

This article was downloaded by:

On: 24 January 2011

Access details: *Access Details: Free Access*

Publisher *Taylor & Francis*

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Journal of Macromolecular Science, Part A

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713597274>

Preparation and Rheological Characterization of Polymer Nanocomposites Based on Expanded Graphite

Mahmoud Abdel-Goad^{ab}; Petra Pötschke^a; Donghui Zhou^c; James E. Mark^c; Gert Heinrich^a

^a Leibniz Institute of Polymer Research Dresden, Dresden, Germany ^b Chemical Engineering Department, Faculty of Engineering, Minia University, Egypt ^c Department of Chemistry, The University of Cincinnati, Ohio, USA

To cite this Article Abdel-Goad, Mahmoud , Pötschke, Petra , Zhou, Donghui , Mark, James E. and Heinrich, Gert(2007) 'Preparation and Rheological Characterization of Polymer Nanocomposites Based on Expanded Graphite', Journal of Macromolecular Science, Part A, 44: 6, 591 – 598

To link to this Article: DOI: 10.1080/10601320701284840

URL: <http://dx.doi.org/10.1080/10601320701284840>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Preparation and Rheological Characterization of Polymer Nanocomposites Based on Expanded Graphite

MAHMOUD ABDEL-GOAD,^{1,2} PETRA PÖTSCHKE,¹ DONGHUI ZHOU,³ JAMES E. MARK,³ and GERT HEINRICH¹

¹Leibniz Institute of Polymer Research Dresden, Dresden, Germany

²Chemical Engineering Department, Faculty of Engineering, Minia University, Egypt

³Department of Chemistry, The University of Cincinnati, Ohio, USA

Received and accepted January, 2007

A new type of conductive filler, namely expanded graphite (EG), was used to prepare novel nanocomposites. The EG was incorporated into several rather different polymers, specifically polycarbonate (PC), low-density polyethylene (LDPE), isotactic polypropylene (PP), and polystyrene (PS), using melt mixing in a small-scale DACA-Microcompounder. The EG content was varied between 1 and 20 wt%. The rheological properties and morphologies of the nanocomposites were characterized by melt rheology and scanning electron microscopy (SEM), respectively. The melt-state linear viscoelastic properties were investigated using an ARES rheometer, with the measurements performed in the dynamic mode at various temperatures over a wide range of frequencies. Addition of the EG increased the linear dynamic moduli and melt viscosity of the materials. Up to a certain critical concentration of EG, the materials exhibited a simple liquid-like behavior. Above this concentration, however, significant changes in the frequency dependences of the moduli and viscosity were observed. In addition, the moduli showed a liquid-solid transition resulting in a second plateau in the low frequency-regime, and the complex viscosity revealed shear-thinning behavior. Specific values of this percolation concentration were found to be at around 4 wt% in the case of PC/EG, 9 wt% for PP/EG and PS/EG, and 12 wt% for PE/EG. This critical concentration was correlated to a network-like structure formed through interactions between the EG platelets and the polymers. The extent of these complications was found to vary from polymer to polymer, presumably due to different degrees of EG exfoliation and dispersion arising from different EG-polymer interactions and from variable shearing forces dependent on the polymer viscosities. The formation of network-like structures is very sensitively displayed using van Gurp-Palmen plots, which are most suitable for identifying “rheological percolation” in our investigated systems.

Keywords: expanded graphite; nanocomposites; rheology; liquid-solid transition; modulus; viscosity

1 Introduction

Layers of expanded graphite have become of intense interest as fillers in polymeric nanocomposites. By way of illustration, one can readily cite studies focusing on such materials with regard to their structures and morphologies (1–7), mechanical properties (8), rheology (9), tribology (10), even piezorestrictive behavior (11). However, most of the studies have concentrated on *electrical conductivity* of the materials (11–22). The primary reason is that only small amounts of expanded graphite are sufficient to reach the threshold concentration for percolation conductivity, because of the strongly anisometric nature of these sheet-like fillers.

The present interest in EG fillers follows and parallels earlier work on polymers containing organo-modified clays or carbon nanotubes. At least some of the polymers mixed with highly expanded EG are certainly nanocomposites since it has been clearly demonstrated that the dimensions of the EG sheets are in the nanometer scale (15, 16), with thicknesses varying from 10 to 50 nm (18). Such systems can provide cost-effective production of materials with improved functional properties such as stiffness, barrier properties and the already mentioned electrical conductivity. Of particular interest is the fact that several investigators reported that a remarkably low volume fraction of EG was sufficient to reach a percolation threshold in different polymeric systems (17). These properties would give EG nanocomposites a wide range of potential applications such as radiation and electromagnetic shielding, antistatic properties, coatings that are shrinkage-resistant and corrosion-resistant, etc. (18).

The present study reports the preparation of EG-reinforced polymer nanocomposites by melt processing, which has the

Address correspondence to: James E. Mark, Department of Chemistry, Crosley Tower, Martin Luther King Drive, The University of Cincinnati, Cincinnati, OH 45221-0172, USA. Tel.: 513-556-9292; Fax: 513-556-9239; E-mail: markje@email.uc.edu

advantage of being an established and practical method for composite preparation for potential industrial applications. The polymers for the investigation were chosen to be significantly different, including amorphous and crystalline polymers and polymers having very different surface tensions, and they were used with a relatively wide range of EG concentrations. The major interest in this part of an extensive series of experiments was the effects of the EG on the melt rheological behavior of the nanocomposites. Of special interest was the determination of the rheological percolation-threshold concentrations. This is obviously closely related to the threshold concentrations required for the electrical conductivity mentioned above. In a broader context, the present study is part of an ongoing extensive effort to use melt rheology as a comparative indirect measure for EG spatial distributions, degrees of dispersion, and interactions across the filler-polymer interfaces.

