

## Layer-by-Layer Assembly for Thin-Film Deposition of Molecular OER Catalysts for Photocatalytic Water-Splitting

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We report a novel and simple method to deposit thin-film of molecular OER (oxygen evolution reaction) catalysts for photocatalytic water-splitting by layer-by-layer (LbL) assembly. Recently, tetracobalt polyoxoanions,  $[\text{Co}_4(\text{H}_2\text{O})_2(\text{PW}_9\text{O}_{34})_2]^{10-}$  were used as a molecular OER catalyst to deposit in the form of thin film by LbL. We found that molecular OER catalysts can be readily deposited onto various electrode surfaces such as gold, titania, and hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) without loss of their catalytic activity. In general, there was a decrease of on-set potential and an increase of current density for electrocatalytic water-splitting at the same time after modification with molecular OER catalysts regardless of types of polyelectrolytes and underlying substrates. We believe that the current study can provide insight to design, build, and test various kinds of electro- and/or photocatalytic systems.