## Accepted Manuscript

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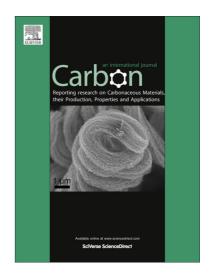
PII: S0008-6223(15)00342-5

DOI: http://dx.doi.org/10.1016/j.carbon.2015.04.051

Reference: CARBON 9869

To appear in: Carbon

Received Date: 20 January 2015 Accepted Date: 20 April 2015



Please cite this article as: Bi, H., Cui, H., Lin, T., Huang, F., Graphene Wrapped Copper-Nickel Nanospheres on Highly Conductive Graphene Film for Use as Counter Electrodes of Dye-sensitized Solar Cells, *Carbon* (2015), doi: http://dx.doi.org/10.1016/j.carbon.2015.04.051

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## **ACCEPTED MANUSCRIPT**

Graphene Wrapped Copper-Nickel Nanospheres on Highly Conductive Graphene Film for Use as Counter Electrodes of Dye-sensitized Solar Cells

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#### **Abstract**

A novel architecture of graphene wrapped copper-nickel (Cu-Ni) nanospheres (NSs)/graphene film was proposed to be TCO- and Pt-free counter electrode (CE) with high electrocatalytic activity for dye-sensitized solar cells (DSSCs). The novel architecture CE is composed of highly conductive graphene film, Cu-Ni alloy NSs and the wrapping graphene on the surface of alloy NSs. The graphene film as an electrically conductive layer was synthesized by chemical vapor deposition (CVD) on the insulating SiO<sub>2</sub> substrate, and graphene wrapped Cu-Ni alloy catalyst NSs on the graphene film were *in-situ* formed by the reduction of Cu-Ni acetate and graphene growth using CVD. The graphene wrapped Cu-Ni NSs/graphene film CE shows much superior electrocatalytic activity, compared with graphene film, and the power conversion efficiency of 5.46% was achieved in DSSC devices, which is close to that of Pt/FTO electrode (6.19%). Therefore, the novel architecture of graphene wrapped

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