends showed a direct relation to their blend ratios, regardless of blending methods used. Iino and co-workers<sup>12</sup> also compared the properties of latex and mill-mixed blends of carboxylated SBR and vinyl pyridine SBR.

Latex blending was found to be better for achieving interactions between these acidic and basic rubbers than mill mixing. A deep understanding of the flow behavior of blends is essential for developing suitable end products. Viscosity of the individual component plays a critical role in the flow and forming of compounds. Several investigations have been made to understand the complicated rheological behavior of polymers in view of its relevance to processing.<sup>13-16</sup> Rheology of polymer blends has been extensively investigated by many research groups. The composition dependence of viscosity of heterogeneous blends is extremely complex.<sup>17</sup>

When two polymers in the latex stage are mixed, the viscosity varies appreciably with composition. King<sup>18</sup> has attempted to improve the compatibility of NR-SBR blends by modification of the stabilizing system. Blackley and Charnock<sup>19-21</sup> have reported on the properties of blends of NR and SBR latices. The viscosity of the blend increases with increasing NR latex content. They proposed that the observed effects arise, essentially, from differences in the rate of interchange of adsorbed stabilizers within the system. However, no serious

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**Table I Materials Used**

Material	Source	Chemical Structure	Total Solid Content (%)	Dry Rubber Content (%)	Styrene Content (%)
Natural rubber latex	Padinjarekara Agencies, Kottayam, India	$\begin{array}{c} \text{---CH}_2 \quad \quad \quad \text{CH}_2\text{---} \\ \quad \quad \quad \diagdown \quad \diagup \\ \quad \quad \quad \text{C}=\text{C} \\ \quad \quad \quad \diagup \quad \diagdown \\ \text{H} \quad \quad \quad \text{CH}_3 \end{array}$	61.6	60	—
Styrene-butadiene rubber latex	Synthetics and Chemicals, Madras, India	$\begin{array}{c} \text{H} \\   \\ \text{---CH}_2\text{---C---CH}_2\text{---CH=CH---CH}_2\text{---} \\   \\ \text{C}_6\text{H}_5 \end{array}$	42.6	38	45.2
Casein	Rubber Board, Kottayam, India	—CO—NH— (Phosphoprotein)	—	—	—
Sodium carboxy methyl cellulose	Rubber Board, Kottayam, India		—	—	—

analysis has been done on the effect of shear rate, temperature, and role of surface-active agents on the flow behavior.

Viscosity is one of the important factors in the manufacture of many latex goods.<sup>22</sup> The NR-SBR latex blends are used in very thick gauge products such as mattresses, where extra strength and elongation are required to facilitate stripping the vulcanized foam rubber from molds containing large lugs.<sup>23</sup>

The flow properties of latices are strongly influenced by shear rate, particle size, presence of electrolytes,<sup>24</sup> temperature, and the incorporation of surface-active agents. Surface-active agents are the substances which can bring about marked modifications in the surface properties of aqueous media even though they are present in small amounts.<sup>25</sup> The principal effect of surface-active agents is lowering of surface and interfacial free energies. The surface-active agents are hydrocolloids soluble in water giving viscous solutions which display the properties of a typical colloid system.<sup>26</sup> The effects of various surface-active agents on the rheological properties of centrifuged and creamed latex compounds have been reported.<sup>27</sup>

The purpose of the present study is to investigate the effect of blend ratio, temperature, shear rate,

and surface-active agents on the flow behavior of NR and SBR latex blends.

## EXPERIMENTAL

Details of the materials used in this study are shown in Table I.

### Blend Preparation

Latices of NR and SBR were blended using a magnetic stirrer. Blends containing 0, 30, 50, 60, 70, 90, and 100% of NR are designated as  $N_0$ ,  $N_{30}$ ,  $N_{50}$ ,  $N_{60}$ ,  $N_{70}$ ,  $N_{90}$ , and  $N_{100}$ , respectively, where the subscripts indicate the weight percent of NR in the blend. The weights were calculated (according to dry rubber content) in such a way that the dry rubber content of each component will be approximately equal. A 5% aqueous solution of each of the surface-active agents (casein and sodium carboxymethyl cellulose) was prepared. The dosage of each surface-active agent was fixed as 0.5 phr (parts per hundred of rubber latex). The surface-active agents were mixed with the polymer latices using the magnetic stirrer. Blends containing casein are designated as  $N_0C$ ,  $N_{30}C$ ,  $N_{50}C$ ,  $N_{70}C$ , and  $N_{100}C$  and those containing

