Carbon Nanotubes Meet Graphene Oxide: Efficient Removal of Chemisorbed water for enhanced electrode materials

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Self-assembled graphene oxide (GO) papers are of increased interest for filtration, sensing and energy storage applications [1-3]. The performance of the paper assemblies, especially upon thermal reduction treatments, is seriously affected by the presence of chemisorbed water, i.e. water molecules inherently bound to the oxygen functional groups (OFGs) of GO [4-5]. In this work we show that oxidized multiwall carbon nanotubes (oCNTs) self-assembled onto GO sheets by an in-situ exfoliation process effectively can remove chemisorbed water and significantly reduce associated damaging effects. This is based on the formation of cooperatively strengthened OH···O=C hydrogen bonds between the carboxylic groups of oCNTs and OFGs on GO. The corresponding GO/CNT hybrid papers reveal enhanced sp^2 character, improved conductivity, and electrochemical capacitance up to 150 F/g with respect to RGO [6]. These findings are of relevance for the development of improved electrochemical devices, such as supercapacitors and (bio-)sensors.

1. R.R. Nair, H.A. Wu, P.N. Jayaram, I.V. Grigorieva, A.K. Geim, *Science* 6067 (2012), 442.
2. J. Zhu, C.M. Andres, J. Zu, A. Ramamoorthy, T. Tsotsis, N.A. Kotov, *ACS Nano* 6 (2012), 8357.
3. L.L. Zhang, X. Zhao, M.D. Stoller, Y. Zhu, H. Ji, S. Murali, Y. Wu, S. Perales, B. Clevenger, R.S. Ruoff, *Nano Letters* 12 (2012), 1806
4. C. Vallés, J.D. Núñez, A.M. Benito, W.K. Maser, Carbon 50 (2012), 835
5. M. Acik, C. Mattevi, C. Gon, G. Lee, K. Cho, M. Chhowalla, Y.J. Chabal, *ACS Nano* 4 (2010), 5861
6. J.D. Núñez, A.M. Benito, S. Rouzière, P. Launois, R. Arenal, P.M. Ajayan W.K. Maser, Chem. Mater., submitted.

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