Dynamic Viscoelastic Properties of Organic / Inorganic Fibres Reinforced LLDPE Composites in Molten State

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Abstract:
Dynamic rheological parameters such as storage modulus, *G'*, loss modulus, *G''*, and dynamic viscosity, η', at 200°C were studied for Kevlar fibres, glass fibres and their hybrids reinforced linear low density polyethylene (LLDPE). Parallel plate rheometer was employed for these tests. *G'*, *G''* and η' increased with the increased reinforcement and angular frequency, ω. Two sets of reinforcement, 10 and 20 vol.% of fibres are used in LLDPE. The composition of fibres in hybrid composites was varied. The replacement of glass fibres with Kevlar increases the values of *G'*, *G''* and η'. The values of these rheological parameters also increased with the thickness of the composite. This increase was associated with the decreased average orientation of fibres present in the composite. The effects of the change in strain amplitude on *G'* and *G''* is also studied and reported here.

Zusammenfassung:
Dynamische rheologische Parameter wie zum Beispiel der Speichermodul *G'*, der Verlustmodul *G"* und die dynamische Viscosität η' wurden bei 200°C an LLDPE gemessen, welches mit Kevlarfasern, Glasfasern oder beidem verstärkt wurde. Für diese Tests wurde ein Parallelplattenrheometer verwendet. Mit zunehmender Faseranteil und Frequenz ω steigen *G'*, *G"* und η' an. Zwei LLDPE Proben mit 10 und 20 vol.% Faseranteilen wurden untersucht wobei die Zusamensetzung der Fasern in Hybridkompositen ebenfalls variert wurde. Durch Ersetzen der Glasfasern durch Kevlarfasern erhöhen sich die Werte für *G'*, *G"* und η'. Die Werte dieser rheologischen Parameter steigen auch mit zunehmender Dicke des Komposits an. Dieses Verhalten wird durch die verminderte mittlere Orientierung der im Komposit anwesenden Fasern verursacht. Im übrigen wurde auch der Einfluss der Belastungsamplitude auf *G'* and *G"* untersucht.

Résumé:
Les paramètres rhéologiques dynamiques, tels que le module de rigidité G', le module de perte G'' et la viscosité dynamique, h', à 200°C, ont été étudiés pour des fibres de Kevlar, des fibres de verre et des polyethylènes linéaires de basse densité (LLDPE) réenforces avec ces fibres. Un rhéometre plan-plan a été employé pour ces tests. G', G'' et h' présentent une augmentation avec le réenforcement et la fréquence angulaire w. Deux types de réenforcement, 10 et 20 % en volume de fibres, ont été utilisés avec LLDPE. La composition en fibres dans les composites hybrides a été variée. Le remplacement des fibres de verre par les fibres de Kevlar, s'accompagne par une augmentation des valeurs de G', G'' et h'. Les valeurs de ces paramètres rhéologiques augmentent aussi avec l'épaisseur du composite. Cette augmentation a été associée à la diminution de l'orientation moyenne des fibres présentes dans le composite. Les effets associés au changement de l'amplitude de la déformation sur les modules G' et G'' furent égalements étudiés et sont ici présentés.

Key words: Kevlar fibre, glass fibre, LLDPE, dynamic rheology, hybrid composites, orientation.

1 INTRODUCTION

Fibre reinforced polymer composites are being used in various industrial applications due to their high specific strength as compared to the metals and ceramics. Two types of polymers; thermoplastics and thermosetting resins are used as matrix material in such composites. The fast production cycle, low tooling cost and ease in processing have broadened the area of application of stampable thermoplastic composites [1-2]. The semi crystalline polyolefins are generally used as matrix due to their high chemical resistance, ease of fabrication, high impact strength, low temperature toughness coupled with the low cost. The addition of inorganic short glass fibres to a polyolefin matrix generally leads to the significant improvement in mechanical properties [3-9]. In some cases, the toughness of matrix material deteriorates on addition of glass

