Highly Concentrated and Conductive Reduced Graphene Oxide Nanosheets by Monovalent Cation-pi interaction: Toward Printed Electronics

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We introduce a novel route to preparing highly concentrated and conductive reduced graphene oxide (RGO) in various solvents by monovalent cation-pi interaction. Previously, the hydrophobic properties of high-quality RGO containing few defects and oxygen moieties have precluded the formation of stable dispersion in various solvents.[1,2] Cation-pi interaction between monovalent cations, such as Na^+ or K^+, and six-membered sp^2 carbons on graphene were achieved by simple aging process of graphene oxide (GO) nanosheets dispersed in NaOH or KOH solution. The noncovalent binding forces introduced by the cation-pi interactions were evident from the chemical shift of the sp^2 peak in the solid ^{13}C NMR spectra. Raman spectra and the I-V characteristics also demonstrated the interactions in terms of the presence of *n*-type doping effect due to the adsorption of cations with high electron mobility (39 cm^2/Vs) as shown in Figure 1. The RGO film prepared without a post-annealing process displayed superior electrical conductivity of 97,500 S/m at a thickness of 1.7 um. Moreover, mass production of GO paste with a concentration as high as 20 g/L was achieved by accelerating the cation-pi interactions with densification process. Our strategy can facilitate the development of large scalable production methods for preparing printed electronics made from high-quality RGO nanosheets.

Mass production of an easily-dispersible cation-pi interacting graphene oxide (CIGO) powder was achieved by improving the effective interactions of alkali ions with a pi system as shown in Figure 2. The cation-pi interactions were accelerated by boiling a GO dispersion in KOH, which increased the number of sp^2 carbons upon mild deoxygenation. Subsequently, the CIGO solution was concentrated by rotary evaporation, then centrifuged three times to remove residual alkali metal ions. The viscosity provided a physical indication of the quality and dispersibility of the paste. The CIGO paste revealed a high viscosity of 10^5 mPas at a 10^{-2} sec^{-1} shear rate under ambient conditions, which is comparable to the viscosity of conventional silver paste.[3] In contrast, condensation of the non-interacting GO was prevented by aggregation. These results demonstrate that monovalent cation-pi interactions are crucial for stabilizing a dispersion of highly concentrated GO and RGO with high quality.

References

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[3] S. P. Wu, L. Q. Zheng, Q. Y. Zhao, X. H. Ding, Colloids and Surfaces A: Physicochem. Eng. Aspects **372** (2010) 120.
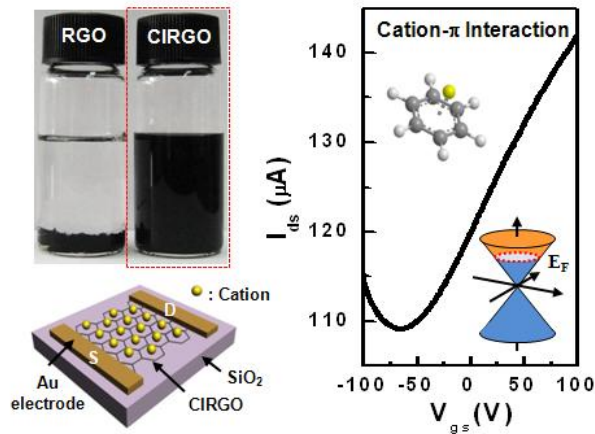
Figures

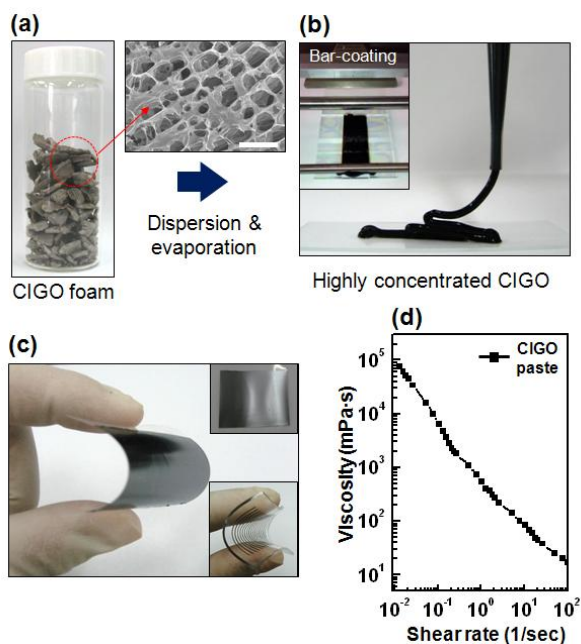## Figure 1

Figure 2

Figure caption

Figure 1

Highly concentrated and conductive RGO nanosheets were prepared by introducing monovalent cation-pi interactions between Na^+ or K^+ ions and six-membered sp^2 carbons on graphene. I-V characteristics demonstrate the interactions in terms of the presence of *n*-type doping effect due to the adsorption of cations with high electron mobility (39 cm^2/Vs).

Figure 2

Mass production of highly concentrated CIGO and its application to printed electronics. a) Production of foam-like CIGO structures after a freeze-drying process (red dashed arrow: SEM image of the CIGO, the scale bar indicates 100 um). b) highly concentrated CIGO paste, prepared by additional condensation processes, to yield a high viscosity (inset: CIGO film formation using bar-coating method). c) Bar-coated CIRGO on a PET substrate (inset: uniform coating on PET and a patterned CIRGO on PDMS substrate). d) Viscosity measurements of the highly concentrated CIGO paste.