A post-synthetic treatment combining applied current with heating to improve the properties of carbon nanotubes and exfoliated graphene

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Many researchers have reported that the healing of crystalline defects in materials improves various properties. For example, the approach to improving the strength by defect healing of carbon fibers has been studied for 50 years. A defect-free carbon nanotube (CNT) and graphene are expected to possess exceptional thermal and electrical properties. However, in reality, crystalline defects are present in CNTs (single-walled carbon nanotubes; SWCNTs), graphene (exfoliated graphene), which decreases the thermal and electrical conductivities and dispersibility [1]. If the crystallinity could be improved (i.e. heal crystalline defects), CNTs and graphene would be available with excellent quality to spur new applications. While high temperature treatment of SWCNTs and in post-synthetic processes, which is considered as an approach to solve this issue, has demonstrated increase in crystallinity, and electrical and thermal properties, unexpected effects have been observed, such as the coalescence of SWCNTs into double-walled CNTs [2].

In this study, to reduce defects and improve thermal and electrical properties of SWCNTs to retain the single-walled structure, we developed an original post-synthetic process (*heat and current process*) which conbined heating treatment and electrical current flow process. At optimum conditions (800 °C, 150 A cm^{-2} (1150 W cm^{-2}) for 1 min in Ar ambient), we achieved a 3.2-fold increase in the Raman G- to D-band ratio (crystallinity), a 3.1-fold increase in electrical conductivity, and a 3.7-fold increase in thermal conductivity. The electrical and thermal conductivities increase simultaneously with the increase in G/D ratio, therefore these properties reflect the improvement in the crystallinity. We clarified that the electrical process contributed to maintain their structure as SWCNTs, achieve the higher property improvement than high temprature process, and decrease process temperatures and process times.

In addition, we adapted this process for 2D-materials with passing in-plane current for graphene sheets and heating. We achieved this by applying the electrical current (current density: ~900 A cm^{-2}) in-plane of an exfoliated graphene sheet with heating (~900 °C) in an argon ambient. At 900 °C, 545 A cm^{-2} for 1 min, we achieved ~30-times increase in the Raman G- to D-band ratio (2.84 to 85.3), and ~2.0-times increase in electrical conductivity (from 1088 to 2071 S cm^{-1}). We also confirmed by using X-ray diffraction (XRD) that the electrical conductivities increase without structure changes (interlayer distance) of exfoliated graphene. In contrast, a decrease in interlayer structure of exfoliated graphene was observed when current was not applied at high temperature (1500 °C in argon ambient above 1 hours), while the electrical conductivity increased up to 1750 S cm^{-1}. These results demonstrate the importance of applying current to improve the crystallinity (defect healing) of exfoliated graphene in short treatment time with maintaining their structure.

References:

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