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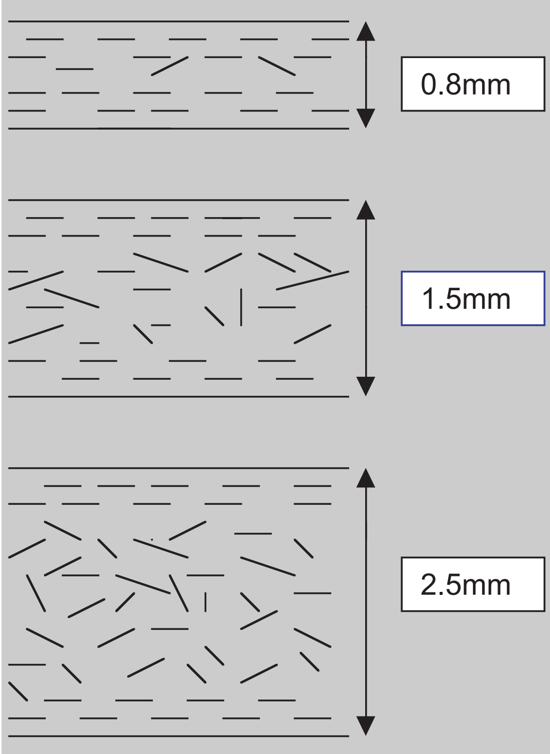

Figure 9 (left): Schematic presentation of distribution and orientation of fibres in composites with different thickness (0.8, 1.5, and 2.5 mm).

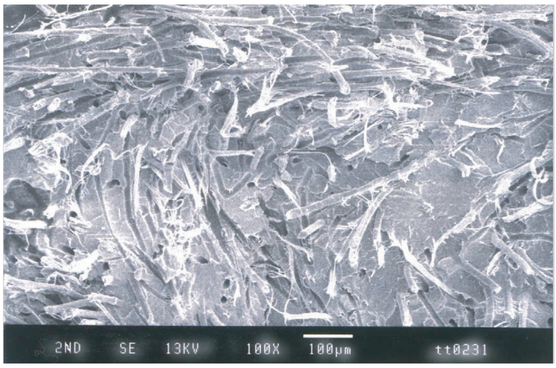

Figure 10 (right above): Cryogenically fractured surface of LL-GF/KF (80-0/100) composite.

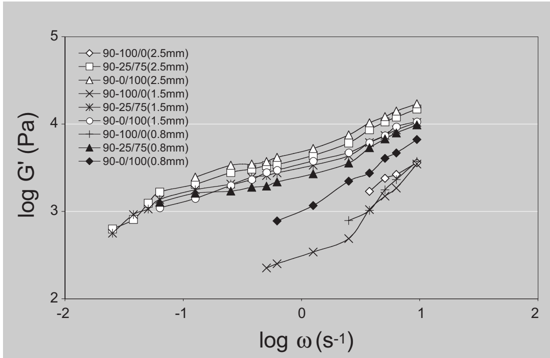

Figure 11 (right below): Storage modulus, G', as a function of angular frequency, ω, for composites having different thickness and composition at θ = ± 1°.

dle layer is composed of randomly oriented fibres. This happens during the fabrication of composite samples. The pressure is applied on the composite material in one direction, say for example, in z direction and thus the fibres preferably took their positions in x-y plane because of the possible movement in that plane only. Secondly, heat is supplied to material through the hot platen. The material in contact with the hot platen of the press would be at a higher temperature as compared to the material at middle position. It is also expected that the fibres can easily move and align in a plane near the platen. The gradient temperature effect would be significant in higher thickness in which skin part of composite sample would show the orientation of fibres in a plane whereas the middle portion would be less affected and shall remain randomly oriented. Fig. 9 shows an schematic presentation of distribution and orientation of fibres in 0.8, 1.5 and 2.5 mm thick composites as discussed above. Fig. 10 shows the fractograph of skin and middle part of composite of 2.5 mm thickness. The top portion of the fractograph shows a part of the top layer in which the direction of fibres is preferably horizontal. The lower part of fractrograph, which represents the middle layer, shows random orientation of fibres.

Figs. 11, 12, and 13 compare the variation of *G'*, *G"*, and *η'* with *ω* respectively for composites of varied average fibre orientation (thickness of composite samples). For Kevlar and glass/Kevlar hybrid fibre reinforced LLDPE, *G'*, *G"*, and *η'* increase with *H* as shown here. An increase in *H* gradually increases the random distribution of fibres in the composites resulted from the fabrication method employed for making these composites. The trend of increasing *G'*, *G"*, and *η'* with *H* was not clear in short glass fibre reinforced LLDPE composites.

Apart from the changed morphology the strain amplitude also varies with *H*. The strain amplitude, *γ*, is related with the oscillatory angle, *θ*, radius of plates, *R*, and the distance between the parallel plates, *H*, of a rheometer by the fol-

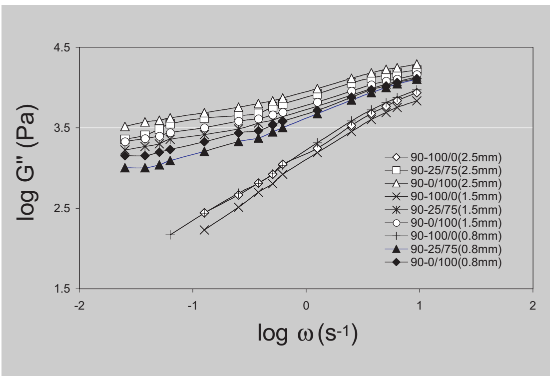

Figure 12 (left): Loss modulus, G", as a function of angular frequency, ω, for composites having different thickness and composition at θ = ± 1°.

