

## Electronic Supplementary Information (ESI)

*Revised ESI: published on 19<sup>th</sup> May 2014*

**An exceptionally facile method to produce layered double hydroxides on a conducting substrate and their application for solar water splitting without an external bias**

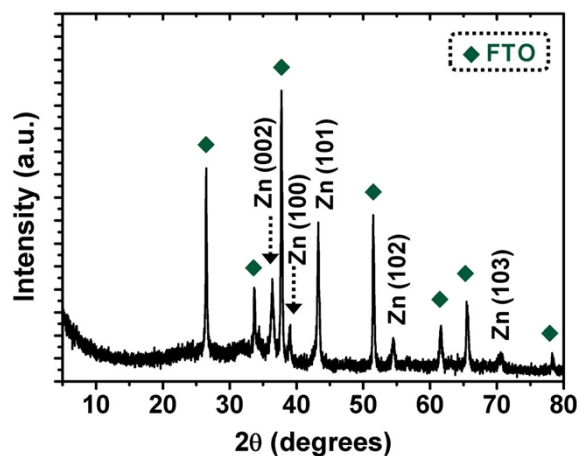
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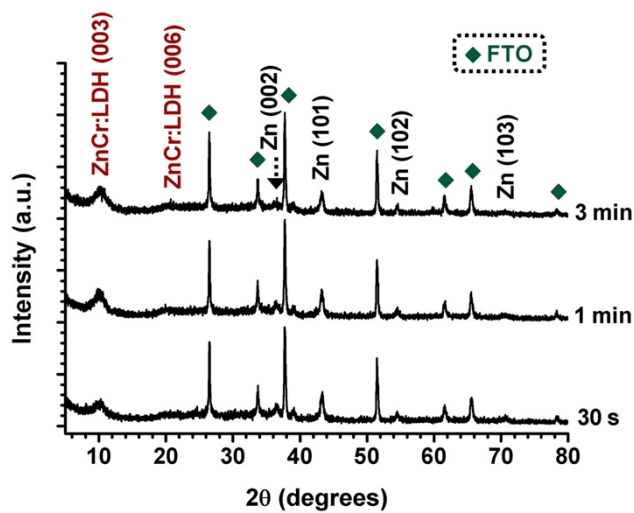
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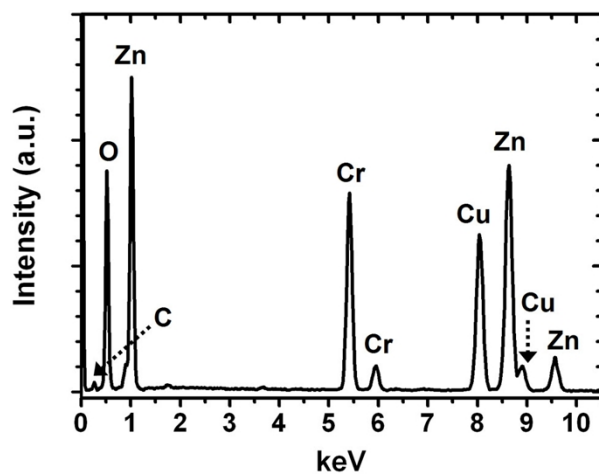
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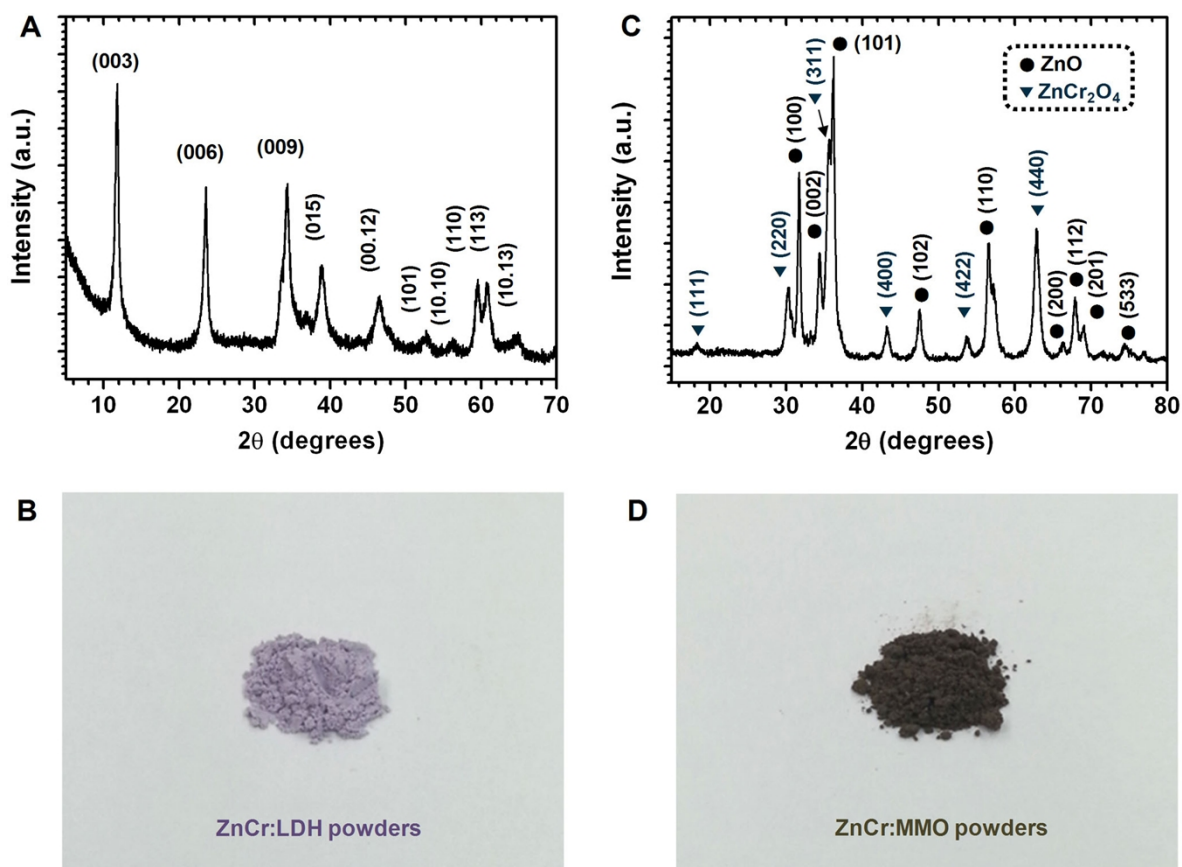
**Figure S1.** XRD pattern of Zn-deposited fluorine-doped tin oxide (FTO) glass.