2 Experimental

The expanded graphite (EG) was prepared at the University of Cincinnati starting from flakes with an average size of 500 μm , provided by the Asbury Graphite Mills, Inc. (Asbury, USA). The expansion was performed in a manner similar to that reported in the literature (15), i.e., by heating graphite flakes, pre-treated and in contact with sulphuric acid, at approximately 1,000°C for 30–60 s.

The expanded graphite was incorporated into samples of (i) (amorphous) polycarbonate (PC, Lupilon E 2000, Mitsubishi, Japan), (ii) isotactic (crystallisable) polypropylene (PP, Moplen HF500N, Basell Polyolefins), (iii) atactic (amorphous) polystyrene (PS, weight-average molecular weight $M_w = 280$ kg/mol, polydispersity index $PD = \sim 2$), and (iv) low-density (crystalline) polyethylene (LDPE, LD252, Exxon Mobil, Belgium). Commercial materials were intentionally chosen to make the results of greater practical importance, in spite of the fact that such samples sometimes contain small amounts of undisclosed additives such as processing aids.

The EG flake material and the polymers in the form of granules or powders were premixed in the dry state, at various compositions, before being fed into an already-running mixer. The mixer was a DACA-Microcompounder (small-scale conical co-rotating twin screw compounder with a capacity of 4.5 cm^3 , from DACA Instruments, Goleta, USA). This corresponded to “melt” mixing, in that temperatures were above the melting points T_m of the two crystalline polymers, or above the glass transition temperatures T_g of the two amorphous polymers. Specifically, the mixing temperatures were 280°C in the case of the PC and 220°C for the PP, PS, and PE. A mixing speed of 50 rpm and a mixing time of 5 min were used. The melt-state linear viscoelastic properties for the polymers were studied using an ARES rheometer (Rheometric Scientifics, USA). In preparation for these tests, the extruded materials were molded

at processing temperatures into sheets with a thickness of 0.9 mm, from which 8 mm diameter discs were punched.

Frequency sweeps (from high to low values) were performed using a parallel-plate geometry, at low strains and under a nitrogen atmosphere. The samples were measured at different temperatures over a wide range of frequencies (100–0.03 rad/s). The results presented here focus on measurements obtained at the mixing temperatures chosen, as cited above. It has to be mentioned that in case of PS/EG, the data points scattered, especially at very low frequencies. For this reason, an unusually large number of points were obtained (10 points/decade), and the trend-fit line was used to give a rough indication of the trends for the PS/EG nanocomposites having different compositions, at very low frequencies.

In addition, scanning electron microscopy (SEM Leo VP 435) was used on the EG itself, and on cryo-fractured surfaces of some of the compression-molded sheets.

3 Results and Discussion

3.1 Morphological Investigations of the Materials

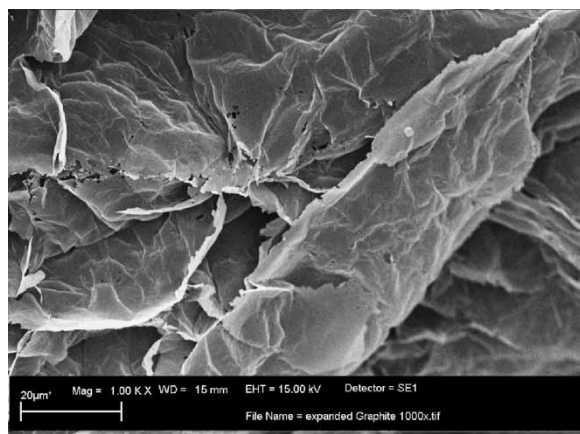
The SEM results on the EG itself are illustrated in Figure 1a, and clearly show a series of layered structures, as expected. The similarly obtained results on the morphologies on the representative PC/EG nanocomposites (Figure 1b) illustrate success in achieving good spatial distributions and dispersions of the EG platelets throughout the entire PC matrix.

3.2 Melt Rheological Behavior of the Polymer-EG Nanocomposites

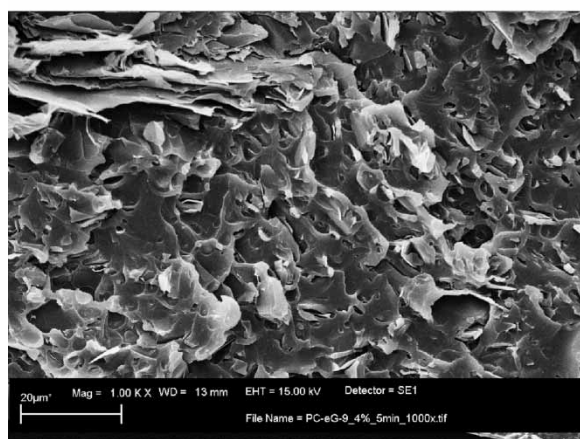
Oscillatory melt rheology is particularly relevant here since it is known that interconnected structures of anisometric fillers lead to marked qualitative changes in the spectra of dynamic moduli and viscosity. Specifically, the complex viscosity changes from a Newtonian plateau to a continuous increase with decreasing frequency, whereas the storage modulus G' and loss modulus G'' flatten significantly and merge into a secondary plateau at low frequencies (23–28).

Figures 2–5 show the frequency (ω) dependence of the shear storage modulus G' and complex viscosity $|\eta^*|$ at 280°C for the EG-based PC and at 220°C for PP, PS and PE nanocomposites, respectively. The addition of EG to the polymers clearly leads to increases in both G' and $|\eta^*|$ in all the systems investigated. The improvements in the dynamic mechanical moduli are an expected outcome in view of the known reinforcement abilities of EG sheets. EG has a high surface area-to-volume ratio based on a high aspect ratio (diameter to thickness) of the platelets (17), and this significantly enhances the interactions between the EG and the polymer chains.

