

## Free volume in strained rubber with carbon black filler by positron annihilation spectroscopy

M. Nippa<sup>1,\*</sup>, Y. Ikeda<sup>2</sup>, A. Kato<sup>3</sup>, A. Komatsu<sup>1</sup>, L. Chiari<sup>1</sup> and M. Fujinami<sup>1</sup>

<sup>1</sup>Department of Applied Chemistry, Chiba University, 1-33 Inage, Yayoi 263-8522, Japan

<sup>2</sup>Kyoto Institute of Technology, Matsugasaki, Kyoto 606-8585, Japan

<sup>3</sup>NISSAN ARC, LTD., Natsushima 1, Yokosuka, Kanagawa 237-0061, Japan

\*email: madoka.nippa@chiba-u.jp

The main component of tires is natural rubber, and various additives are added to increase the strength of the rubber. Carbon black (CB), which is dispersed in the rubber, is known to remarkably improve its mechanical properties. This reinforcing effect is thought to be related to the CB network, in which CB particles are connected to each other within a 3-nm thick layer, the so-called bound rubber, at the CB/rubber interface [1]. However, the detailed reinforcement mechanism is still unknown. In this study, we aimed to investigate the mobility of molecular chains in rubber by measuring the change in free volume within strained rubber with various amounts of carbon black filler by positron annihilation spectroscopy.

The rubber consisted of isoprene rubber (IR) filled with various amount of CB (N330). The rubber specimens were prepared by mixing IR, CB and 1 phr (phr: parts by weight per 100 parts by weight of rubber) DCP (dicumyl peroxide) as the crosslinking agent. The CB content was varied from 0 to 80 phr. The rubber samples were strained to a ratio of 160% with a tensile tester and several consecutive positron lifetime measurements were carried out in situ for about 100 hours in total. After releasing the samples, they were measured once again.

The o-Ps lifetime dependence on the CB content is shown in Fig.1. The o-Ps lifetime showed a minimum at 40 phr and generally no large changes were observed. The results shown in Fig. 1 were very similar to the dependence of the branch chain fraction of the CB network structure (number of branched chains / (number of branched chains + number of crosslinked chains))\_on the CB content [1]. Afterwards, the o-Ps lifetime of the strained samples measured in situ. The o-Ps lifetime of the rubber samples hardly changed with and without strain. On the other hand, a decrease in the intensity of the o-Ps component was observed in the strained samples and the maximum decrease of 3 % was measured in the sample containing 40 phr CB. After releasing the sample, the intensity recovered. It is believed that the molecular chains in the amorphous phase become aligned when subject to tensile stress, thus leading to a decrease in Ps formation.

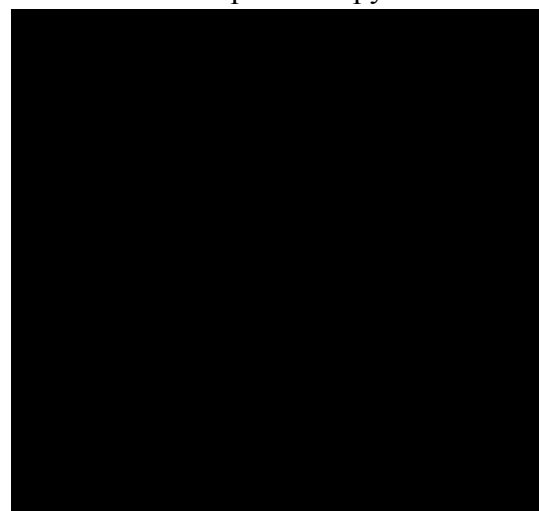


Fig.1. o-Ps lifetime dependence of CB content

[1] A. Kato *et al.*, *Japan Rubber Association Magazine* 87, 447 (2014).