**Figure S2.** XRD patterns of Zn-deposited FTO glass with different Cr nitrate solution-dipping times.



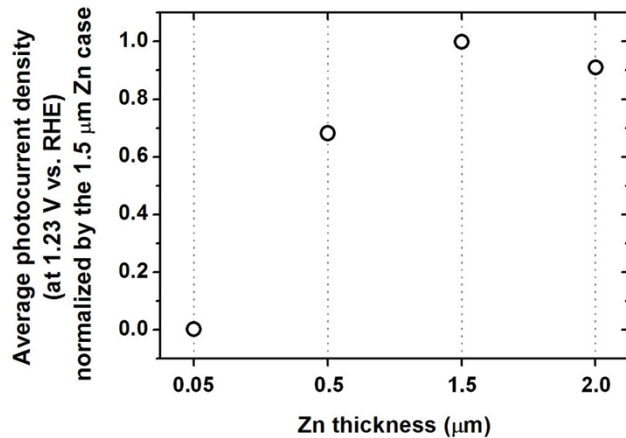
**Figure S3.** EDX pattern of particles detached from the substrate fabricated by the Cr nitrate solution-dipping process followed by calcination at 527°C in air for 2 h.



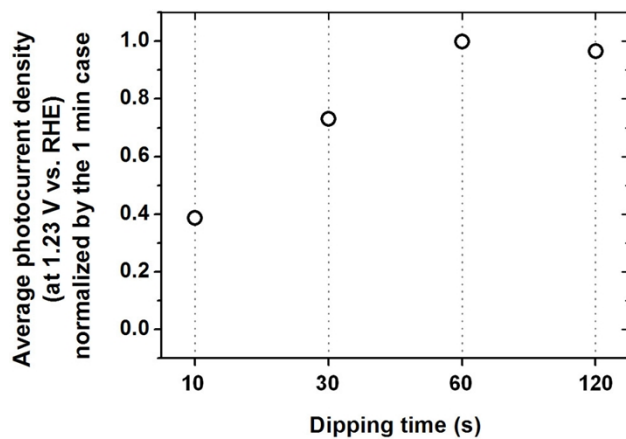
**Figure S4.** (A and B) XRD pattern and photograph of zinc chromium layered double hydroxide (ZnCr:LDH) powders synthesized by mixing of a 30 mL aqueous solution containing 1.4 M zinc nitrate hexahydrate and 0.6 M chromium nitrate nonahydrate and a 40 mL aqueous solution containing 3 M sodium hydroxide and 2.5 M sodium carbonate at 60°C for 24 h. (C and D) XRD pattern and photograph of zinc chromium mixed metal oxide (ZnCr:MMO) powders prepared by calcination of the ZnCr:LDH powders at 527°C in air for 2 h.



