

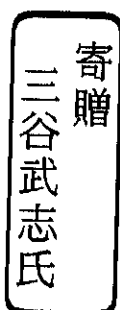
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Photo-Induced Structural Transformation of C<sub>60</sub> Single  
Crystals and C<sub>60</sub> Compounds under High Pressure

Doctoral Program in Engineering  
University of Tsukuba

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## Abstract

Photo-induced structural transformation of  $C_{60}$  single crystal and its compounds has been investigated under high pressure up to around 30 GPa by means of optical measurements. Formation of three-dimensional  $C_{60}$  network structure was achieved by the photo-irradiation to  $C_{60}$  single crystals under high pressure. Iodine doping worked as a structural template of the photo-dimerization because  $C_{60}$  dimers were selectively fabricated irrespective of magnitude of the pressure applied during photo-irradiation. Alkali metal doping completely suppressed the structural change of  $C_{60}$  molecules.

Electronic and vibrational properties of  $C_{60}$  products which were photo-irradiated under pressure showed that structural transformation of  $C_{60}$  single crystals significantly depended on the pressure applied during the photo-irradiation. The dependences can be interpreted as imprints of contracted structures under pressure due to formation of intermolecular bonds whose number is continuously increased with the applied pressure. The number of intermolecular bond has been estimated by comparative analyses using high pressure optical properties of the  $C_{60}$  photopolymer which was photo-irradiated at ambient pressure. The analyses reveal that the number of the intermolecular bond increases up to about 5 with the applied pressure to 25 GPa. The activation energy of the photo-transformation at 3 GPa has been estimated to be 0.23 eV which is equivalent to the activation energy of the photo-polymerization at ambient pressure. Therefore, the interlinking mechanism can be considered to be a [2+2] cycloaddition scheme similar to the photo-polymerization at ambient pressure. The three-dimensional structure of the polymerized phase originates from the inherent randomness of the photo-polymerization of  $C_{60}$  molecules.

Iodine doping effects on the photo-induced structural transformation have been investigated by Raman spectroscopy for iodine doped  $C_{60}$  compounds crystallized into FCC and hexagonal structures. The photo-irradiation to the two samples yielded only  $C_{60}$  dimers irrespective of the applied pressure. The decomposition of the polyiodine molecules that exist in  $C_{60}$  lattice has close relationship with the dimerization of  $C_{60}$  molecules. Alkali metal doping effects have also been investigated with  $A_3C_{60}$  and  $A_6C_{60}$  compounds, where A denotes potassium and rubidium. In spite of some hopeful precedents, structural transformation of  $C_{60}$  was completely suppressed by alkali metal doping even though the  $A_xC_{60}$  was subjected to the photo-irradiation at high pressures and high temperatures.

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