In general, the changes are much lower than those observed when using multi-walled carbon nanotubes (26). In the present case, the measurements were carried out on pressed samples in which a preferential orientation of the EG platelets



(a)

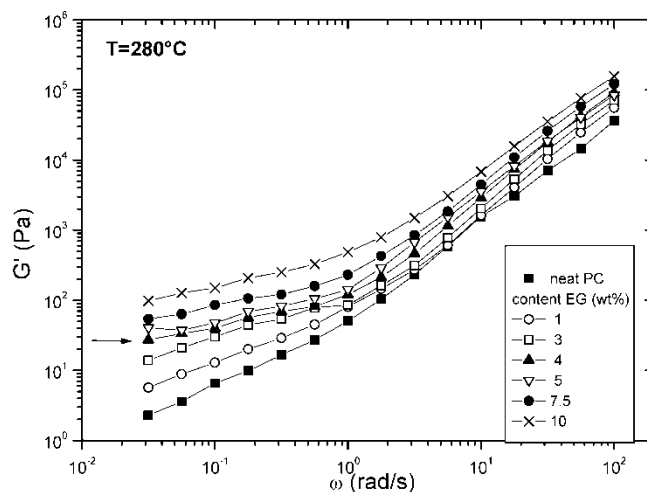


(b)

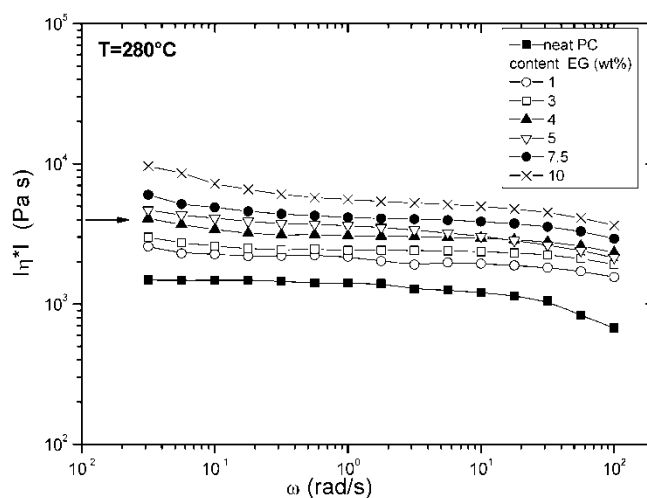
Fig. 1. Scanning electron micrographs (without sputtering) for (a) pure EG, (b) cryofractured surfaces of a compression-molded sample of polycarbonate having 4 wt% expanded graphite (EG).

and sheets parallel to the plate direction may have occurred, as suggested by some of the results in Figure 1b. This is an inherent problem and difficult to overcome when using melt processing. However, the impact of EG, i.e., the magnitude of the increases, and the dependences of G' and $|\eta^*|$ on measuring frequency, are different in the different polymer matrices. Even if the matrix viscosities do not differ very much, the highest impact was seen in PC, followed by PP and PS. In PE, only very low enhancements were observed up to 15 wt% EG.

The relative changes in storage modulus G' and complex viscosity $|\eta^*|$ at low frequencies for all four polymers are summarized in Figure 6, where the values at a given concentration are related to those for the neat material (or for the lowest EG content prepared). It is clearly seen that the impact in G' was much higher than that for the complex viscosity. For PC, a G' value 43 times higher than for the neat PC was found at 10 wt% EG addition. In PP and PS the addition of 9 wt% EG lead to an increase by a factor of about 3, whereas in PE no change at all was found at 9 wt% EG. At



(a)



(b)

Fig. 2. Frequency dependence of (a) shear storage modulus G' and (b) complex viscosity $|\eta^*|$ measured at 280°C for the PC/EG nanocomposites.

higher content (20 wt% EG), PP showed larger improvement than PS. Addition of 15 wt% EG in PE lead to a G' increase of 2.8. Similar trends were found for the complex viscosity at low frequencies, but the increase at 10 wt% EG in PC was by a factor of only 6. The different impacts of EG in the polymers used is expected to be mainly influenced by different exfoliation of the EG sheets during the mixing procedure. The different degrees of exfoliation, in turn, may be caused by different wetting behavior and interfacial tensions between the EG material and polymers. Different extents of exfoliation would of course change the active surface areas of the EG platelets and thus the degrees of interaction between the polymer chains and the EG filler.

Even if the melt viscosities of the matrix polymers at high shear rates at the corresponding mixing temperatures do not differ very much (all were in the range between 200 and 700 Pa · s), small changes may lead to different shearing

