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International Journal of Polymeric Materials

Publication details, including instructions for authors and subscription information: http://www.informaworld.com/smpp/title~content=t713647664

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To cite this Article Anely, J. N. and Khananashvili, L. M.(1995) 'Effect of Ultrasound on Conductivity of Polymeric Composites', International Journal of Polymeric Materials, 28: 1, 99 – 102 To link to this Article: DOI: 10.1080/00914039508012092 URL: http://dx.doi.org/10.1080/00914039508012092

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Effect of Ultrasound on Conductivity of Polymeric Composites

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(Received July 15, 1994)

The mechanism of the influence of ultrasound (US) field on conductive properties of filled polymeric materials based on poly(dimethylmethylsiloxan) and polypropylene was investigated at room temperature and above up to 80°C. It has been shown, that the effect of US on the conductivity of composites essentially depends on such factors as density of crosslinked bonds, concentration and type of fillers, degree of crystallinity and interaction between polymer and filler. It is found that composites conductivity decreases as US power increases because of the increase in intensivity of Brownian motion of filler particles. This motion is connected with segmental movement of macromolecules which essentially depends on the character of macromolecular microstructure and the value of interaction between composites components.

KEY WORDS Ultrasound, effect, polymeric composites, Brownian motion, interaction, fillers concentration, crystallinity, conductivity, current-voltage characteristic.

The conductivity properties of polymeric materials, including some conductive fillers, depend essentially on a number of physical factors (for example mechanical deformations, temperature *et al.*).^{1,2} The explanation of experimental data lies generally in the character of distribution of conductive filler particles in the polymeric matrix. On the other hand, it is known that the pass of ultrasound through the substance is connected to the moving of their particles near equilibrium points.³

The main purpose was to define the effect of ultrasound on conductive properties of composites based on different polymers:insulators and conductive disperse fillers.

The experiments were conducted using two types of polymer composites—rubbers and plastics—filled by carbon materials. Polymer silicon elastomer polydimethylvinilsiloxane SKTV and polypropilen were used as fillers—highconductive carbon black P357-E and lowconductive carbon black P803.

Rubber samples were obtained by peroxide vulcanization and by cold vulcanization (at room temperature). Peroxide of dicumile and diethylaminomethyltree-toxysilan as curing agent were used in this work.⁴ The samples sizes $40 \times 10 \times 1$ mm equipped with electrodes according 4 electrodes method⁵ were irradiated in the twofold distilled water by US waves from source with magnetostriction radiator at fixed frequency (30 cs) with regulated power of the radiation. The experiments were carried out mainly at room temperature. In separate cases, temperatures up to 80°C were used.

The influence of US on the conductive properties of polymeric composites expressed by their current-voltage characteristic (CVC) was studied. US do not damage linearity of CVC, but the slope of corresponding lines relative to voltage axis increase when the irradiated power increases (Figure 1). At this time, the effect of US field, expressed by decrease of conductivity, illuminated almost immediately. An increase of environment temperature generates a weakening of US field effect on the conductivity value (Figure 2). The experiments show that the effect of US on the conductivity values γ (calculated by CVC) depends essentially on structural factors, 1) density of crossed macromolecular bonds (partial, on density of vulcanization set in the rubbers), 2) concentration and dispersity of fillers, 3) degree of crystallinity of polymeric matrix and 4) the character of the interaction between polymer and filler (Figure 2).

Let's discuss the experimental results more in detail.

1. As Figure 1 illustrates, using CVC for rubber based on elastomer SKTV and carbon black P803, the conductivity of the composite decrease as US power increases (the slope of dependence CVC at this time increases). This may reflect an increase in the intensity of Brown movement of filler particles. According to References 1 and 2, the variation of the conductive particles configuration affect the conductivity of composite. In addition, US field as a factor tool to investigate a conductive system, leads to the increase of volume resistivity of the material. The CVC for all samples in used voltage interval shows reversibility, which indicates a full regeneration of initial filler configuration in the polymeric matrix. The results obtained with other samples agree qualitatively with the reported trends, but may differ quantitatively as a consequence of their specific structure.

2. CVC obtained at above room temperature are characterized by decrease of



FIGURE 1 Current-voltage characteristics for rubber based on SKTV and P803 (80 mass.part) obtained at US power: 0(1), 2(2), 3(3) watt at room temperature.



FIGURE 2 Effect of temperature (a), density of vulcanization network (b), degrees of filling (c), interaction (d) and crystallinity (e) of composites on dependence of relative special volume conductivity on US power for rubbers based on SKTV and P803 at consisting filler 80 (1235), 120 (4) mass.part, P357-E at consisting of latter 60 mass.part (6) and for plastics based on polypropilen filled by P357-E (60 mass.part) with degree of crystallinity 37(7) and 56(8)%. The lines 1, 3, 4 correspond to rubbers, vulcanized by use cure agent diethylaminomethyltrietoxisilan, lines 2, 5, 6—to rubbers vulcanized by use dicumil peroxide as a cure agent.

conductivity, which is associated with an increase of Brown movement of conductive filler particles and with the damage of conductive system. On simultaneous exposure of US and heating on conductivity of composites the field effect weakens at elevated temperatures because these two treatments have similar effects on conductivity (Figure 2a).

3. It is shown, that the effect of microstructure of polymeric matrix on conductivity properties of rubbers, vulcanized by two methods (cold vulcanization and peroxide vulcanization) is clearly discernible in the response of these systems to US field (Figure 2b). At could vulcanization the crosslinking of SKTV molecules proceeds only on the end groups, while the peroxide vulcanization—involves the participation of unsaturated functional groups. These differences in the nature of crosslinking affects many properties of vulcanizates. So, the macromolecules linked by end groups are characterized by low density of vulcanization network. These systems have the higher elasticity and mobility, than the vulcanizates with high density of cross-link density. The fillers particles placed in more flexible vulcanizates are therefore, more mobile, than those in highly crosslinked rubbers. This is the reason for the observed dependence of conductivity of rubbers on the density of vulcanization network.

4. Important factor, which also effects the mobility of macromolecules is the concentration of filler. It is known,⁶ that an increase of dispersed particles content in composite reduces the segmental movement through an increase of matrix density. This leads to a decrease of mobility of filler particles and consequently to the weakening of US effect on conductivity of the material (Figure 2c).

5. The behavior of composites, containing crystal phases in the molecular system, resembles that of samples having a high density polymeric matrix. In polymers with definite well developed crystallinity the fillers particles are excluded from the crystalline phase, which leads to an increase of fillers concentration in the amorphous phase. Therefore, an increase of conductivity and density is observed,⁷ the US effect on conductivity, however, decreases with increases in crystallinity and density of material (Figure 2d).

6. According to Figure 2e, an essential contribution to the US effect is the extent of polymer-filler interaction. This interaction is due to adsorption and has a physical as well as a chemical nature.8 For example, the particles of carbon blacks P357-E are characterized by strong interaction with the elastomer molecules because of their surface characteristics which contains many active chemical groups. These interactions lead to a decrease of segmental mobility of macromolecules. The rubber, filled with carbon black P803 exhibits weak interaction with macromolecules. This leads to high mobility of fillers particles and more strong effect of US.

The obtained results allow us to make one general conclusion: the main factor in the effect of US field on conductivity of polymeric composites is the intensivity of Brown movement of fillers particles. The Brown movement is however controlled by segmental mobility of macromolecules, which reflects the nature of microstructure of macromolecules and the value of interaction between the components of the composite system.

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