Electronic Supplementary Material (ESI) for Energy & Environmental Science

Electronic Supplementary Information for

Self-Assembly of Layered Double Hydroxide 2D Nanoplates with Graphene Nanosheets: An Effective Way to Improve the Photocatalytic Activity of 2D Nanostructured Materials for Visible Light-Induced O_2 Generation

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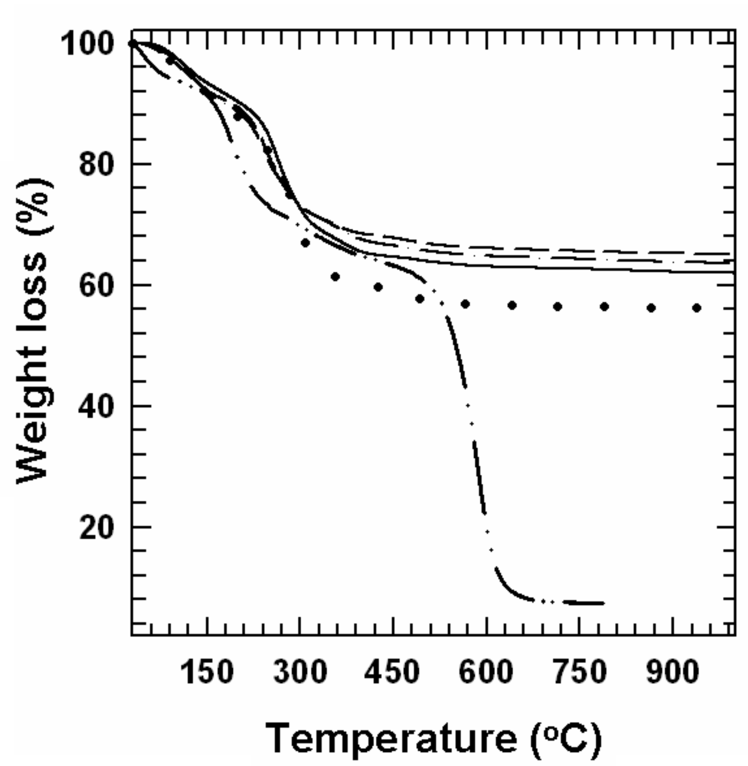
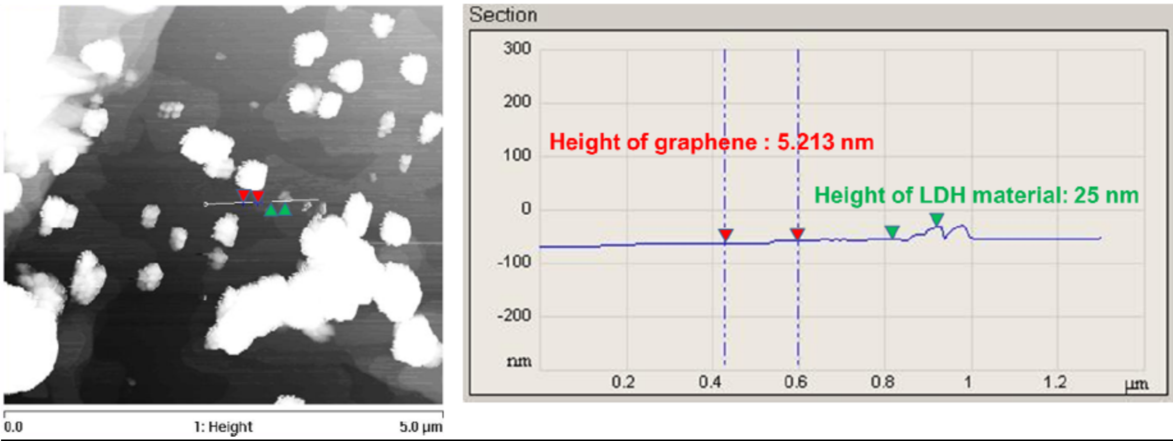
Fig. S1. Thermogravimetric analysis (TGA) data of the self-assembled nanohybrids of **ZCGO-1** (solid lines), **ZCGO-2** (dotted lines), **ZCGR-1** (dashed lines), **ZCGR-2** (dot-dashed lines), and the reference GO (dot-dot-dashed lines).