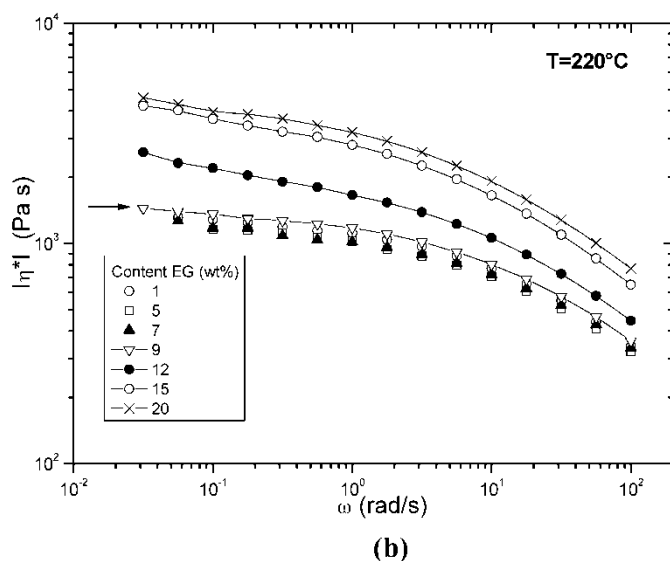
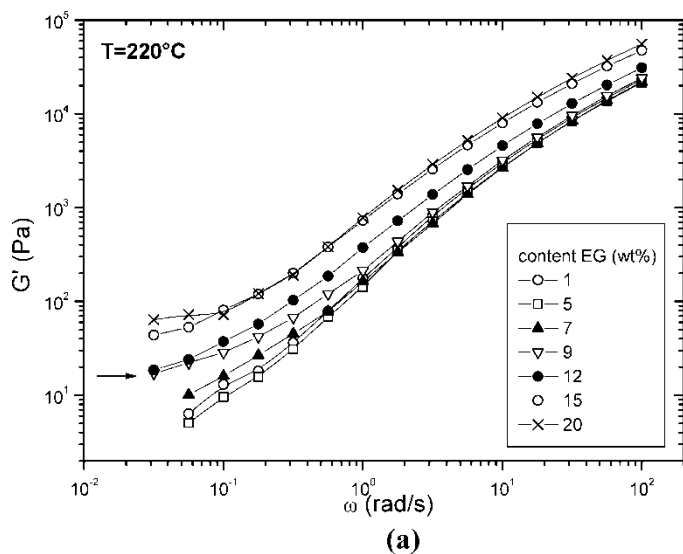


Fig. 3. Frequency dependence of (a) shear storage modulus G' and (b) complex viscosity $|\eta^*|$ for the PP/EG nanocomposites at 220°C.

forces during mixing, also tending to yield different degrees of exfoliation. We have measured the surface energy of the EG used here and found a dispersive value of 27.3 mJ/m² (30). All polar contributions were estimated to be negligible. This value is similar to values for other graphite structures such as carbon blacks and carbon nanotubes. Recently, using different methods, we have carefully estimated the free surface energy (and its corresponding dispersive and polar parts) for several types of carbon blacks (N121, N234, N234 graphitized, N339, N990) and several types of multi-walled carbon nanotubes. Values for the total free surface energy were found to be in the range 25–30 mJ/m², and all polar contributions were estimated to be negligible (30).

According to the literature, the surface energies of the polymer melts are as follows: at 280°C the value for PC

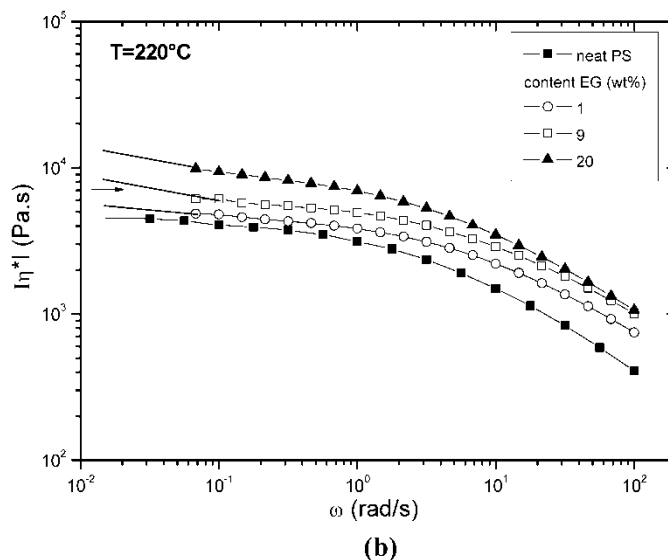
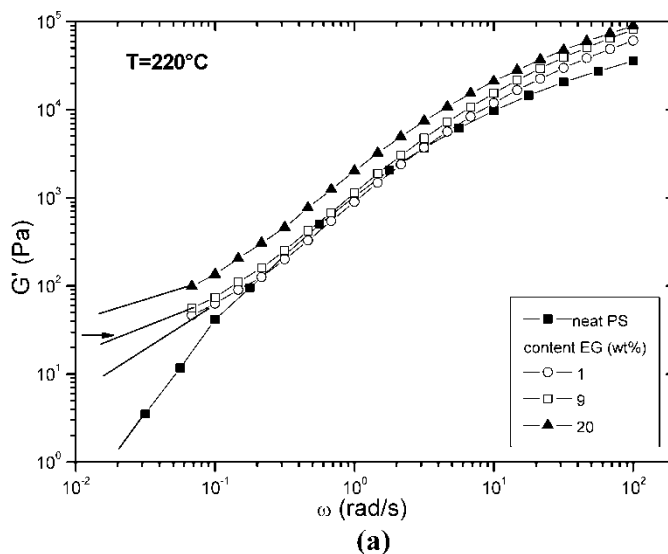


Fig. 4. Frequency dependence of (a) shear storage modulus G' and (b) complex viscosity $|\eta^*|$ for the PS/EG nanocomposites at 220°C.

was 23.8 mJ/m², and at 220°C the values for PP were 16.7 mJ/m², for PS 26.3 mJ/m², and for PE 24.3 mJ/m² (31). These values indicate that surface tension alone cannot explain the large differences between the properties of the polymer-EG pairs. In addition, the polarities of the materials have to be taken into account; values are zero for the polyolefins, but in the range of 20% for PC and 15% for PS (31). Thus, it may be expected that PP and PE would show the best compatibility with EG. Interestingly, the nonpolar, low surface tension PP, having also the lowest melt viscosity in the frequency range that is characteristic for the mixing in our equipment, showed a much higher effect on G' and complex viscosity $|\eta^*|$ than did PE.