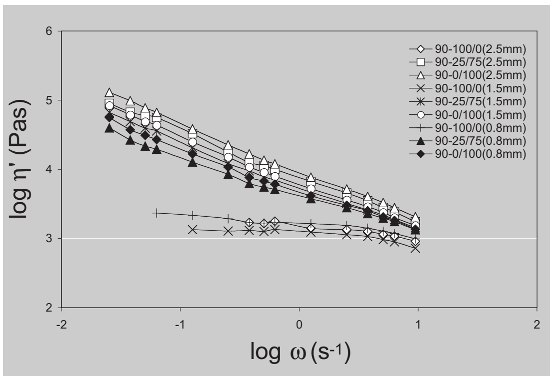

Figure 13 (right): Dynamic viscosity, η', as a function of angular frequency, ω, for composites having different thickness and composition at θ = ± 1°.

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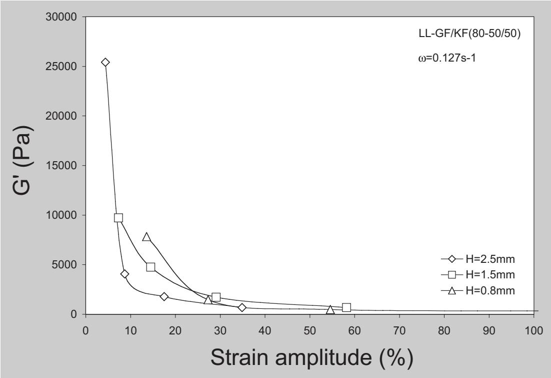

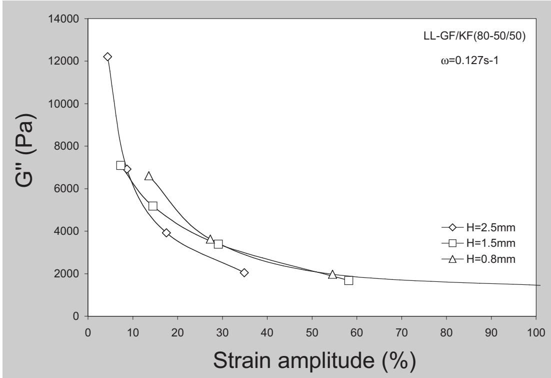

Figure 14 (left): Storage modulus, G', as a function of strain amplitude, γ, for LL - GF/KF (80-50/50) at $\omega = 0.127\ s^{-1}$.

Figure 15 (right): Loss modulus, G'', as a function of strain amplitude, γ, for LL - GF/KF (80-50/50), at $\omega = 0.127\ s^{-1}$.

lowing relation

$$\gamma = \frac{\theta R}{H} \tag{4}$$

In present heterogeneous system, the change in *H* is associated with the change in average orientation of fibres in the composite and hence affects the dynamic rheological properties of composites differently as compared to those affected by changing the oscillatory angle.

In Fig. 14 and 15, *G'* versus γ and *G''* versus γ, respectively, are shown. These data were obtained at different thickness and oscillatory angles. The strain amplitude, γ, was calculated by using Eq. 4. The values of γ at ± 0.5° θ angle for 0.8, 1.5 and 2.5 mm *H* values were 13.6, 7.3 and 4.4 %. Similarly values corresponding to ± 1, ± 2, ± 4° θ angle were obtained and plotted in Figs. 14 and 15. The values of *G'* and *G''* decrease sharply with the increased strain amplitude percentage at lower values of γ which however become weakly sensitive to the increased strain amplitude.

CONCLUSIONS

Dynamic viscoelastic properties of Kevlar fibres, glass fibres and their hybrid reinforced LLDPE composites were studied at 200° C. Kevlar fibres, glass fibres and their hybrids increase the values of *G'*, *G''*, and η' of the composite. The replacement of glass fibre by Kevlar fibres increases *G'*, *G''*, and η' at constant loading of fibres in LLDPE. The decreased average orientation of fibres in the composites increases the values of *G'*, *G''*, and η'. The values of *G'* and *G''* decrease sharply with the increased strain amplitude.

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