Surface modification of reinforced silica by plasma polymerization Preparation and characterization

University of Twente
Enschede - The Netherlands

M. Tiwari, W. K. Dierkes, J. W. M. Noordermeer
University of Twente, Elastomer Technology and Engineering (ETE)
Enschede, The Netherlands
W. J. van Ooij
University of Cincinnati, Chemical and Materials Engineering
Cincinnati, USA

1. Objective

The objective of the work is to control the distribution of silica fillers in blends of incompatible elastomers by matching the surface energy of the silica and the polymers. The surface modification of silica fillers is achieved by plasma polymerization with different monomers.

Figure 1. Schematic of the tumbler RF plasma reactor

2. Introduction

The physical properties of a polymer blend, in general, are strongly influenced by the heterogeneity of the blend and the distribution of additives (especially, reinforcing fillers like carbon black and silica, curatives etc.) in each of the polymer phases [1]. With filled elastomer blends, controlled blend structure, i.e., controlled filler distribution in the individual rubber phases, is of primary importance [2]. Mismatches in polarity result in an unbalanced distribution and a bad dispersion of the fillers. Extensive transfer of silica from a polymer with low unsaturation (e.g. EPDM and IIR) to rubbers with high unsaturation (e.g. SBR, NBR) was found [3]. A good and stable dispersion and distribution is a prerequisite for a high degree of reinforcement and good properties of the material [4].

3. Experimental

For the surface modification of silica fillers, a radiofrequency (13.56 MHz) electrode-less tumbler plasma reactor consisting of a Pyrex cylinder chamber, with a motor-driven shaft in the centre and two vanes was used. The schematic picture of the reactor is shown in Figure 1. Plasma-polymerization is carried out after charging of dried silica powders into the tumbler reactor, vacuuming to 13 Pascal and then introducing a monomer for plasma polymerization. The plasma power is varied during the plasma polymerization of silica fillers. The monomer used for the experiments was acetylene (C_2H_2).

4. Results

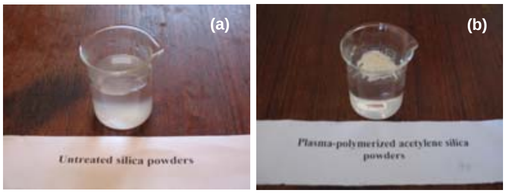

Figure 2. Immersion tests of (a) untreated and (b) acetylene plasma-polymerized silica fillers

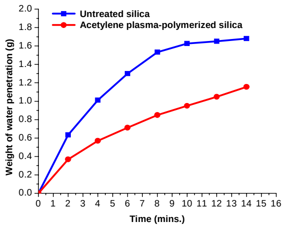

Figure 3. Water penetration into powder beds of untreated and acetylene plasma-polymerized silica fillers [5]

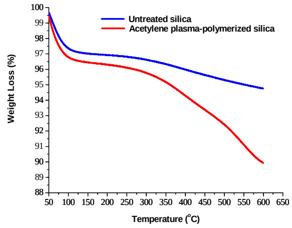

Figure 4. Thermo-gravimetric analysis (TGA) curves of untreated and acetylene plasma-polymerized silica fillers

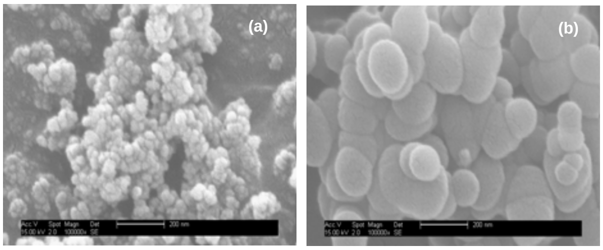

Figure 5. SEM images of (a) untreated and (b) acetylene plasma-polymerized silica fillers (a and b: 100,000X)

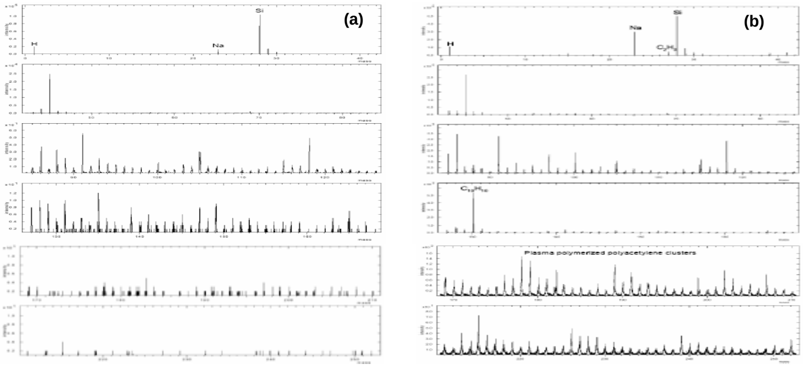

Figure 6. Positive TOF-SIMS spectra of (a) untreated and (b) acetylene plasma-polymerized silica fillers

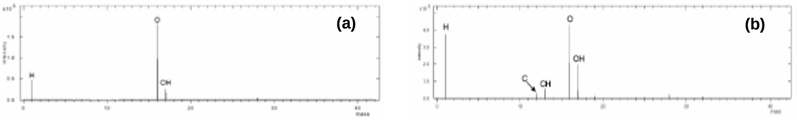

Figure 7. Negative TOF-SIMS spectra of (a) untreated and (b) acetylene plasma-polymerized silica fillers

5. Conclusions

The immersion tests and the water penetration measurements show a decrease in the surface energy of silica coated with polyacetylene film. TGA and TOF-SIMS measurements confirmed the film deposition on the surface of silica. SEM pictures show that the silica particles are covered with a polymer film, but basically still has the typical filler structure which is an important aspect for rubber reinforcement.

6. References

[1] J. E. Callan, W. M. Hess and C. E. Scott, Rubber Chem. Technol., 44, 814 (1971).
[2] M.-J. Wang, Rubber Chem.Technol., 71, 520 (1998).
[3] I. H. Jeon, H. Kim and S. G. Kim, Rubber Chem. Technol., 76, 1 (1971).
[4] B. L. Lee, Polym. Eng. Sci.,12, 25 (1985).
[5] N. Inagaki, S. Tasaka, and H. Abe, J. Appl. Polym. Sci., 46, 595 (1992).

7. Acknowledgement

The financial and technical support of **STW**, **HEXAGON POLYMERS** and **TIMCAL GRAPHITE and CARBON** is acknowledged.