As also indicated in Figures 2–5, there were typical changes in the frequency dependences of the moduli and viscosity at a certain (critical) concentration of EG. Below this

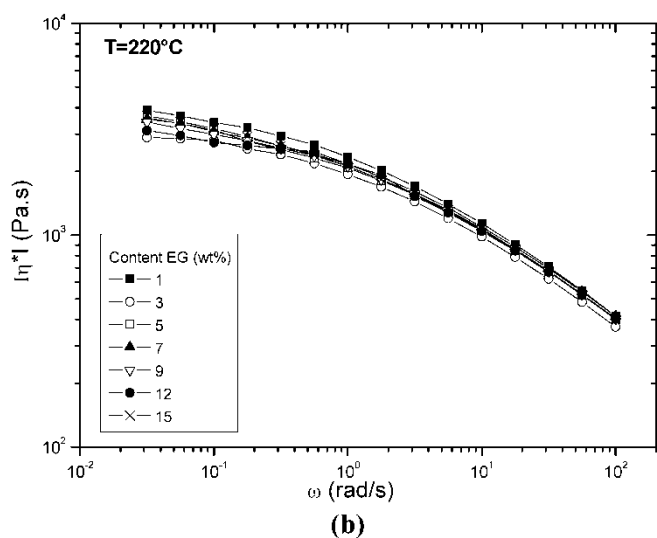
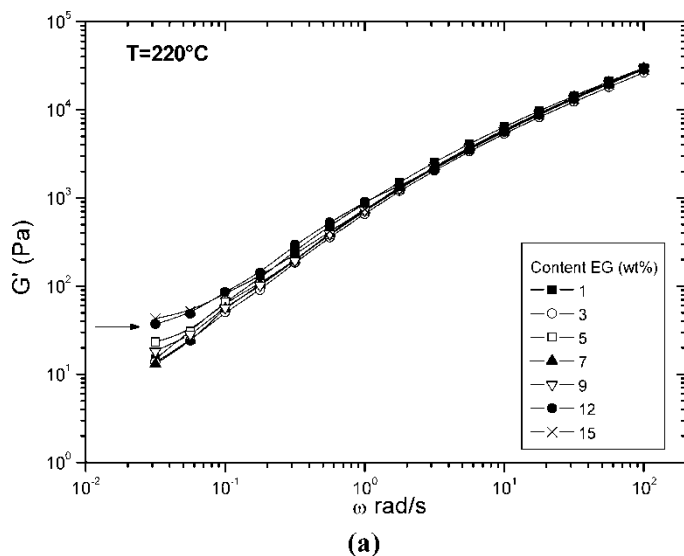


Fig. 5. Frequency dependence of (a) shear storage modulus G' and (b) complex viscosity $|\eta^*|$ for the PE/EG nanocomposites at 220°C.

concentration the materials exhibited a liquid-like behavior, but above it the qualitative behavior of G' and $|\eta^*|$ changed markedly. The moduli showed a liquid-solid transition resulting in a second plateau in the low-frequency regime, and the complex viscosity revealed shear-thinning behavior. These changes are expected from the formation of a network-like structure resulting from interactions between the EG platelets and the polymers. This critical concentration is called the “rheological percolation threshold concentration”. Such percolation could be clearly observed for many other nanocomposite systems, such as layered double hydroxides (25) or multiwalled carbon nanotubes (26). In the systems investigated here, however, it was more difficult to identify such a transition unequivocally since, for a presently unknown reason, the changes were more gradual. The rheological percolation composition is seen to occur between 3 and 4 wt% in

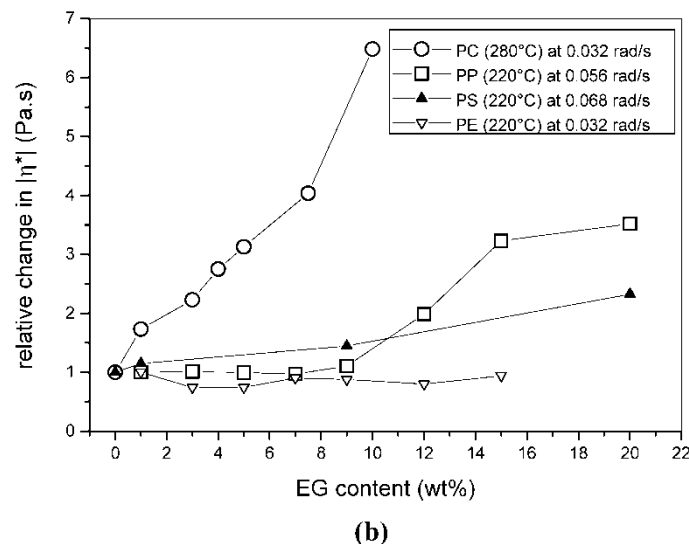
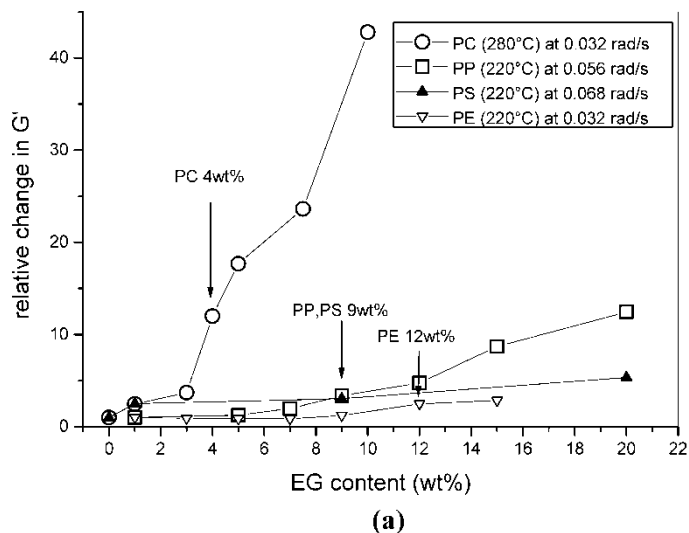