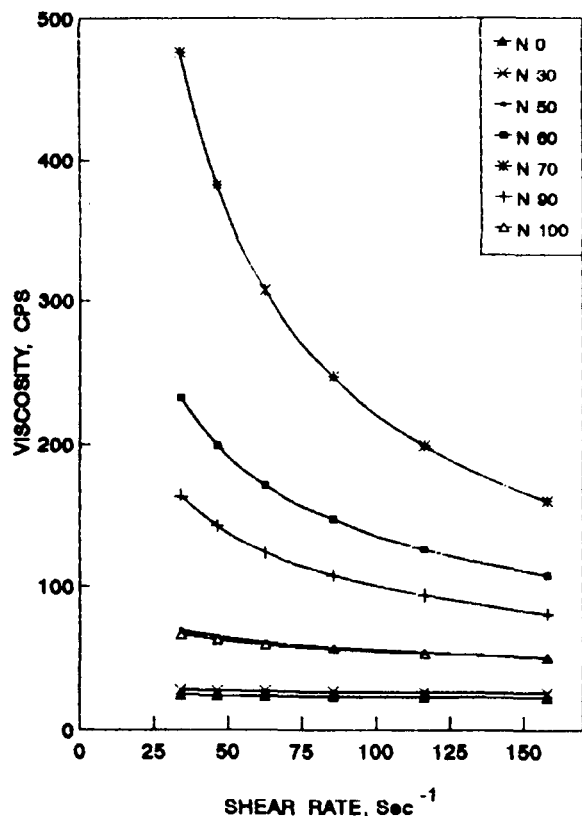


Figure 1 Viscosity vs. shear rate of NR-SBR blends at 30°C.

sodium carboxymethyl cellulose (NaCMC) are designated as  $N_0N$ ,  $N_{30}N$ ,  $N_{50}N$ ,  $N_{70}N$ , and  $N_{100}N$ .

### Rheological Measurements

The rheological measurements of the blends with and without surface-active agents were made using a Contravers viscometer, Rheomat-30. A concentric cylinder with "B" cup was used for making viscosity measurements. The experiments were carried out at three different temperatures, i.e., 30, 40, and 50°C.

## RESULTS AND DISCUSSION

The rheological behavior of the blends has been analyzed using the Power law equation:<sup>28</sup>

$$\tau = K\dot{\gamma}^n \quad (1)$$

where  $\tau$  is shear stress (Pa),  $K$  is the viscosity index,  $\dot{\gamma}$  is the shear rate ( $s^{-1}$ ), and  $n$  is the pseudoplasticity index. The  $n$  and  $K$  values were obtained by regression analysis of the values of  $\tau$  and  $\dot{\gamma}$  obtained from

experimental data. The apparent viscosity ( $\eta$ ) was calculated using the equation

$$\eta = K(\dot{\gamma})^{n-1} \quad (2)$$

Each rheogram was analyzed for yield stress ( $\tau_0$ ), viscosity index ( $K$ ), and pseudoplasticity index ( $n$ ).

### Effect of Blend Ratio and Shear Rate on Viscosity

The effect of blend ratio and shear rate on the viscosity of NR-SBR latex blends at 30°C is shown in Figure 1. NR latex shows higher viscosity than SBR because of its high solid content. The NR and SBR latices and blends containing lower proportion of NR ( $\leq 50\%$ ) show almost Newtonian behavior. As the NR content increases ( $> 50\%$ ), the blends show non-Newtonian behavior. The viscosity decreases with increasing shear rate, indicating pseudoplastic behavior. The extent of decrease in viscosity with shear rate (pseudoplasticity) increases as NR content increases. It is interesting to note that, over the entire range of shear region, when the SBR content is less than or equal to 50%, the viscosity of the blends is a nonadditive function of the viscosities of

