An Intelligent Nano-Composite: Strain-Induced Crystallization of Natural Rubber

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Natural rubber (NR) has been know to be able to crystallize upon elongation. Though studies on crystallization behaviors of NR appeared at the early stage of the advent of polymer science, the details of crystallization mechanism are still to be elucidated. X-ray radiation from synchrotron sources is powerful enough to conduct in situ measurements of wide-angle X-ray diffraction (WAXD) of NR under streching. NR vulcanizates, which were completely amorphous (rubbery state), gave crystalline reflections upon streching and returned to the original amorphous state by retraction. Time resolved measurements of this crystallization-melting cycles by streching-retraction were carried out extensively. The analyses of the experimental results indicate that (1) the crystallization-melting cycle was apparently reversible, but crystallization index vs. strain ratio polts showed hysteresis. (2)The onset of crystallization was at fairly large elongations, e.g. at strain ratio of 3.5, and the onset strain did not depend on network chain density (or degree of crosslinking) of the vulcanizate. (3) The melting of the formed crystallites was delayed when compared with the crystallization, which may be one of the reasons of the hysteresis. (4)The highest crystallinity index was at most 40 %, and the oriented amorphous fraction was less than 10 %. Therefore, majority of network chains remain amorphous even as high as strain ration of 7 or 8. (5) The crystallite formed was of nm size, and the lattice constants were changed during the elongation and retraction. These results suggest that NR self-forms many reinforcing nano-meter sized 'filler' (crystallite) upon elongation (when it is necessary to be strong under strains), and they automatically disappear when the strain is decreased. In other works, NR is intellignet enough to become tough when it is necessary, and relaxed when under no stress.