Fig. 6. Relative changes in the shear storage modulus and complex viscosity at very low frequencies (value at given EG content divided by that for the neat material or lowest EG content) vs. EG content: (a) G' and (b) $|\eta^*|$.

the case of PC (Figure 2), between 7 and 9 wt% in the case of PP (Figure 3, clearly apparent only in G'), below 9 wt% for PS (Figure 4), and between 9 and 12 wt% for PE (Figure 5, clearly apparent only in G'). The concentration at which percolation occurred is marked by an arrow in the plots. As already explained, this order correlates with the physical interactions between the polymer chains and the EG. The higher the degree of exfoliation of the EG, the larger the interactions, and the lower the concentrations at which a rheological network between polymer chains and platelets was formed. Similar results concerning rheological percolation concentration become evident when looking at the relative changes at low frequencies vs. EG content, as shown in Figure 6. These figures reveal the effects of EG loadings on G' and $|\eta^*|$ before and after reaching the rheological percolation concentration. Below this concentration (again marked

by an arrow) the moduli and viscosities increase gradually and only slightly with increasing EG content. Beyond this concentration, G' and $|\eta^*|$ increase more strongly. Possible methods for a more exact determination of the rheological percolation threshold composition are discussed below in greater detail.

3.3 Determination of the Rheological Percolation Concentration

In order to determine the rheological percolation concentration more accurately, the data for PC and PP showing the biggest impact on EG addition were analyzed and plotted in different ways. First, the slope of the storage modulus G' at low frequencies (known to be very sensitive to network formation) was plotted vs. EG content. For this, Figures 2a and 3b were used and the range of linear dependence of G' on frequency was selected. Linear fits in double-logarithmic scales were obtained for PC data in the range lower than 1 rad/s, and for the PP results were based on three to five values. The fitted values are plotted in Figure 7. First, it is obvious that our neat materials do not exhibit the terminal slopes of G' and G'' of 2 and 1, respectively (i.e., $G' \sim \omega^2$ and $G'' \sim \omega$ as $\omega \rightarrow 0$). It should be noted that these are industrial materials that are not polydisperse and, as previously mentioned, may contain small amounts of additives such as processing aids. The slope values obtained for G' , irrespective of whether presenting a power-law dependence of G' , were much smaller than the expected value of approximately 2. For PC, we found a decrease in the power-law dependence of G' already at low EG concentrations, which levels off to a constant value of about 0.4, starting at 4 wt% EG addition. For PP, the slope of G' in the terminal regime decreased only slightly up to 7 wt% but showed a sharp decrease starting at 9 wt%, where the values also started to level off to approximately the same value as that for PC.

Another way to look at structural changes in polymeric materials is to plot G'' versus G' , in what is known as a

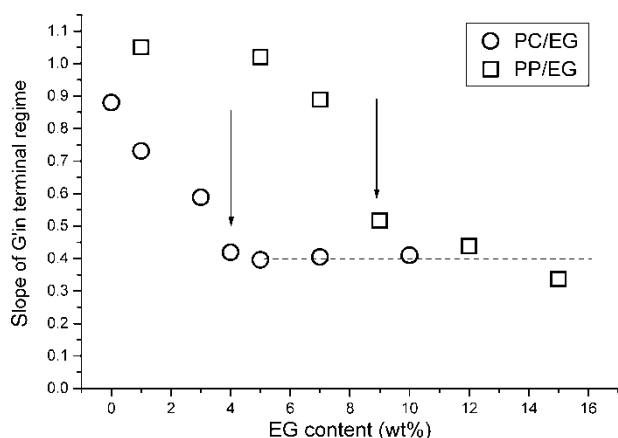


Fig. 7. Slope of G' at low frequencies (terminal regime) for PC/EG and PP/EG composites as a function of EG content.

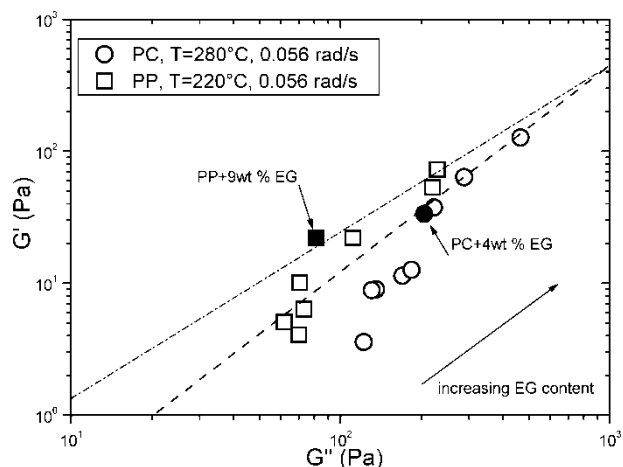


Fig. 8. Relationships between G' and G'' at low frequency for PC and PP nanocomposites.

modified Cole-Cole-plot. Such plots can be used to elucidate structure differences between different polymer modifications. For example, Harrell and Nakayama (32, 33) used plots of $\log G''$ vs. $\log G'$ to explore the influence of branching and the broadening of the molecular weight distribution of PE on microstructure. In our case, we plotted a type of reduced Cole-Cole plot by presenting G' vs. G'' values at 0.056 rad/s and different compositions (Figure 8). Both G' and G'' increased with EG content, as is expected for solid filler additions. Above a specific composition, there appeared a linearity in G' vs. G'' . For PC this composition was 4 wt% and all concentration above that show linearity between G' and G'' , whereas for PP this occurred at 9 wt%. All lower concentrations give values lying significantly below the line for linearity between G' and G'' .