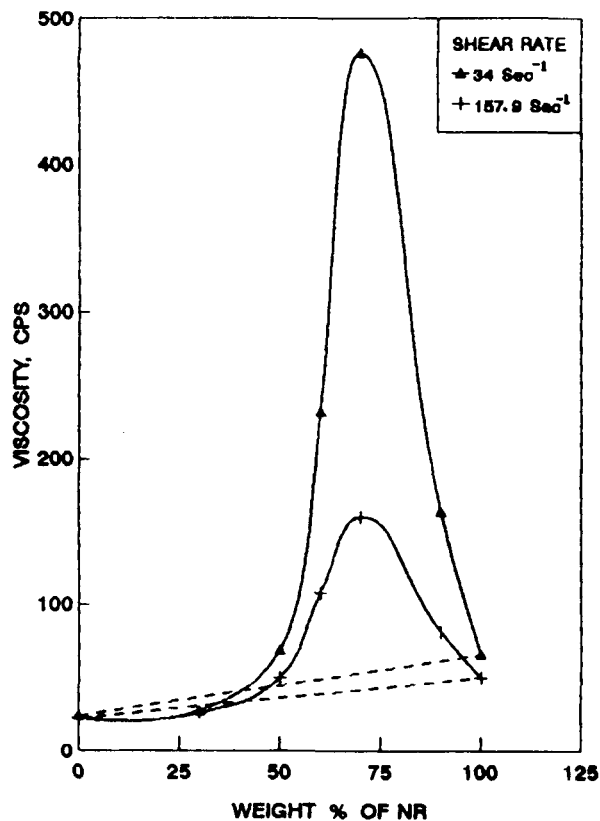
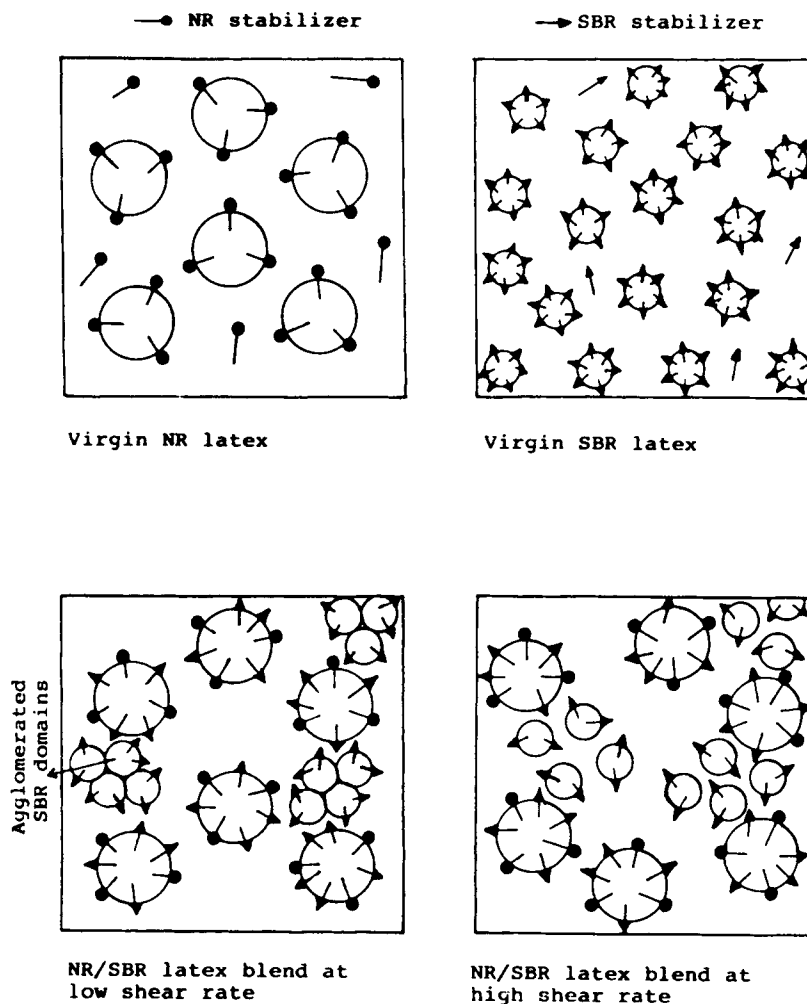


Figure 2 Viscosity vs. weight percent of NR of NR-SBR blends at 30°C.



**Figure 3** Speculative model for structural buildup in NR-SBR latex blends.

the component polymers; i.e., a positive deviation is observed. This behavior can be well understood from Figure 2, where the variation of viscosity with blend ratio is presented at two different shear rates. It can be seen that this behavior is observed only at lower proportions of SBR ( $\leq 50\%$ ). At high SBR contents ( $> 50\%$ ) the observed viscosities do not differ from the additivity rule, i.e., those expected on the basis of a more viscous liquid (NR latex) being diluted with a less viscous liquid (SBR latex) with which it is miscible. It is important to note the shear rate dependence of the positive deviation; i.e., the extent of positive deviation is reduced at high shear rates (Fig. 2). The nonadditive nature of the viscosity of NR-SBR blends at low and high shear rate can be explained as follows: When the proportion of SBR in a blend is less than or equal to 50%, the SBR domains undergo agglomeration and consequently the so-called microfloculation of SBR domains takes place. This accounts for the large increase in

viscosity from additivity line. Since the shear rates encountered in the viscometer are rather low, there would be little deformation of the agglomerated domains. This generates shear-dependent structural buildup in the NR matrix. As shear rate increases, the domains will be partially separated and the structural buildup is reduced. A speculative model is presented in Figure 3 to illustrate the structural buildup at low and high shear rates.

The mechanism of microfloculation as suggested by Blackley and Charnock<sup>19</sup> is based on the exchange of stabilizers between the synthetic and natural rubber latices. When natural and synthetic latices are mixed, the adsorbed stabilizers exchange with one another, until all the particles of the blend are stabilized. The stabilizers of SBR latex are more rapidly desorbed and reabsorbed on the NR particle. At low NR content there is little increase in viscosity from additivity. This is because the interfacial area of NR present is very small compared with that of