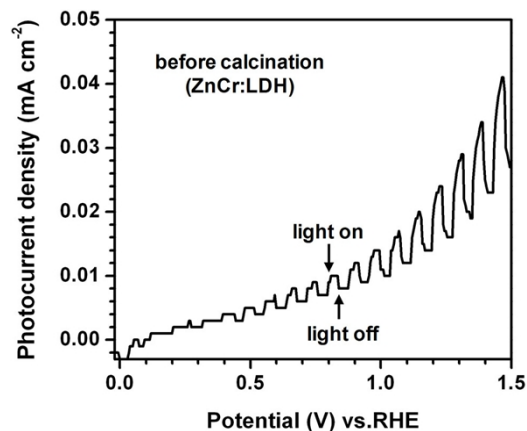
**Figure S5.** Photograph of the ZnCr:MMO/FTO glass fabricated by the Cr nitrate solution-dipping process followed by calcination at 527°C in air for 2 h.



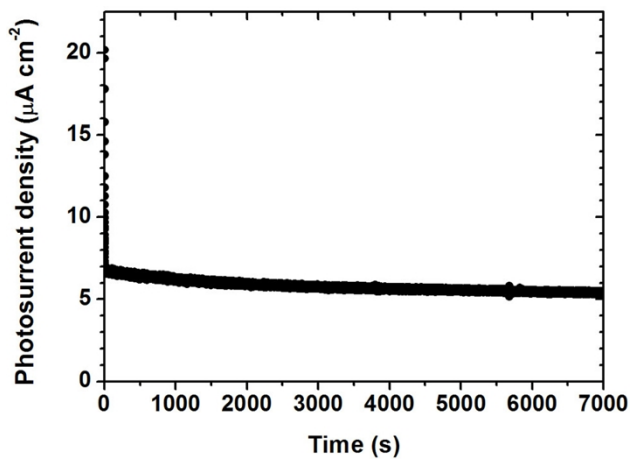
**Figure S6.** Average photocurrent densities at 1.23 V (vs. RHE) normalized by that of the 1.5 μm case as a function of Zn thicknesses. The photocurrent densities were measured in 0.2 M Na<sub>2</sub>SO<sub>4</sub> solution (pH 6.5) under visible light irradiation ( $\lambda > 420$  nm) for the ZnCr:MMO/FTO glass fabricated by dipping of a Zn-deposited FTO glass in a Cr nitrate solution for 1 min, followed by calcination.



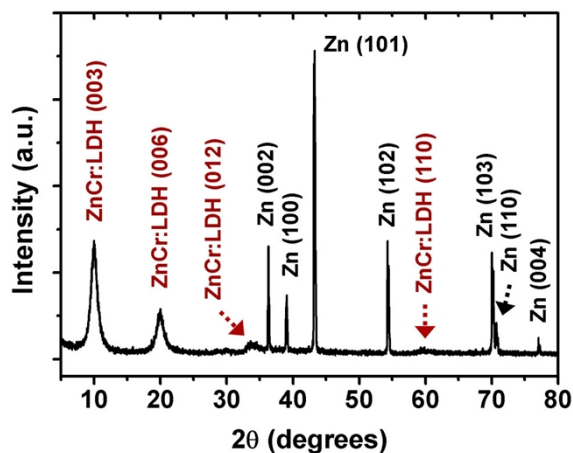
**Figure S7.** Average photocurrent densities at 1.23 V (vs. RHE) normalized by that of the 1 min dipping case as a function of dipping times. The photocurrent densities were measured in 0.2 M Na<sub>2</sub>SO<sub>4</sub> solution (pH 6.5) under visible light irradiation.



**Figure S8.** Current–voltage curve in 0.2 M Na<sub>2</sub>SO<sub>4</sub> solution (pH 6.5) under intermittent visible light irradiation ( $\lambda > 420$  nm) for the ZnCr:LDH/Zn/FTO glass fabricated by dipping of a Zn-deposited FTO glass in a Cr nitrate solution for 1 min.



**Figure S9.** Current–time curve of CoO<sub>x</sub>-deposited ZnCr:MMO photoelectrode in a no-bias, two-electrode configuration with a Pt wire cathode.



**Figure S10.** XRD pattern of a ZnCr:LDH/Zn substrate fabricated by immersing a Zn metal foil in Cr nitrate solution.