Growth of Microvoids in Fluorine Elastomer Composites during Stretching

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1 Introduction

The fluorine elastomers are used for coatings, O-rings and valves due to their excellent thermal and chemical resistance. However, their high chargeability causes dust deposit and their mechanical strength is low. For modifying their properties, attempts have been made to disperse conductive and high strength fillers into fluorine elastomers. This study compares the influences of carbon nanotubes (CNTs) and graphene oxide nanoribbons (GONRs) on the mechanical properties and the growth of microvoids during stretching for the fluorine elastomer composites containing these fillers. [1]

2 Experimental

A copolymer of tetrafluoroethylene with perfluorovinyl ether (FFKM, Daikin Industries, Ltd.) was used for the matrix elastomer. Multi-walled carbon nanotubes (MWCNTs, Showa Denko K.K., VGCF-H), with diameters of 150 nm and lengths of 10–20 μ m, were used for the fillers and the parent MWCNTs used for producing the GONRs via the unzipping method. [2]

The FFKM was swollen with a solvent, added with the CNTs or GONRs and mixed using a planetary centrifugal mixer. After the solvent was evaporated, the mixture was pressed into films 500 μ m thick using a hot press. During the hot pressing, Si₃N₄ and urea were placed around the films for producing crosslinking by the formation of triazine rings.

The small-angle X-ray scattering (SAXS) measurements during stretching of the elastomer composites were performed at BL-6A in PF. The X-ray wavelength was 0.15 nm, the sample-to-detector distance

was 2 m and the scattering intensity was measured with Pilatus 3. The tensile tests and the wear tests were performed on the elastomer composites.

3 Results and Discussion

Fig.1 shows the tensile modulus, the tensile strength, the tensile strain at break and the wear resistance for the elastomer composites relative to these values for the elastomer without containing fillers where the wear resistance is defined as the inverse of the wear rate. The elastomer without containing fillers had the tensile modulus of 1.1 MPa, the tensile strength of 3.3 MPa, the tensile strain at break of 2.1 and the wear rates of 7.6×10^{-11} Pa⁻¹. As compared with CNTs, GONRs more effectively increase the tensile strength and the wear resistance without causing larger increase in the tensile modulus.

In Fig.2, the integrated intensity of SAXS is plotted against the tensile strain for the elastomer composites. The integration was carried out by weighting the intensity at individual pixels with $2\pi |s|$ where *s* is the separation, measured perpendicularly to the stretching direction, between each pixel and the center beam position. This integrated intensity is proportional to the total volume of the microvoids formed at the filler-matrix interfaces. This figure shows that the growth of microvoids during stretching is markedly suppressed by dispersing GONRs in place of CNTs. This result can be attributed to the larger surface area and the larger amount of functional groups for the GONRs as compared with the CNTs.

In some application fields of fluorine elastomers, it is desired to increase mechanical strength without



Fig. 1: Comparison of tensile and wear properties of fluorine elastomer composites containing 1 wt% of CNTs and GONRs shown by relative values.



Fig. 2: Integrated intensity of SAXS versus tensile strain for fluorine elastomer composites containing CNTs and GONRs by the weight fractions shown in the figure.

increasing the tensile modulus and the GONR-dispersion is more suitable than the CNT-dispersion for such applications.

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References

K. Kubo, S. Maruta, M. Shioya, T. Noguchi, *Fiber preprints, Japan*, **72**, No. 1, 1H09 (2017).
D. V. Kosynkin, et al., *Nature*, 458, 872 (2009).

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