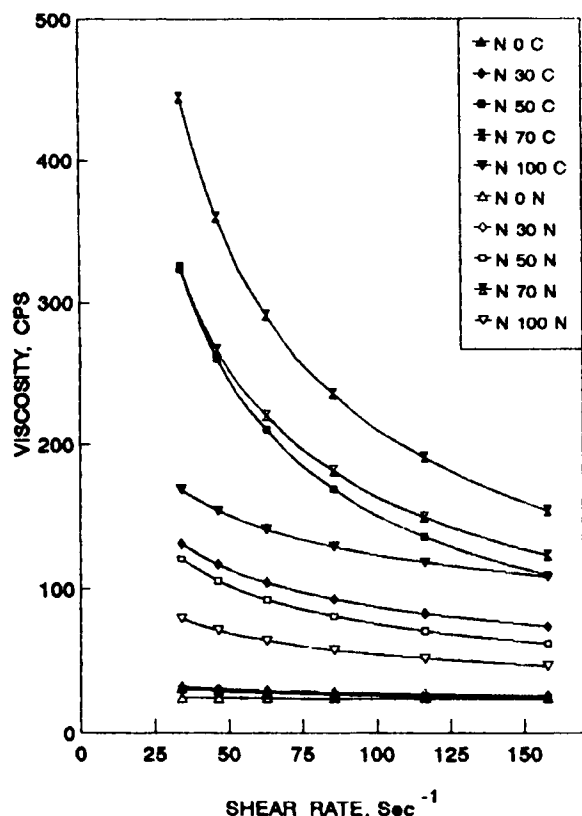


Figure 4 Viscosity vs. shear rate of NR-SBR blends containing surface-active agents at 30°C.

the SBR interfacial area, and therefore small amounts of stabilizer, which is rapidly desorbed from the SBR latex, has very little effect upon the stability of the latex. However, as the NR content increases more and more, stabilizers will be lost from the SBR particles and the same will be adsorbed on the NR domains. This will lead to the destabilization of SBR domains resulting in agglomeration. Thus the large increase in viscosity is due to the agglomeration of SBR domains. The observed fall in viscosity when NR content exceeds a certain level ( $> 80\%$ ) is due to the diminishing concentration of destabilized SBR latex particles. In other words at very low SBR concentrations ( $< 80\%$ ) each SBR particle is increasingly likely to have a stabilized NR particle as its neighbor rather than a destabilized SBR particle. It can be noticed that the structural buildup is maximum at 20 and 30% of SBR. At high shear rates the agglomeration behavior is reduced and therefore the viscosity decreases (Fig. 2). This sort of shear-dependent positive deviation in the viscosities of polymer blends has been reported by many researchers. For example, Zhang et al.<sup>29</sup> studied the agglomeration of polymer latex by another polymer latex. They found that when two lattices are mixed,

the particles will adhere together or adhere between the particles of same latex leading to agglomeration. In some cases the agglomeration behavior is advantageous since it provides a balance of properties which cannot be achieved with a rheologically simple straightforward blend.<sup>30</sup> Lee<sup>31</sup> and Munstedt<sup>32</sup> observed an increase in viscosity in the low-shear-rate region in the case of elastomer-modified thermoplastic. They concluded that agglomeration and network formation by the rubber domains is the cause of the existence of yield stress. Ablazova<sup>33</sup> reported that the viscosity-vs.-composition curve of polyoxymethylene (POM)/copolyamide (CPA) goes through a maximum at low shear stress levels and to a minimum at high shear stress levels. Koshy et al.<sup>34</sup> observed similar positive deviations in viscosity in natural rubber (NR)-ethylene vinyl acetate (EVA) blends. They found that the increase in viscosity is due to the clustering of EVA domains in the continuous NR medium.

#### Effect of Surface-Active Agents on Viscosity

Figure 4 indicates the effect of surface-active agents and shear rate on the viscosity of NR-SBR latex

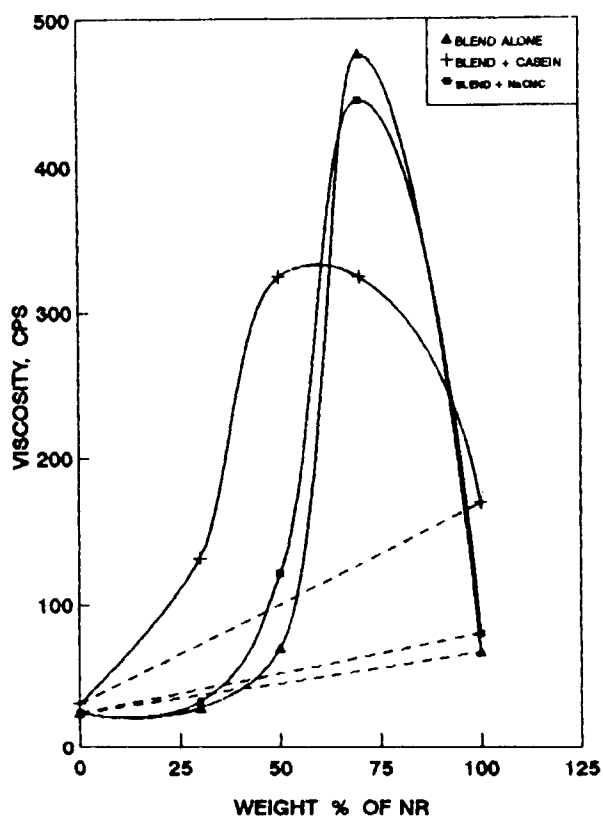


Figure 5 Viscosity vs. weight percent of NR of NR-SBR blends containing surface-active agents.

