

MATERIAL DESIGN FOR PHOTOCATALYTIC H₂ PRODUCTION FROM WATER SPLITTING

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INTRODUCTION

Energy has been defined as the top of the ten challenges that our society faces and exploitation of renewable energy has become a vital task. Hydrogen has attracted researchers' attention as a green alternative fuel. There are two green ways for hydrogen production, electrocatalytic and photocatalytic water splitting. Compared with electrocatalytic water splitting, photocatalytic water splitting (PCWS) bears the advantages of using abundant solar light instead of electricity and therefore, can be considered as a way more sustainable technology. The working principles of PCWS can be summarized by Fig.1. Under light illumination, semiconducting catalysts generate photo excited electrons and holes. These excitons can separate and become free-moving charges. Then, the charges can transport to the surface of the catalyst and transfer to the adsorbed species to form H₂ and O₂. For the moment, PCWS still suffers from rather low overall efficiency as a result of insufficient charge separation, charge transportation to the interfaces, or charge transfer from the interfaces to the reactant medium that all contribute to the final quantum yield values. Besides, catalyst deactivation^[1] can also happen and result in strong sudden drops of the energy conversion efficiency during the photocatalytic process.

FUNDAMENTAL OF THE PROBLEM

To improve the efficiency of photo-driven water splitting, to face those above-mentioned challenges, we follow several strategies. (a) To harvest more photo-generated charges, catalysts with more suitable bandgaps have been investigated. The value of the band gap defines the threshold energy of absorbed photons, while its position has a strong impact on electron and hole injection over-potentials. (b) Hybridization^[2] of photocatalysts with various materials (e.g. nanocarbons and other metal oxides) has been proven promising to facilitate the charge separation process and reduce charge recombination due to the formation of p-n junctions or quick extraction of one of the charges by the second component. (c) Meanwhile, nano structuring of photocatalysts is favourable for efficient charge extraction from bulk to the surface where the ultimate catalytic reaction takes place. Moreover, mesoporous catalysts with accessible pores provide more reactive sites between catalysts and reactants^[3].

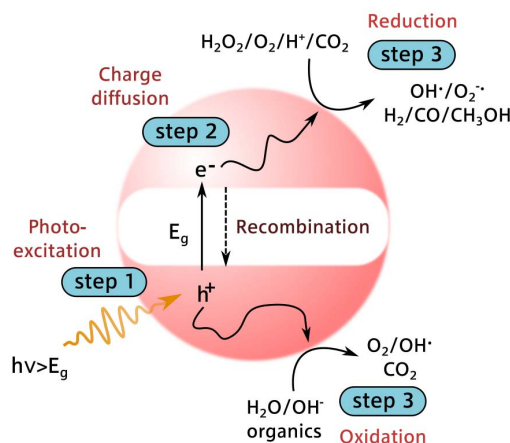


Figure 1. Schematic illustration of working principles of photocatalytic water splitting

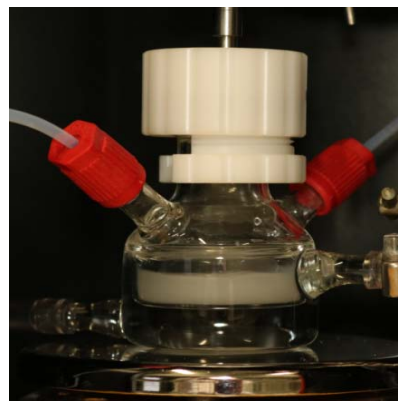


Figure 2. Reactor for water splitting

RESULTS AND DISCUSSION

Based on the above discussions, we have rationally designed a number of materials for efficient H_2 generation. As shown in Fig.3, hybrid Ta_2O_5/CNT (Fig.3a) was prepared that performed superior compared with pure Ta_2O_5 . The presence of carbon nanotube (CNT) indeed facilitated charge separation and reduced charge recombination leading to more charges available for the catalytic conversion. By using the self-assembly of triblock-terpolymer PI-PS-PEO, 3D mesoporous structure of Nb_2O_5 with suitable 10-30 nm pore sizes and long-range order were prepared (Fig.3d). Our photocatalytic H_2 evolution experiments demonstrate that nanostructured photocatalysts with high and accessible surface and short migration lengths for photoexcited charge carriers are primary candidates for high-performance photocatalysts. Therefore, mesoporous Ti-based metal-organic framework (MIL 125-MOF) and its metal oxides (Fig. 3c) were also investigated for PCWS. Furthermore, defects were found to trigger early-stage deactivation in the traditional photo-catalyst TiO_2 with photodeposited Pt as co-catalyst (Fig.3b). Key to this phenomenon is the ratio of Pt atoms to oxygen vacancies, which were created through ultrasonic pretreatment and in situ UV irradiation in the bulk and surface, respectively. Additionally, a possible surface passivation of Pt was observed that could be analogue to known strong metal-support interactions of reducible metal oxides. Pre-calcining the photocatalyst resulted in a stable rate during the course of the experiment.

CONCLUSION

Actually, these strategies inter-played with each other and are complimentary to each other. Rational material design based on these strategies contributed to minimize the challenges and to improve water splitting efficiency.

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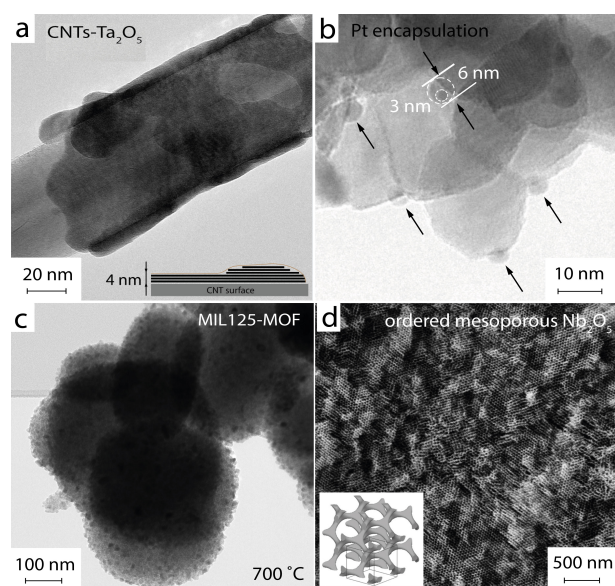


Figure 3. TEM images of novel catalysts designed for more efficient water splitting