EUROFILLERS 03

Alicante (SPAIN) – September 8-11th, 2003

CARBON BLACK: MICRO-DISPERSION

by

Dr. M. Gerspacher

Sid Richardson Carbon Company, Fort Worth, United States,

mgerspacher@sidrich.com

Introduction

One of the most important parameters of a given rubber compound reinforced with Carbon Black is the micro dispersion of the filler. This micro dispersion governs the fundamental viscoelastic response of the compound.

Far from solving all of the aspects of an important segment of tire technology, the present paper proposes to shed some light on the influence of the Carbon Black micro-dispersion on the compound properties.

The Rubber Compound

The main component of a pneumatic tire is the rubber compound. Composed of a mixture of an elastomeric matrix and rigid filler, its exact composition will depend on its role in the tire architecture.

In spite of all these variations, all rubber compounds can be described as two interpenetrating networks Figure 1 (TEM image) it clearly appears that some microvolume of polymeric networks are totally unaffected by the filler whereas the filler itself appears as a network of clusters of different sizes. One still remaining important questions is the presence or absence of polymeric chains in the filler cluster voids.

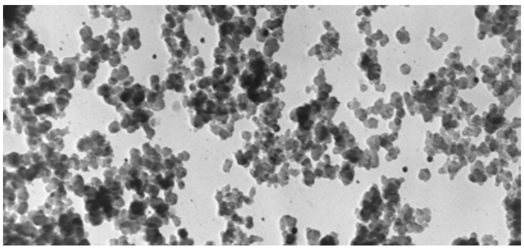

Figure 3: TEM image.

The local variation of filler cluster density may be considered as the "filler micro-dispersion" and it is being shown in this paper that this dispersion is by far the most important parameter influencing the physical behavior of the rubber compound.

It is also obvious that a lower local cluster density, a good micro-dispersion, will allow more filler surface to be in contact with the polymeric matrix, forming a less compact filler network.

It is also to be noted that the filler mono-unit, in this case an aggregate of carbon black, is a rigid object. The clusters or agglomerate, which make up the carbon black network, is what is being deformed upon strain energy input.

The polymeric network is composed of elastomeric chains characterized by a degree of mobility (depending on the elastomer structure, crosslink density, etc) and chemical unsaturation. These polymeric chains also undergo deformation upon strain energy input.

The filler-filler and the polymer-filler interactions are recognized as the important parameters of the physical properties of the final product. The micro-dispersion of the filler will govern the ratio of these two types of interactions.

Filler Micro-Dispersion

Carbon black is used in form of pellets. These pellets are obtained by mechanical densification of the original "fluffy" material and have an apparent specific gravity of 0.4g/cm3.

These pellets are normally introduced into the rubber mixer (i.e., banbury) once the polymer has been masticated. The "destruction" of the pellets is essentially obtained by shearing forces created by the mixer blades. It is possible to imagine that, once the pellet has been reduced to a much smaller size, the shearing forces are no longer effective for further size reduction. This may result in aggregates which are not distributed in the matrix of polymer since they are part of an original agglomerate.

It is most likely that at this stage of the process the interactions between the filler surface and the polymer chains has the largest influence. The more interactions, the more carbon black will be taken away from the original pellet and be distributed in the rubber matrix. This mechanism may explain the mode of disintegration of the pellets. [1] It is therefore appropriate to evaluate filler dispersion at a scale adapted to the aggregate size, the submicron length.

Several techniques exist [2] to approximate the degree of dispersion, but an absolute quantification of that dispersion of aggregates in 3-D space is still missing. A review of the possible methods [3] to assess dispersion highlight the following techniques:

Microscopy
Reflectometry
Mechanical Scattering Microscope
Electrical Measurements…

As indirect method, electrical resistivity measurement, have been used successfully to assess the degree of dispersion of carbon black in rubber. In particular it has been shown [4] that the electrical percolation threshold Ep is linked to dispersion. Ep being the smallest amount of carbon black required to attain percolation (Figure 2). The larger this Ep for a given carbon black, the better the dispersion.

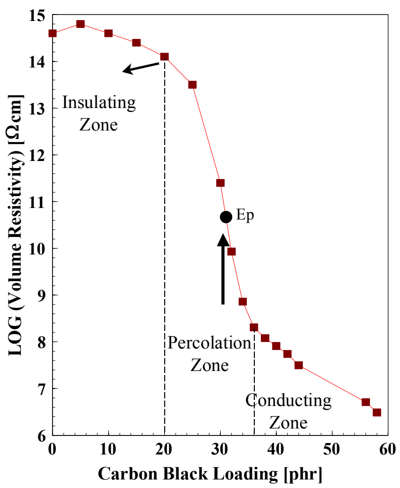

Figure 2: Percolation Point.

The result on Figure 3 indicates that this threshold is polymer dependent. The results show that Sn-SSBR disperses a given carbon black much better than NR for example. It is also remarkable to realize (Figure 4) that this percolation threshold is directly related to the polymer solubility parameter [5]. This solubility parameter δ being related to the unsaturation of the polymer chain it is reasonable therefore to relate the elastomer ability to disperse carbon black to the interactions between the carbon black surface and the polymeric unsaturations. In particular as shown in Table 1 the styrene content of the solution SBR is related to the percolation threshold Ep.

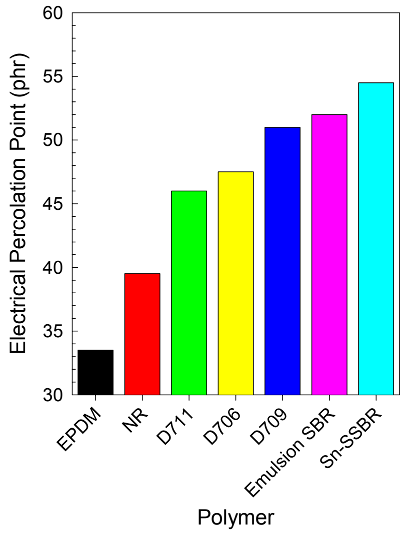

Figure 3: Percolation Point by Polymer.