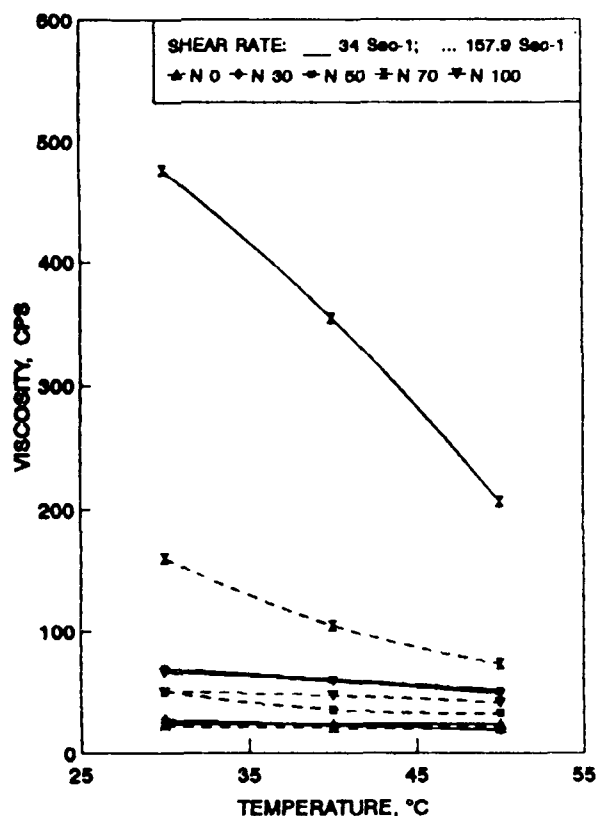


Figure 6 Viscosity vs. temperature of NR-SBR blends.

blend. When surface-active agents are added to NR latex, the viscosity is increased. This is due to the fact that parts of the surface-active agents are adsorbed on the surface of NR particles and the rest goes to the aqueous phase. There will be an interconnection between the surfactants in the rubber phase and aqueous phase in the form of a network in which the rubber particles are occluded.<sup>35</sup> This will lead to an increase in viscosity. As compared to sodium carboxymethyl cellulose (NaCMC), casein is more effective in increasing the viscosity of NR latex. As shear rate increases, the network becomes loose and viscosity will be reduced. The pseudoplasticity of NR is increased by adding surface-active agents. In the case of SBR the surface-active agents do not increase the viscosity much. They show almost Newtonian behavior. NaCMC has little effect on the viscosity of SBR latex since it is immiscible with SBR. Casein increases the viscosity of SBR, but the increase is not very significant as compared to NR.

The behavior of blends in the presence of surface-active agents is similar to that of the virgin blends (without surface-active agents). When SBR content is less than 50%, the blend shows positive deviation; i.e., the viscosities are higher than the homopolymer

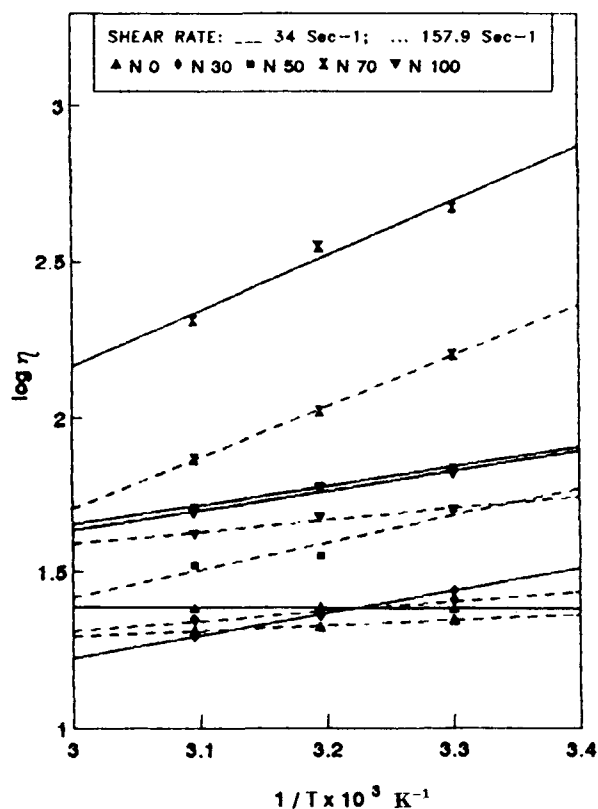


Figure 7  $\log \eta$  vs.  $1/T$  of NR-SBR blends.

viscosities. This is presented in Figure 5. One can note that the addition of surface-active agents marginally reduces the microfloculation behavior. Among the two surface-active agents, casein is found to be more effective in reducing the agglomeration of SBR domains. This is because NaCMC is immiscible with SBR phase.