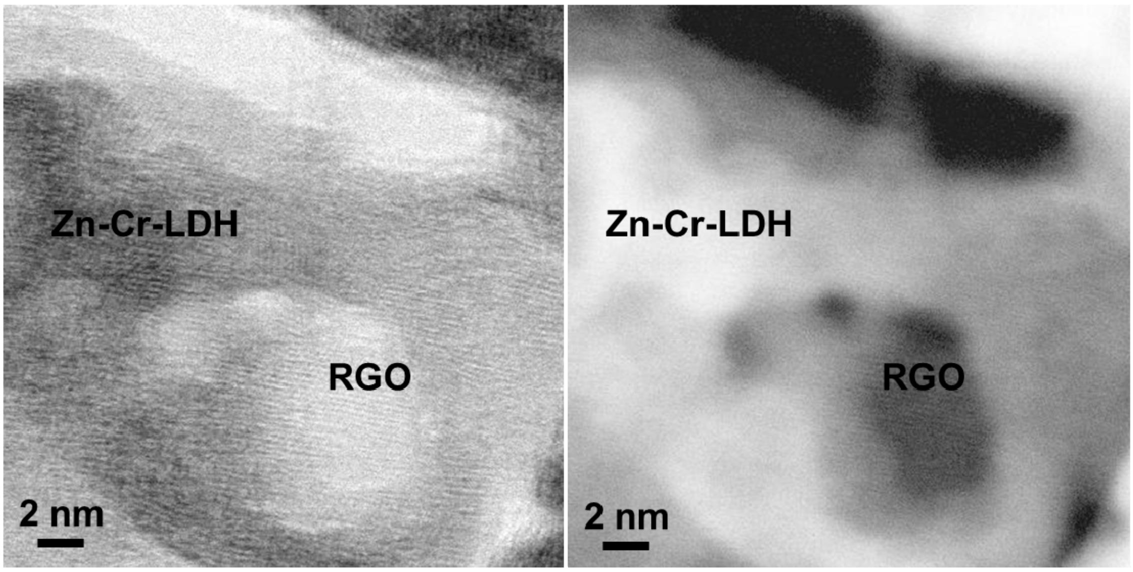
: As plotted in **Fig. S1**, the reference GO displays three steps of weight loss. The first weight loss of ~8% occurs at 30–150 °C, which is attributable to the removal of physisorbed water molecules. The second weight loss of ~22 % at 150–300 °C is related to the decomposition of oxygenated functional groups into gaseous CO_x and H_2O species. In the temperature region of 450–690 °C, there is another abrupt weight loss of ~56% corresponding to the combustion of graphitic carbon layers. Conversely, all of the present Zn-Cr-LDH–graphene nanohybrids show two steps of weight loss. A distinct weight loss of ~9% corresponding to the removal of surface-adsorbed or intercalated water molecules occurs in the temperature range of 30–180 °C. This weight decrease is followed by a larger weight loss of ~23–26% at 180–450 °C, which is attributable to the removal of oxygenated functional groups. In contrast to the reference GO, no weight loss is observable beyond 450 °C, strongly suggesting the enhancement of the thermal stability of graphene nanosheets after the hybridization.

Fig. S2. Atomic force microscopy (AFM) data of the self-assembled **ZCGR-1** nanohybrid.



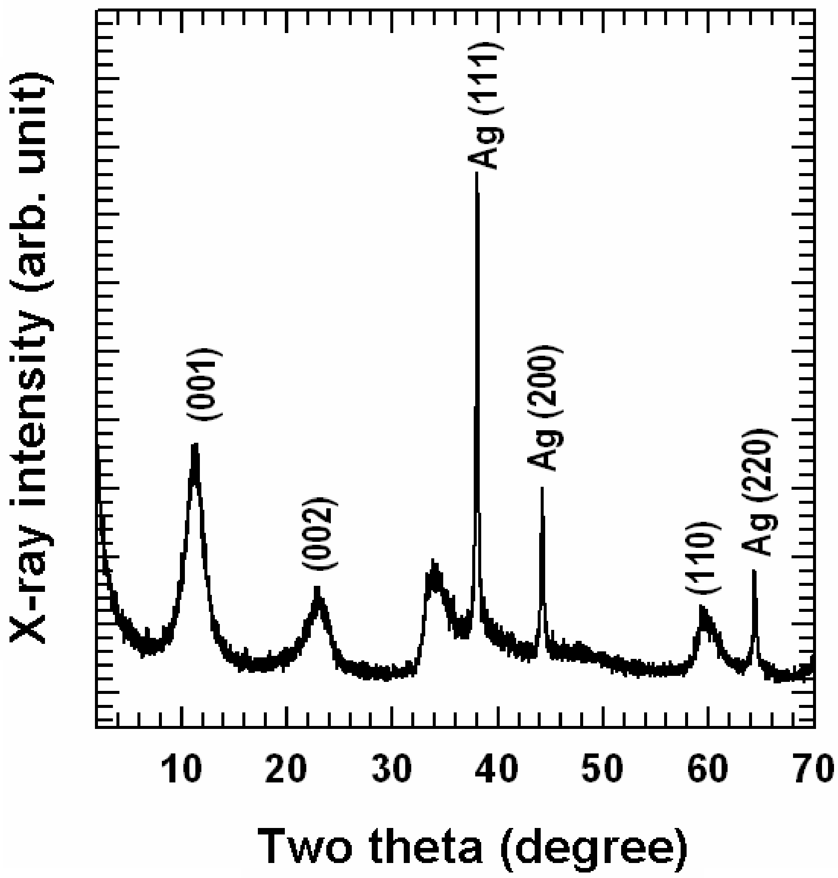
: The formation of the self-assembled nanohybrids of LDH and graphene is confirmed by the atomic force microscopy (AFM). As plotted in **Fig. S2,** the present **ZCGR-1** nanohybrid clearly shows the plate-type morphology with step-like height profile. The lower part with the black color corresponds to the substrate whereas the upper parts with gray color corresponds to the graphene nanosheets. The white-colored domains on the top of gray colored domains can be assigned as the Zn-Cr-LDH material. The present AFM image provides clear evidence for the anchoring of the Zn-Cr-LDH nanoplates on the top of graphene nanosheets.

