

Novel graphene based hybrid material with tunable electronic properties

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Chemical functionalization of graphene (or a few-layered graphene) is an advanced approach for creation of complementary materials with wide spectrum of electronic properties. This method is used for a band gap opening and a managing with carrier concentration, type and mobility. The band gap has a central role in modern device physics and technology and governs the operation of semiconductor devices. A tunable band gap would be highly desirable because it would allow great flexibility in design and devices optimization. In present study we have used intercalation of N-methylpyrrolidone (NMP, C₅H₉NO) into a few-layered graphene with the aim to create a new graphene-based material with variable properties in controllable manner. Such approach as intercalation provides a pure and two-side functionalization of graphene. A choose of intercalation agent is based on well-known property of the NMP to penetrate between every graphene layers. As a result, we have created a new hybrid material with variable properties.

Fabrication process of our hybrid structures includes the follows steps: (1) an electrostatic exfoliation of the few-layered graphene film with thickness about $d \sim 2-5$ nm and transfer it on the 300 nm SiO₂/Si substrate; (2) an intercalation of the NMP into graphene flakes; (3) annealing of the intercalated structures at temperature in the range of 125–250°C was the final operation which creates the hybrid structures.

Tunable electronic properties with strong and non-monotonic dependence on the fabrication temperature were revealed for this hybrid material. Opening of the band gap, wide variation of resistivity (up to 10⁷ times), relatively high mobility and sp² or sp³ hybridization of carbon atoms are attributed to our hybrid material. Hybrid structures created in temperature ranges of $T < 200^\circ\text{C}$ and $T > 200^\circ\text{C}$ were found to demonstrate different properties. The first type of hybrid structures created at the temperatures lower 200°C have high resistivity ($\sim 10^2-10^6$ Ohm.cm), p-type conductivity, a strong temperature dependence of resistivity in the range of 77–300 K, and the band gap $\sim 3 - 3.5$ eV (for structures created at temperature of 150°C). We have presumed for these structures that bonds between NMP and graphene are formed through oxygen atoms. The second type of hybrid structures corresponded to $T > 200^\circ\text{C}$ is distinguished by appearance of the sp³ hybridization of carbon atoms typically observed in the case of graphane formation. In our case this process can be caused by NMP polycondensation with releasing of hydrogen atoms. The band gap ($\sim 3.5 - 4.0$ eV) is revealed for structures created at 250°C.