The destabilization and adsorption of surface-active agents in NR-SBR blend is very complex. As compared to NR particles, the SBR particles are almost completely saturated with surface-active agents. Therefore, the added surface-active agents

Table II Activation Energy for Flow of Blends in Absence of Surface-Active Agents

Shear Rate (s <sup>-1</sup> )	Activation Energy (kJ/mol)				
	N <sub>0</sub>	N <sub>30</sub>	N <sub>50</sub>	N <sub>70</sub>	N <sub>100</sub>
34	0.110	14.022	12.004	33.903	12.289
46.3	0.614	12.440	13.022	33.450	11.337
62.9	1.332	10.899	14.032	33.00	10.393
85.5	2.053	9.292	15.046	33.540	9.449
116.2	2.771	7.717	16.060	32.098	8.504
157.9	3.490	6.144	17.071	31.646	7.559

will preferentially locate at the NR phase and the aqueous phase: only a small portion goes to the SBR phase. Let us now look at the situation at low and high NR content. At low NR content ( $< 50\%$ ) the added surface-active agent is sufficient to saturate the NR phase. As a result there will be no net transfer of stabilizer from the SBR phase to the NR phase. Therefore, no microfloculation of SBR domains takes place. But as NR content increases, the added stabilizer is not sufficient to saturate the NR particles. Therefore, exchange of stabilizer takes place from the SBR phase to the NR phase. This leads to the destabilization of SBR latex resulting in agglomeration. However, as compared to a system containing no surface-active agents, the extent of agglomeration is less in the presence of surface-active agents. The high pseudoplasticity (the sharp decrease in viscosity with shear rate) of the system in the presence of surface-active agent is due to the structural buildup becoming weak at high shear rates.

#### Effect of Temperature on Viscosity

The effects of temperature on the viscosity of the blends at two shear rates are shown in Figure 6. Viscosities of all the blends decrease with increasing temperature. As temperature increases the free volume increases. As a result the flow units become less restricted, more highly energized, and less organized.<sup>36</sup> In the case of  $N_{70}$  blend there is a sharp decrease in viscosity with temperature at both shear rates. This happens because the structural buildup of SBR domains, as reported earlier, becomes weak with the rise of temperature.

To further understand the effect of temperature on viscosity of the blends, Arrhenius plots at two different shear rates were made (Fig. 7). In this figure the logarithm of viscosity is plotted as a function of reciprocal temperature. The activation energies of flow, calculated from the slopes of the lines, are given in Table II. The activation energy of a material provides valuable information on the sensitivity of the material toward the change in temperature. The higher the activation energy, the more temperature sensitive the material will be. In the case of NR and SBR, for  $N_{30}$  and  $N_{50}$  blends the activation energy values are not very high. But the very high activation energy value of the  $N_{70}$  blend indicates the high-temperature sensitivity of the agglomerated SBR domains.

The effects of temperature on the viscosity of NR-SBR latex blends in the presence of surface-active agents at a shear rate of  $157.9 \text{ s}^{-1}$  is shown

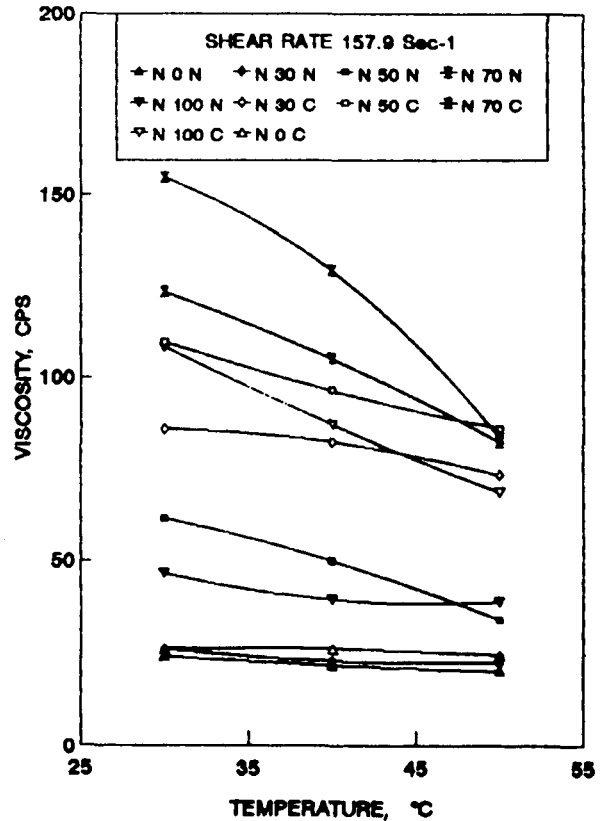


Figure 8 Viscosity vs. temperature of NR-SBR blends containing surface-active agents.

in Figure 8. In all cases the viscosity values decrease with increase in temperature. As shown earlier in Figure 7, in the case of blends without surface-active agents, only  $N_{70}$  shows a sharp decrease in viscosity. But when surface-active agents are added, all the blends show sharp decreases in viscosity with rise of temperature. It is important to note that decrease of viscosity with the increase of temperature is sharper in the cases of  $N_{70}C$ ,  $N_{70}N$ , and  $N_{50}C$ . This is associated with the temperature-induced breakdown of the SBR structural agglomerates.