Fig. S3. (Left) Scanning transmission electron microscopy-bright field (STEM-BF) and (right) Scanning transmission electron microscopy-high angle annular dark field (STEM-HAADF) image of the self-assembled **ZCGR-1** nanohybrid.



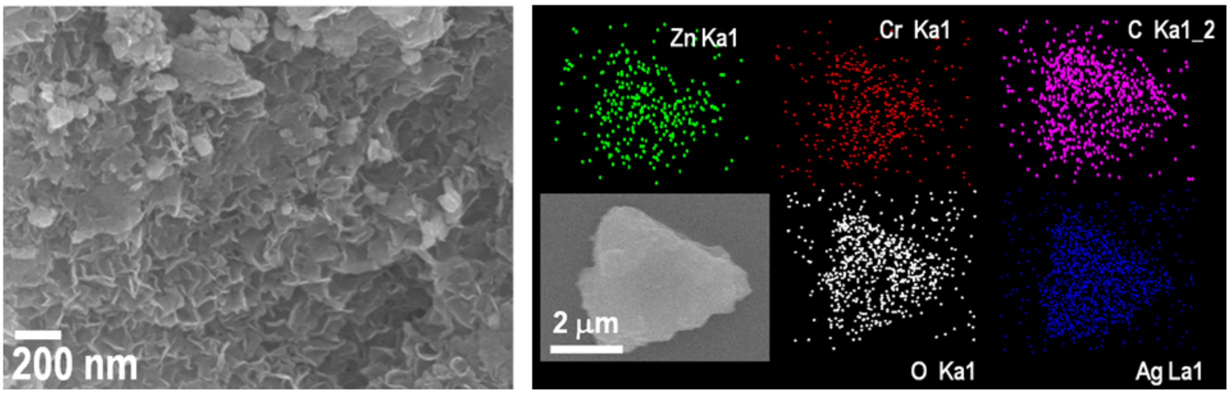
: The formation of the self-assembled nanohybrids of LDH and graphene is confirmed by the HR-TEM and the STEM-HAADF analysis. As plotted in the left panel of **Fig. S3**, the HR-TEM image clearly demonstrate the hybridization of Zn-Cr-LDH domains showing stronger contrast with the RGO domains showing weaker contrast. In the corresponding STEM-HAADF image of the right panel of Fig. S3, the Zn-Cr-LDH domains show a brighter signal than the RGO ones. Since the brightness of STEM-HAADF signal is proportional to the square of atom number of the elements distributed in the corresponding domain, the present result provides straightforward evidence for the existence of heavier elements (Zn and Cr) in the Zn–Cr-LDH domains. The present experimental data provide straightforward evidence for the nanoscale hybridization between Zn-Cr-LDH and RGO species.

Fig. S4. Powder X-ray diffraction (XRD) pattern of the **ZCGR-1** nanohybrid restored after the photocatalyst test.



: The effect of photoreaction on the crystal structure of the Zn-Cr-LDH–graphene nanohybrid is examined with powder XRD analysis. As plotted in **Fig. S4**, the **ZCGR-1** nanohybrid restored after the photocatalyst test still shows its original XRD pattern with the appearance of the XRD peaks of Ag metal, confirming the maintenance of its original crystal structure after the photoreaction and the surface deposition of elemental silver. This finding clearly demonstrates that the present **ZCGR-1** nanohybrid is structurally stable against the photocatalytic reaction.

Fig. S5. (Left) Field emission-scanning electron microscopic (FE-SEM) image and (right) elemental mapping/FE-SEM data of the **ZCGR-1** nanohybrid restored after the photocatalyst test.



: The variations of the crystal morphology and chemical composition of the Zn-Cr-LDH–graphene nanohybrid upon the photoreaction are probed with FE-SEM and energy dispersive spectrometry (EDS)–elemental mapping analysis. As plotted in **Fig. S5**, the corresponding FE-SEM image clearly demonstrates the negligible influence of photoreaction on the porous stacking structure of the **ZCGR-1** nanohybrid. Also, the elemental mapping analysis for the **ZCGR-1** nanohybrid restored from the photoreactor provides clear evidence not only for the maintenance of the original composition of the **ZCGR-1** nanohybrid upon the photoreaction but also for the Ag deposition on the surface of **ZCGR-1** nanohybrid.