Another type of plot very sensitively displays the formation of network-like structures in nanofilled systems. In it, the phase angle δ is plotted against the absolute value of the complex modulus $|G^*|$. This plot is known as “van Gurp-Palmen plot” and has been recommended for identifying ‘rheological percolation’. An example is given by Meincke et al. (34) for polyamide 6/multiwalled carbon nanotube composites. This plot is shown for the PC and PP nanocomposites in Figures 9 and 10, respectively. These results establish with greater accuracy the critical concentration at which the so-called “rheological” (or “mechanical”) percolation threshold occurred. This threshold value was taken to be the concentration at which the phase angle decreased significantly with decreasing complex modulus. This drop in phase angle is related to the development of the elasticity due to the network structure formed by the EG layers and the polymer chains. The values of the critical concentrations as obtained from the van Gurp-Palmen plots were between 3 and 4 wt% for PC and between 7 and 9 wt% for PP. In contrast to the previous plots, the percolation concentration can be located unequivocally from this type of plot.

In a next step it would be interesting to compare these rheological results with electrical conductivity measurements. It

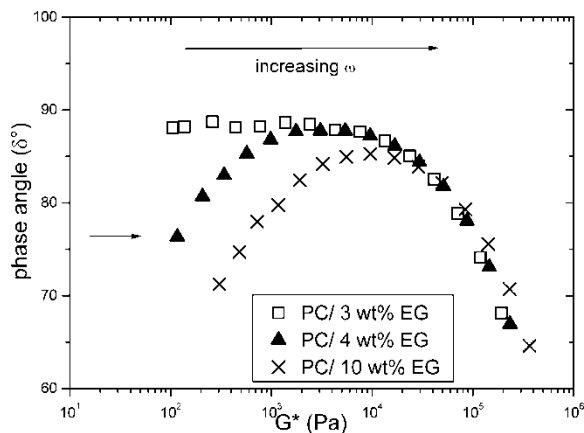


Fig. 9. Phase angle vs. G^* for PC nanocomposites at 280°C.

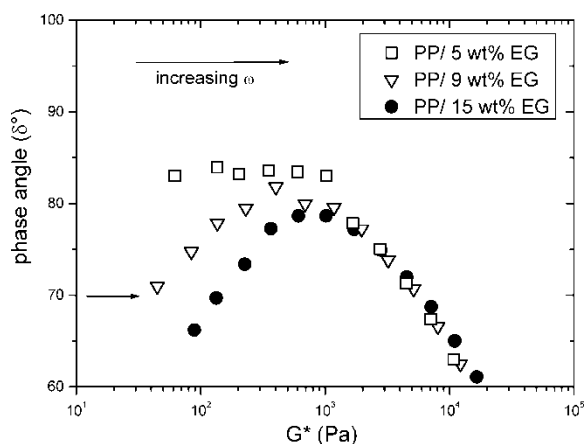


Fig. 10. Phase angle vs. G^* for PP nanocomposites at 220°C.

was already observed that the PC sample with 4 wt% EG had percolated sufficiently to show such conductivity, and studies on the other matrix polymers are in progress.

4 Conclusions

The present results documented the effects of expanded graphite (EG) content on the rheological properties of nanocomposites prepared by melt mixing with several rather different types of polymers.

The rheological measurements showed that the incorporation of EG into the polymeric matrixes greatly changed some of their properties. In particular, the linear dynamic moduli and viscosities of the materials were considerably increased. In some polymers, the overall improvements in the rheological properties were more pronounced than in others.

There were significant changes in the frequency dependences of the moduli and viscosity at a certain critical concentration of EG called the rheological percolation threshold concentration. This concentration was associated with formation of a network-like structure arising from interactions

between the EG platelets and the polymers. However, the magnitudes of the effects of the EG were found to vary among the polymers. The effects were most pronounced in PC, followed by PP, and PS. In PE, almost no changes took place up to 15 wt% EG. Specific values of this percolation concentration were found to be at around 4 wt% in the case of PC/EG, 9 wt% for PP/EG, and PS/EG, and 12 wt% for PE/EG.

The differences in the critical compositions are most probably caused by differences in the exfoliation behavior, but this has to be investigated in more detail in morphological studies. Preliminary explanations for different degrees of exfoliation could be differences in the wetting behaviors and viscosities of the polymer melts. In particular, the wetting behavior depends on the surface tensions of the polymers and the resulting interfacial tensions between the molten polymers and the solid EG surfaces. Here, differences in the polarity of the polymer melts can play an important role. Effects from different entanglement densities probably are presumably significant as well.

Comparing different kinds of plots of the same data revealed that the the formation of network-like structures is very sensitively displayed using van Gurp-Palmen plots, which are most suitable for identifying “rheological percolation” in our investigated systems.

5 Acknowledgments

JEM wishes to acknowledge with gratitude the financial support he received from the National Science Foundation through Grant DMR-0314760 (Polymers Program, Division of Materials Research). He would also like to thank his colleagues at the Leibniz Institute of Polymer Research Dresden for their help and hospitality during a two-month visit there during the summer of 2004. Finally, the authors are pleased to acknowledge the help of Dr. K.-W. Stoeckelhuber (IPF Dresden) for his measurements of the surface energies of the EG samples employed in this investigation and the assistance of S. Pegel (IPF Dresden) in SEM studies.