The Arrhenius plots of the blends containing surface-active agents are shown in Figure 9. The activation energies calculated from the slopes of the plots at a shear rate of  $157.9 \text{ s}^{-1}$  are shown in Table III. By comparing the behavior with the blends without surface-active agents (Table II), it can be noticed that addition of casein decreases the activation energy of all blends, especially that of  $N_{70}$ . The ability of casein to reduce the agglomeration of SBR domains is clear from the lowering of activation energy values. Carboxymethyl cellulose also decreases the activation energy of the  $N_{70}$  blend.

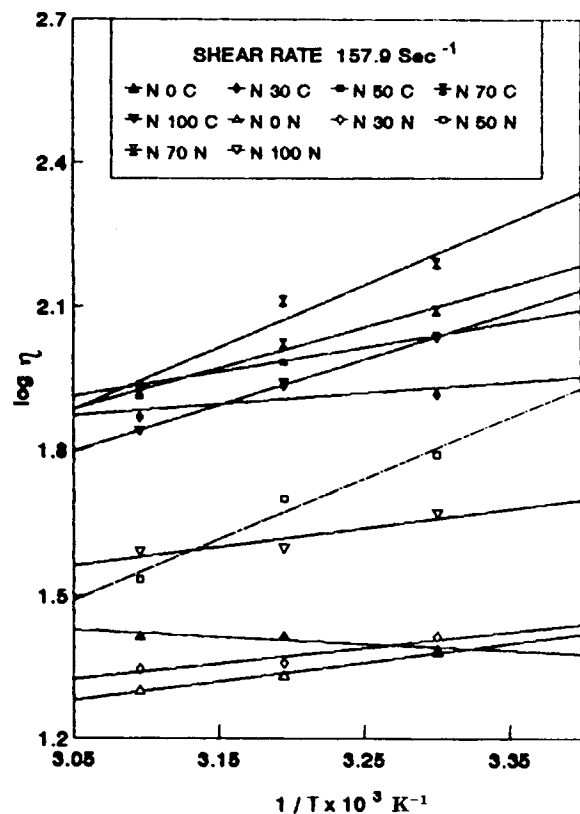


Figure 9  $\log \eta$  vs.  $1/T$  of NR-SBR blends containing surface-active agents.

### Zero Shear Viscosity ( $\eta_0$ )

Figure 10 shows the zero shear viscosity of the blends. NR shows higher zero shear viscosity than SBR. As NR content increases, the  $\eta_0$  also increases and reaches a maximum at  $N_{70}$  and then decreases. The  $\eta_0$  values of the system (in the presence and absence of surface-active agents) show composition-

Table III Activation Energy for Flow of Blends Containing Surface-Active Agents at Shear Rate  $157.9 \text{ s}^{-1}$

Blend	Activation Energy (kJ/mol)
$N_0C$	2.775
$N_{30}C$	6.256
$N_{50}C$	9.850
$N_{70}C$	16.368
$N_{100}C$	18.357
$N_0N$	7.617
$N_{30}N$	6.179
$N_{50}N$	24.077
$N_{70}N$	24.896
$N_{100}N$	7.414

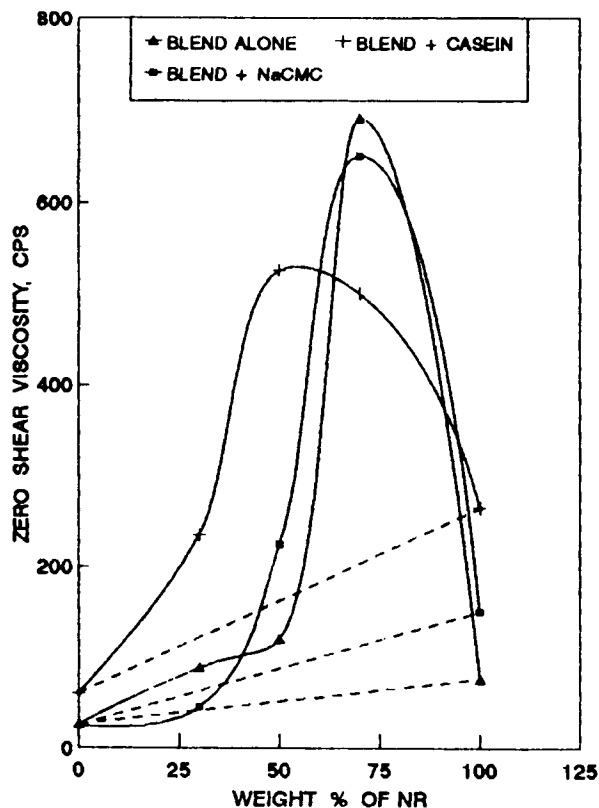


Figure 10 Zero shear viscosity vs. weight percent of NR of NR-SBR blends.

dependent positive deviations due to the agglomeration of the SBR domains. The existence of positive deviation in the zero shear viscosity vs. composition curve of high density polyethylene (HDPE)-ethylene vinyl acetate (EVA) blends has been reported by Fujimura and Iwakura.<sup>37</sup>

### Pseudoplasticity Index ( $n$ )

The effects of temperature and blend ratio on the flow behavior indices of the samples are given in Table IV. The extent of non-Newtonian behavior of

Table IV Pseudoplasticity Index Values in Absence of Surface-Active Agents

Blend	Pseudoplasticity Index, $n$		
	30°C	40°C	50°C
$N_0$	0.949	0.903	0.892
$N_{30}$	0.903	0.977	0.874
$N_{50}$	0.792	0.650	0.713
$N_{70}$	0.290	0.206	0.329
$N_{100}$	0.824	0.855	0.899



the system can be judged from the  $n$  values. Pseudoplastic materials are characterized by  $n < 1$ . Therefore, a high value of  $n$  shows low pseudoplastic nature of the material.

