Edge-selective graphene nanoplatelets as high performance and stability metal-free catalysts in DSSCs

Myung Jong Ju[1], Jae Cheon Kim[2], Kicheon Yoo[2], Jong-Beom Baek[1], and Jae-Joon Lee[2]

[1]Ulsan National Institute of Science and Technology (UNIST), Interdisciplinary School of Green Energy/Institute of Advanced Materials & Devices, 100 Banyeon, Ulsan 689-798, Korea

[2] Departmaent of Energy & Materials Engineering, Dongguk University, 26 Pil-dong, 3-ga, Jung-gu, Seoul, 100-715, Korea

Metal-free carbon-based electrocatalysts for dye-sensitized solar cells (DSSCs) are sufficiently active in Co(II)/Co(III) electrolytes but are not satisfactory in the most commonly used iodide/triiodide (I^-/I_3^-) electrolytes. Thus, developing active and stable metal-free electrocatalysts in both electrolytes is one of the most important issues in DSSC research. Edge-selectively heteroatoms-doped graphene nanoplatelets (XGnPs, X=N, halogens, Se, Te, Sb etc.) were prepared by a simple mechanochemical driven reaction between graphite and target materials. The edge-selective doping and the preservation of the pristine graphene basal plane in the XGnPs were confirmed by various analytical techniques, including atomic-resolution transmission electron microscopy (AR-TEM) and/or time of flight secondary ion mass spectrometry (TOF-SIMS). Here we report the synthesis of edge-selective graphene nanoplatelets (XGnPs) prepared by a simple mechanochemical reaction between graphite and target materials, and their application to counter electrodes (CE) for DSSCs in both Co(II)/Co(III) and I^-/I_3^- electrolytes. Specifically, tested as the DSSC CE in both $Co(bpy)_3^{2+/3+}$ (bpy = 2,2′-bipyridine) and I^-/I_3^- electrolytes, among the XGnPs, the SeGnP-CEs exhibited outstanding electrocatalytic performance with ultimately high stability. The SeGnP-CE–based DSSCs displayed a higher photovoltaic performance than did the Pt-CE–based DSSCs in both SM315-sensitizer with $Co(bpy)_3^{2+/3+}$ and N719-sensitizer with I^-/I_3^- electrolytes. Furthermore, the I_3^- reduction mechanism, which has not been fully understood on carbon-based CE materials to date, was clarified by an electrochemical kinetics study combined with density functional theory (DFT) and nonequilibrium Green’s function calculations (NEGF). These new findings may pave the way for further development of more practical Pt-free CEs, which may help to accelerate the practical applications of DSSCs.