

COMPARISON OF VISCOELASTIC BEHAVIOUR IN SILICA FILLED CURED AND UNCURED CPE/NR BLENDS WITH VARIOUS MIXING TIME

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ABSTRACT:

Blends of 30-phr silica filled elastomeric chlorinated polyethylene (CPE) and natural rubber (NR) at the blend composition ratio of 80/20 CPE/NR were prepared with various mixing time from 120 to 600 s. Viscoelastic behaviours of cured and uncured blends were determined using two rheometers with different shear modes, i.e., the oscillatory rheometer (Rubber Process Analyser, RPA2000) and the rate-controlled capillary rheometer (Goettfert Rheotester 2000). Results obtained reveal that the viscoelastic behaviour of blends is influenced by the formation of silica transient network, particularly in cured blends. Mixing time affects viscoelastic properties of vulcanised blends to some extent which is due probably to the high extent of thermal degradation, but plays no significant role in viscoelastic properties of unvulcanised blends. The superimposition of oscillatory and steady shear results is possible when the elastic component is eliminated from the results.

ZUSAMMENFASSUNG:

Blends aus mit Silika (30phr) gefülltem, elastomerem und chloriertem Polyethylen und natürlichem Gummi (NR) bei einer Blendzusammensetzung von 80/20 CPE/NR wurden bei verschiedenen Mischzeiten zwischen 120 und 600 s hergestellt. Das viskoelastische Verhalten der unvernetzten und vernetzten Blends wurde mit Hilfe von zwei Rheometern in unterschiedlichen Schermoden bestimmt, d. h. einem Oszillationsrheometer (Rubber Process Analyser RPA 2000) und einem scherraten-kontrollierten Kapillarrheometer (Goettfert Rheotester 2000). Die Resultate verdeutlichen, dass das rheologische Verhalten der Blends durch die Bildung eines transienten Silikanetzwerkes beeinflusst wird, insbesondere in den vernetzten Blends. Die Mischzeit beeinflusst bis zu einem Grad die viskoelastischen Eigenschaften der vulkanisierten Blends, der möglicherweise zu einem hohem Mass aus der thermischen Degradation resultiert. Dagegen spielt die Mischzeit keine signifikante Rolle bei den viskoelastischen Eigenschaften der nichtvulkanisierten Blends. Die Superposition der oszillatorischen und stationären Scherergebnisse ist möglich, wenn der elastische Anteil von den Ergebnissen eliminiert wird.

RÉSUMÉ:

Des mélanges de caoutchouc naturel (NR) avec du polyéthylène chloré (CPE) renforcé par de la silice 30-phr ont été préparés pour un ratio de composition de 80/20 CPE/NR en variant le temps de mélange entre 120 et 600 s. Les comportements viscoélastiques des mélanges réticulés et non réticulés ont été déterminés en utilisant deux rhéomètres avec deux modes de cisaillement, c-à-d, le rhéomètre oscillatoire (Rubber Process Analyser, RPA2000) et le rhéomètre capillaire à vitesse contrôlée (Goettfert Rheotester 2000). Les résultats obtenus révèlent que le comportement viscoélastique des mélanges est influencé par la formation d'un réseau transitoire de silice, particulièrement dans les mélanges réticulés. Le temps de mélange affecte significativement les propriétés viscoélastiques des mélanges vulcanisés, ce qui est probablement du au grand degré de dégradation thermique, tandis qu'il ne joue pas de rôle significatif pour les propriétés viscoélastiques des mélanges non vulcanisés. La superposition des résultats oscillatoires et de cisaillement en régime établi est possible quand la composante élastique est éliminée des résultats.

KEY WORDS: Viscoelastic properties, rheology, chlorinated polyethylene, natural rubber, silica

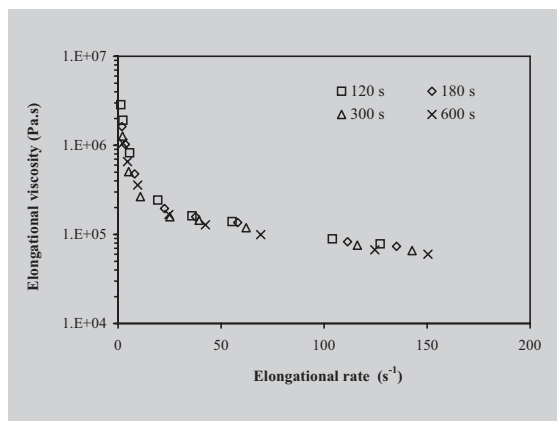


Figure 13 (left): Elongational viscosity as a function of elongational rate of uncured blends as determined from capillary results.

