The role of reduced graphene oxide as a secondary filler in reducing the energy dissipation of silica-filled styrene-butadiene rubber composites under dynamic deformation

M. Nabavian Kalat[1,2], M. Razzaghi-Kashani[2], E. Pieczyska[1], M. Staszczak[1]

[1]*Institute of Fundamental Technological Research, Polish Academy of Sciences, Pawińskiego 5b, 02-106, Warsaw, Poland, email: mnabavian@ippt.pan.pl*

[2]*Polymer Engineering Department, Faculty of Chemical Engineering, Tarbiat Modares University, P.O. BOX: 14115-114, Tehran, I.R. Iran*

Abstract

Various quantities of reduced graphene-oxide (rGO) were added into silica-filled styrene-butadiene rubber (SBR) to make hybrid-filler composites. The non-linear viscoelastic behavior and corresponding energy dissipation of silica/rGO hybrid-filler and silica-filled composites were compared. Dynamic-mechanical analysis (DMA) of the hybrid-filler composites in strain-sweep showed that adding small quantity of rGO results in diminished silica network and reduced storage and loss modulus in silica-filled composites. Moreover, the synergy between silica and small quantities of rGO results in reduction of heat build-up (23%). The hybridization of silica/rGO was done without surface modification. This hybrid-filler system can be considered as an alternative to silane-modification of silica in industry.

1. Introduction

Silica is a common reinforcing filler using in the rubber compounds. But the high surface energy of unmodified silica particles (due to the presence of hydroxyl groups on the surface) leads to strong filler-filler interactions, poor dispersion of silica and formation of a filler network in the in the silica-filled rubber compounds. Breakage and reformation of the filler network under small-medium dynamic deformations result in non-linear viscoelastic behavior and corresponding energy dissipation [1]. In order to overcome this problem, "hybrid filler systems" have been widely used as a new approach to obtain homogeneous filler dispersion and lower energy dissipation resulted from dynamic deformation as well as superior mechanical properties [2-4]. In the present study, unmodified silica and rGO are used simultaneously as a hybrid filler system, without any surface modification and coupling agent, as other researchers have done. Different contents of rGO are simply added to the compounds containing constant volume fraction of un-modified silica and the hybrid filler compounds of silica/rGO are compared with the reference compound filled with silica (without any rGO), in order to detect any reduction of energy dissipation in hybrid filler compounds under the dynamic deformation.

2. Materials and characterization methods

The (rGO/silica)-SBR compounds were prepared in two steps. First, rGO-SBR compounds were prepared by the latex mixing method. Second, the silica-SBR compounds, with constant volume fraction of silica (14.66 vol.%), was prepared using an internal mixer (W50ETH, Brabender, Duisburg, Germany). Afterward, the prepared "rGO-SBR" and "silica-SBR" compounds were mixed on a two-roll mill (Brabender PM2000). Dynamic-mechanical properties of the compounds in strain sweep were measured using the MonTech D-RPA 3000. The heat build-up tests were performed on the vulcanized cylindrical shape samples with a diameter and height of 17.8 mm and 25 mm, respectively. Three specimens of each sample were tested under the constant load of 200 N and 30 Hz frequency at room temperature by Doli compression flexometer.

3. Dynamic-mechanical properties of silica/rGO-SBR compounds

Figure 1a displays the effect of adding different quantities of rGO to silica-filled compounds on the storage and loss modulus of compounds. In comparison to the silica-filled compound, the compound containing 0.25 phr (per hundred rubber) (0.09 vol.%) of rGO has the lowest initial storage modulus (G'). Decreased values of initial storage modulus is an indication of diminished filler network and improved filler dispersion in the presence of rGO platelets. Figure 1b represents loss modulus (G") and energy dissipated by the breakage and reformation of a filler network in a highly-filled compound under dynamic deformation. Peak of G" decreases in the presence of 0.25 (0.09 vol.%) and 0.5 phr (0.18 vol.%) of rGO, indicating weakened network of silica.

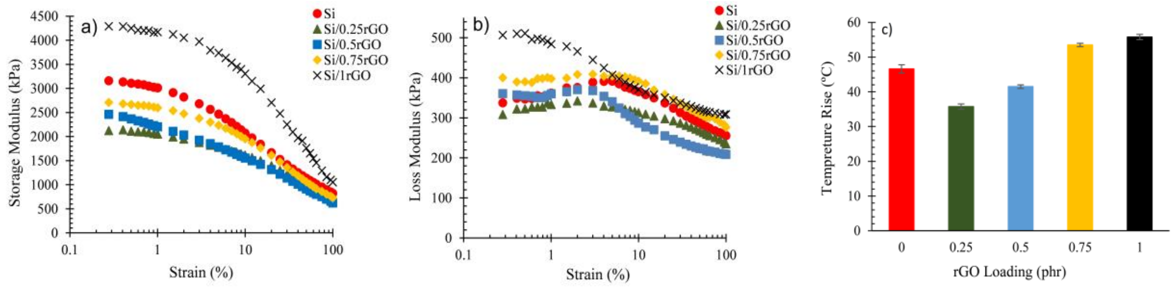

Fig. 1. a) Strain dependency of storage modulus, **b)** Strain dependency of loss modulus in the silica rGO-SBR compounds, **c)** Dependence of heat build-up on rGO content in silica-filled compounds

Temperature-rise in the heat build-up test is remarkably reduced in the presence of 0.25 and 0.5 phr rGO (Figure 1c). This is in line with the results of the storage and loss modulus showed in Figures 1. The presence of small content of large rGO platelets keeps the silica particle away from each other, hinders the reformation of silica network after mixing and reduces the friction between the silica particles, leading to decrease of energy dissipation.

4. Conclusion

Different contents of rGO platelets were incorporated into silica-filled compounds to investigate the energy dissipation of the silica/rGO-SBR hybrid filler compounds under dynamic deformation. By the addition of 0.25 phr rGO, heat built-up reduced 23%, which was in line with the reduction of storage and loss modulus of the compounds. Reduction of energy dissipation of compounds under dynamic deformation is a sign of better filler dispersion and less filler network extent.

Acknowledgments: The research was performed with support of the NCN - Grant 2017/27/B/ST8/03074.

References

[1] Sun, J.; Song, Y.; Zheng, Q.; Tan, H.; Yu, J.; Li, H. Nonlinear Rheological Behavior of Silica Filled Solution-Polymerized Styrene Butadiene Rubber. J. Polym. Sci. Part B Polym. Phys. **2007**, 45, 2594–2602, doi:10.1002/polb.21263.

[2] Feng, W.; Tang, Z.; Weng, P.; Guo, B. Correlation of filler networking with reinforcement and dynamic properties of SSBR/carbon black/silica composites. Rubber Chem. Technol. **2015**, 88, 676–689, doi:10.5254/rct.15.84881.

[3] Tang, Z.; Zhang, C.; Wei, Q.; Weng, P.; Guo, B. Remarkably improving performance of carbon black-filled rubber composites by incorporating MoS2 nanoplatelets. Compos. Sci. Technol. **2016**, 132, 93–100, doi:10.1016/j.compscitech.2016.07.001.

[4] Sattayanurak, S.; Sahakaro, K.; Kaewsakul, W.; Dierkes, W.K.; Reuvekamp, L.A.E.M.; Blume, A.; Noordermeer, J.W.M. Synergistic effect by high specific surface area carbon black as secondary filler in silica reinforced natural rubber tire tread compounds. Polym. Test. **2020**, 81, 106173, doi:10.1016/j.polymertesting.2019.106173.