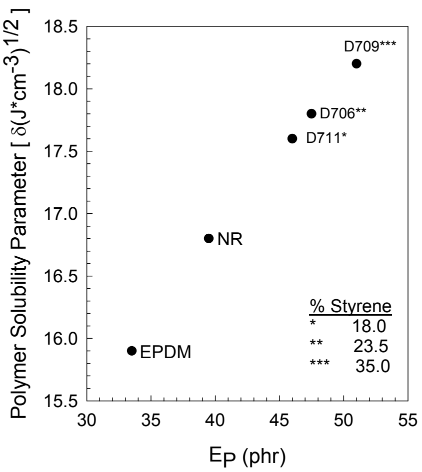

Figure 4: Percolation Point by Polymer Solubility.

Table 1.

% Styrene	EP (phr)	δ (Sol. Parameter)
18.0	46	17.6
23.5	47.5	17.8
35	51	18.2

Role of Carbon Black Dispersion

The Carbon Black dispersion governs the compound material interactions.

At low frequency and for strains below 50%, the filler-filler interactions play the major role. The effect of these interactions is to create a filler network which upon straining dissipates energy (friction) in the form of heat.

This may be beneficial if one wants to avoid any undo amount of stored elastic energy, which could be responsible for crack formation and propagation. But in today's world, where fossil fuel has to be spared, this energy dissipation can be viewed as a negative (i.e. high hysteresis).

This negative can be addressed by a less dense filler network, which may be achieved by a better dispersion of the filler (Figure 5).

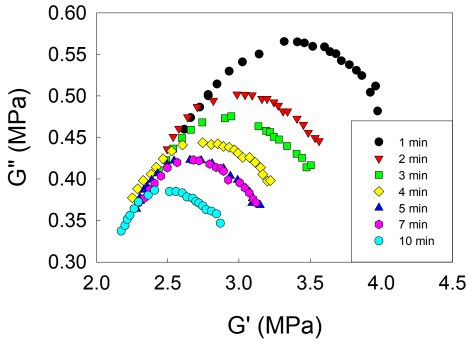

Figure 5: Effect of Mixing Time on Low Strain Dynamic Properties.

At high frequency, characteristic of the road/tire tread interface where the deformation is very minute, the reduced mobility of the polymeric chains, due to their interaction with the carbon black, is responsible for energy dissipation required for good traction (Figure 6).

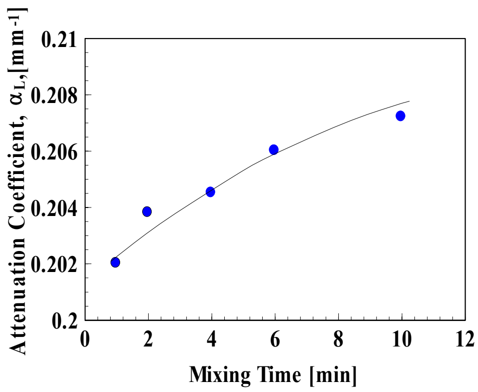

Figure 6: Effect of Mixing Time on High Frequency Viscoelasity.

In a previous study [6] it was also shown that a tread compound exhibiting a better micro-dispersion (Ra) has improved wear performance (Table 2 and Figure 7).

Table 2. Tread Wear.

Wear Rating	Ra
100	0.42
109	0.35
117	0.26
110	0.21
111	0.30
109	0.31

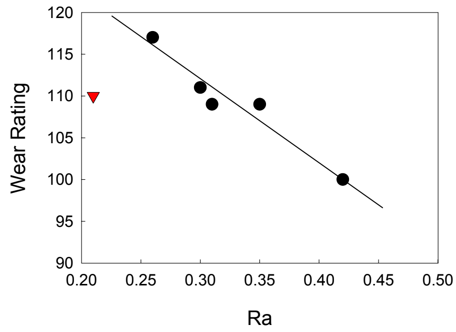

Figure 7: Tread Wear

It appears therefore that a better filler dispersion will be beneficial for these fundamental characteristics of a tire: Rolling Resistance Traction and Wear.

It is interesting to note that in order to achieve a better dispersion during mixing, the interactions between the polymer and the fillers may play an important role besides the mechanical aspect of mixing.

Conclusions

Considering a modern tire compound, it has been shown that the materials interactions play a very significant role. This paper also indicates that a good dispersion may well solve the problem of the so-called magic triangle. Indeed a good dispersion improves all fundamental tire properties.

To achieve a good dispersion obviously both the carbon black and the polymer have to be fine-tuned. This is certainly one of the key objectives of the producers of these materials.

References

1. Qi Li, D. L. Feke, and I. Manas-Zloczower, *Rubber Chem. Technol.* **1995**, 68(5), 836-841.
2. M. Gerspacher, L. Nikiel, H. Yang, and C. P. O'Farrell, *Rubber Chem. Technol.* **1998**, 71(1), 17-25.
3. L. Nikiel, M. Gerspacher, H. Yang, and C. P. O'Farrell, *Rubber Chem. Technol.* **2001**, 74(4), 249-259.
4. M. Gerspacher, C. P. O'Farrell, and L. Nikiel, ACS Rubber Division Paper 155, Orlando **1999**.
5. R. Schuster – Private Communication.
6. M. Gerspacher and C. P. O'Farrell, *Rubber & Plastic News.* **1993**, 23(6), 85-87.