6 References

- Tracz, A., Jeszka, J., Kucinska, I., Chapel, J.-P. and Boiteux, G. (2001) *Macromol. Symp.*, **169**, 129–135.
- Alexandre, M., Pluta, M., Dubois, P. and Jerome, R. (2001) *Macromol. Chem. Phys.*, **202**, 2239–2246.
- Causin, V., Marega, C., Marigo, A., Ferrara, G. and Ferraro, A. (2006) *Eur. Polym J.*, **42**, 3153–3161.
- Bissessur, R., Liu, P.K.Y., White, W. and Scully, S.F. (2006) *Langmuir*, **22**, 1729–1734.
- Stankovich, S., Piner, R.D., Chen, X., Wu, N., Nguyen, S.-B.T. and Ruoff, R.S. (2006) *J. Mats. Chem.*, **16**, 155–158.
- Palacios-Lidon, E., Perez-Garcia, B., Abellan, J., Miguel, C., Urbina, A. and Colchero, J. (2006) *Adv. Functional Mater.*, **16**, 1975–1984.

7. Fukushima, T. (2006) *Polymer Journal*, **38**, 743–756.
8. Yang, J., Tian, M., Jia, Q.-X., Zhang, L.-Q. and Li, X.-l. (2006) *J. Appl. Polym. Sci.*, **102**, 4007–4015.
9. Yang, Y., Grulke, E.A., Zhang, Z.G. and Wu, G. (2005) *J. Nanosci. Nanotech.*, **5**, 571–579.
10. Xian, G., Walter, R. and Hauptert, F. (2006) *J. Appl. Polym. Sci.*, **102**, 2391–2400.
11. Lu, J., Chen, X., Lu, W. and Chen, G. (2006) *Eur. Polym. J.*, **42**, 1015–1021.
12. Chen, G.-H., Wu, D.-J., Weng, W.-G., He, B. and Yan, W.-L. (2001) *Polym. Int.*, **50**, 980–985.
13. Chen, G.-H., Wu, D.-J., Weng, W.-G. and Yan, W.-L. (2001) *J. Appl. Polym. Sci.*, **82**, 2506–2513.
14. Shen, J.-W., Chen, X.-M. and Huang, W.-Y. (2003) *J. Appl. Polym. Sci.*, **88**, 1864–1869.
15. Chen, G., Weng, W., Wu, D. and Wu, C. (2003) *Eur. Polym. J.*, **39**, 2329–2335.
16. Weng, W., Chen, G., Wu, D., Chen, X., Lu, J. and Wang, P. (2004) *J. Polym. Sci., Part B: Polym. Phys.*, **42**, 2844–2856.
17. Zheng, W., Lu, X. and Wong, S.-C. (2004) *J. Appl. Polym. Sci.*, **91**, 2781–2788.
18. Shen, J.-W., Huang, W.-Y., Zuo, S.-W. and Hou, J. (2005) *J. Appl. Polym. Sci.*, **97**, 51–59.
19. Wu, G., Lin, J., Zheng, Q. and Zhang, M. (2006) *Polymer*, **47**, 2442–2447.
20. Lu, W., Lin, H., Wu, D. and Chen, G. (2006) *Polymer*, **47**, 4440–4444.
21. Wang, W.-P., Liu, Y., Li, X.-X. and You, Y.-Z. (2006) *J. Appl. Polym. Sci.*, **100**, 1427–1431.
22. Celik, C. and Warner, S.B. (2007) *J. Appl. Polym. Sci.*, **103**, 645–652.
23. Pötschke, P., Bhattacharyya, A.R., Janke, A. and Goering, H. (2005) *GAK*, **58-1**, 45–51.
24. Pötschke, P., Abdel-Goad, M., Alig, I., Dudkin, S. and Lellinger, D. (2004) *Polymer*, **45**, 8863–8870.
25. Costa, F.R., Abdel-Goad, M., Wagenknecht, U. and Heinrich, G. (2005) *Polymer*, **46**, 4447–4453.
26. Abdel-Goad, M. and Pötschke, P. (2005) *J. Nonnewt. Fluid Mech.*, **128**, 2–6.
27. Shenoy, A.V. *Rheology of Filled Polymer Systems*; Kluwer Academic Publishers: Dordrecht, Boston, London, 1999.
28. Utracki, L.A. *Rheology and Processing of Multiphase Systems. In Current Topics in Polymer Science, Vol. II. Rheology and Processing/Multiphase Systems*; Ottenbrite, R.M., Utracki, L.A. and Inoue, S. (eds); Carl Hanser: Munich, Vienna, New York, 149–165, 1987.
29. Zhang, Y., Chung, I.S., Huang, J., Matyjaszewski, K. and Pakula, T. (2005) *Macromol. Chem. Phys.*, **206**, 33–42.
30. Stoeckelhuber, K.W., Das, A., Jurk, R. and Heinrich, G. Publication in preparation.
31. <http://www.surface-tension.de/solid-surface-energy.htm>, assessed 22.12.06.
32. Harrell, E.R. and Nakayama, N. (1984) *J. Appl. Polym. Sci.*, **29**, 995–1010.
33. Nakayama, N. and Harrell, E.R. Modified cole-cole plot as a tool for rheological analysis of polymers. In *Current Topics in Polymer Science; Rheology and Polymer Processing/Multiphase Systems*; Vol. II, Ottenbrite, R.M., Utracki, L.A. and Inoue, S. (eds); Carl Hanser: Munich, Vienna, New York, 149–165, 1987.
34. Meincke, O., Kaempfer, D., Weickmann, H., Friedrich, C., Vathauer, M. and Warth, H. (2004) *Polymer*, **45(3)**, 739–48.