From Table IV it is clear that NR is more pseudoplastic than SBR. The pseudoplasticity of blends is higher than the constituent polymers. The pseudoplasticity increases in the order  $N_{30} < N_{50} < N_{70}$ . The maximum pseudoplasticity of  $N_{70}$  is indicated by the lowest value of  $n$ . This is associated with the shear-dependent structural buildup of the SBR domains. This result suggests that one can easily control the pseudoplasticity of NR-SBR blends by adjusting the blend composition.

Table V shows the pseudoplasticity of NR, SBR, and blends containing surface-active agents at three different temperatures. In most cases, when surface-active agents are added, the pseudoplasticity is increased. However, a few systems show the reverse trend.

### Yield Stress ( $\tau_0$ )

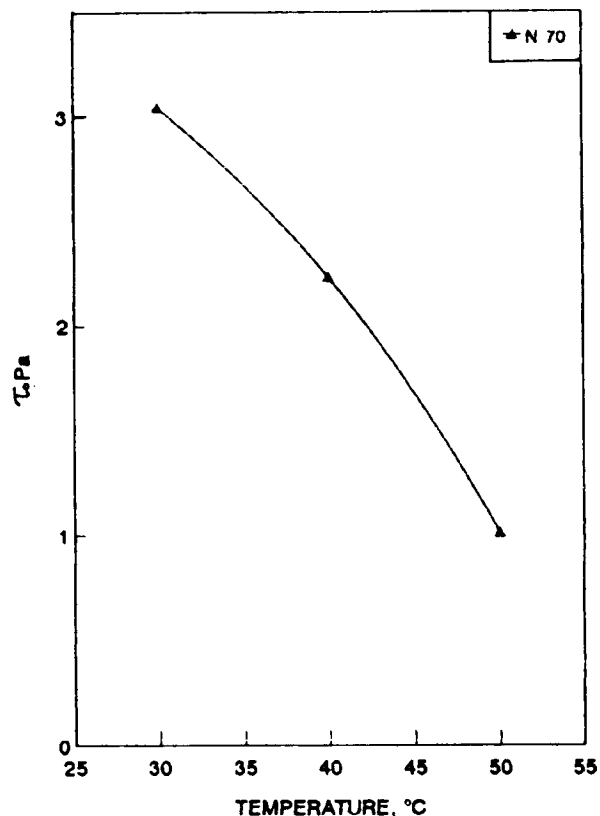
Figure 11 shows the yield stress values of  $N_{70}$  blend as a function of temperature. Yield stress was exhibited by systems containing a higher proportion of NR ( $> 50\%$ ) where the blend undergoes microfloculation. Therefore, a finite positive stress is to be applied to initiate the flow in these systems. It is seen that as temperature increases, the yield stress decreases.

## CONCLUSION

The effects of blend ratio, temperature, shear rate, and surface-active agents on the rheological behavior

**Table V Pseudoplasticity Index Values of Blends Containing Surface-Active Agents**

Blend	Pseudoplasticity Index, $n$		
	30°C	40°C	50°C
$N_0C$	0.846	1.02	1.02
$N_{30}C$	0.622	0.522	0.663
$N_{50}C$	0.295	0.287	0.370
$N_{70}C$	0.372	0.382	0.510
$N_{100}C$	0.712	0.695	0.805
$N_0N$	1	1.04	1.02
$N_{30}N$	0.856	1.09	1.070
$N_{50}N$	0.562	0.600	0.495
$N_{70}N$	0.312	0.311	0.358
$N_{100}N$	0.652	0.663	0.712



**Figure 11** Yield stress vs. temperature of NR-SBR blend.

of NR-SBR latex blends have been studied. The viscosity-composition curves of the low-SBR-content ( $< 50\%$ ) blends (in the presence and absence of surface-active agents) showed composition-dependent positive deviation. The changes in viscosity arise essentially from the differences in the rates of interchange of adsorbed stabilizers within the system. A difference in rates of desorption/adsorption of stabilizers could lead to destabilization of the SBR domains, leading to the development of agglomerated network structures throughout the blend. Presence of this agglomerated structure imparts yield stress and high pseudoplasticity values to the blends. Increase of shear rate and temperature reduces the agglomeration of the SBR domains. The destabilization of SBR domains can be marginally reduced by the addition of surface-active agents. Casein is more effective than NaCMC for this purpose.

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