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**Abstract**

Solar redox flow cells (SRFCs) have emerged as an alternative all-in-one technology for harvesting and storage solar energy. However, the availability of high performing and cost-effective photoelectrodes is limiting the practical application of SRFCs. Tantalum nitride (Ta3N5) is one of the most prominent semiconductor materials, particularly due to its narrow bandgap (2.1 eV) that allows to reach a maximum theoretical photocurrent-density of 12.9 mA⸳cm-2.[1] For photoelectrochemical (PEC) water splitting, a maximum current density of 12.1 mA⸳cm-2 was reported using opaque Ta3N5 photoelectrodes, gaining advantage over other extensively studied semiconductors.[2] The low photopotential and chemical stability of Ta3N5 are current challenges. Moreover, the transparency is another drawback since it prevents its use in tandem systems, which are needed to increase photopotential for addressing higher potential redox cells (> 0.8 V). [3] Electrophoretic deposition (EPD) is one of the most reported techniques for the synthesis of semi-transparent Ta3N5 photoelectrodes. Domen *et al*. [4] pioneered the use of EPD for depositing Ta3N5 films over fluorine-doped tin oxide (FTO) glasses further coated with a IrO2 co-catalyst; a photocurrent density of 2.0 mA∙cm-2 at 0.2 VAg/AgCl using a 0.1 M Na2SO4 solution as electrolyte was reported for PEC water splitting.

The present work aims at studying the role of each operating and design conditions for preparing efficient and stable semi-transparent Ta3N5 photoelectrodes by EPD over FTO glass substrates. A Ta-doped TiO2 (TTO) underlayer was deposited by atomic layer deposition (ALD), which promotes a better adhesion of the Ta3N5 film to the FTO layer and improves the electron back-contact transport. Different concentrations and thicknesses of TTO were studied and its influence on the Ta3N5 performance was assessed using *J*-*V* curves. The Ta3N5 filmwas also optimized following a systematic procedure that allowed studying the effect of time and applied potential during the EPD as well as different annealing temperatures (500 ºC, 525 ºC and 550ºC) under a flowrate of 100 mL⸳min-1. The best-performing photoelectrode reached a photocurrent-density of *ca.* 4.01 mA⸳cm-2 at 0.2 VAg/AgCl and a photovoltage of 0.5 V (using 0.1 M K4Fe(CN)6 in 1 M KOH as electrolyte); to the best knowledge of the authors, this value is the highest ever reported for semi-transparent Ta3N5 photoelectrodes without dopants and co-catalysts. Following, a high photocurrent of 5.18 mA⸳cm-2 was obtained using a ferrihydrite overlayer and a Co3O4 co-catalyst, which also demonstrated improved stability. Therefore, the optimized multilayered approach proved to be an effective strategy to enhance the performance of Ta3N5 photoelectrodes.