Figure 14 (middle): Plots of apparent shear viscosity measured by capillary rheometer (■) and complex viscosity determined from RPA 2000 (□) as a function of shear rate or frequency in filled uncured CPE/NR blends with various mixing time.

Figure 15 (right): Plots of true shear viscosity measured by capillary rheometer (■) and dynamic viscosity determined from RPA 2000 (□) as a function of shear rate or frequency in filled uncured CPE/NR blends with various mixing time.

transient network. Additionally, results of all blends fall on a single correlation, implying that the time of mixing used in this study does not influence significantly the elongational behaviour.

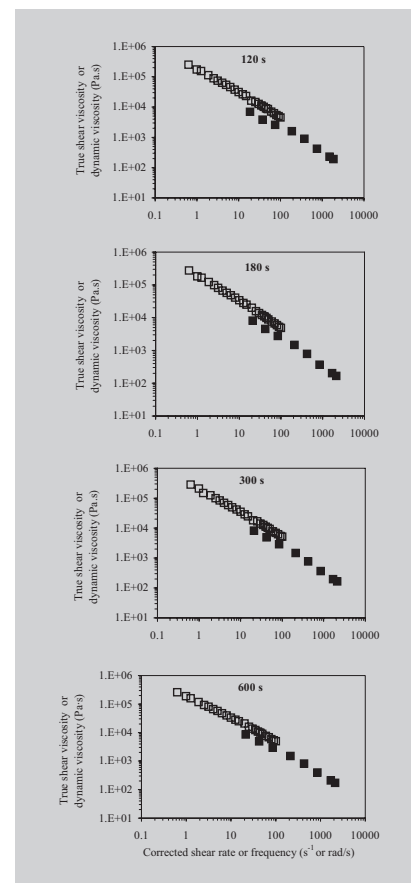
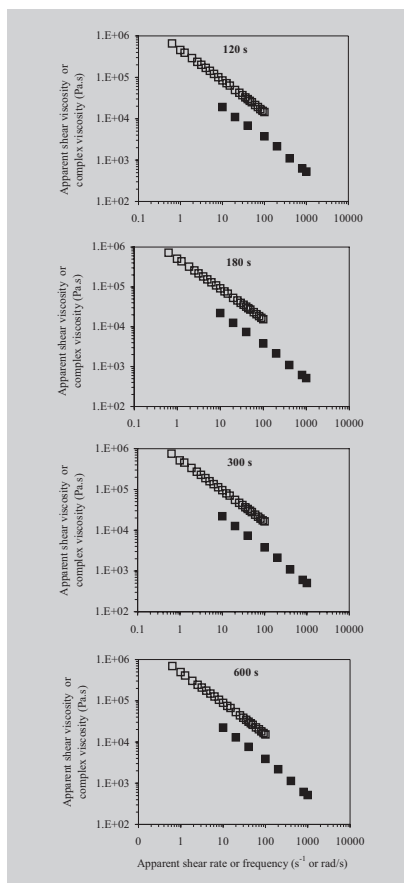
Fig. 13 exhibits a comparison of $\eta^*(\omega)$ and $\eta_a(\dot{\gamma}_a)$ of the blends with different mixing time deter-

mined from oscillatory and steady shear tests, respectively. It is obvious that all blends do not obey the Cox-Merz concept, i.e., $\eta^*(\omega)$ is greater than $\eta_a(\dot{\gamma}_a)$. The explanation is based on the fact that the strain imposed to the blends in the oscillatory flow with in the LVE region is much smaller than that in the capillary flow. Consequently, the presence of silica transient network would promote elastic response and thus the $\eta^*(\omega)$. Similar observation has been reported in CPE/NR blends with different blend ratio and silica loading [12, 13].

To correlate the oscillatory to steady shear data more effectively, the dynamic viscosity, η' , and true shear viscosity, η_{true} , are plotted, as illustrated in Fig. 15. Apparently, the discrepancy of steady and oscillatory shear results is markedly reduced, which suggests that the Cox-Merz concept is valid to some extent in filled systems when the elastic component is eliminated. The result is in good accordance with previous work reported elsewhere [13].

4 CONCLUSIONS

From all obtained rheological results of 80/20 CPE/NR blends with various mixing time, the following conclusions could be drawn: The formation of silica transient network associated with the strong interaction between silanol group on silica surfaces and chlorine atoms of CPE plays role in viscoelastic properties of blends to some



extent, especially in the case of cured blends. Mixing time significantly influences viscoelastic properties of vulcanised blends which is due probably to the high extent of thermal degradation, but does not affect significantly in uncured blends. The validity of a correlation between oscillatory and steady shear results via the Cox-Merz concept is mainly controlled by the presence of silica transient network effect rather than the mixing time effect, and the superimposition of oscillatory and steady shear results is possible when the elastic component is eliminated from the apparent results.

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