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Ternary Rare Earth Sulfide CaCe<sub>2</sub>S<sub>4</sub>: Synthesis and Characterization of Stability, Structure, Photoelectrochemical Properties in Aqueous Media

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#### **ACCEPTED MANUSCRIPT**

# Ternary Rare Earth Sulfide CaCe<sub>2</sub>S<sub>4</sub>: Synthesis and Characterization of Stability, Structure, and Photoelectrochemical Properties in Aqueous Media

Paola Sotelo<sup>a</sup>, Melissa Orr<sup>a</sup>, Miguel Tayar Galante<sup>a,c</sup>, Mohammad Kabir Hossain<sup>a</sup>, Farinaz Firouzan<sup>a</sup>, Abbas Vali<sup>a</sup>, Jun Li<sup>b</sup>, Mas Subramanian<sup>b</sup>, Claudia Longo<sup>c</sup>, Krishnan Rajeshwar<sup>a</sup>, Robin T. Macaluso<sup>a\*</sup>

#### **Abstract**

A red-orange rare earth ternary chalcogenide, CaCe<sub>2</sub>S<sub>4</sub>, was prepared in powder form by solid-state synthesis. The structural details of this compound were determined by powder X-ray diffraction. The optical band gap of CaCe<sub>2</sub>S<sub>4</sub> was determined by diffuse reflectance spectroscopy (DRS) to be ~2.1 eV, consistent with the observed red-orange color. Quantitative colorimetry measurements also support the observed color and band gap of CaCe<sub>2</sub>S<sub>4</sub>. Both direct and indirect optical transitions were gleaned from Tauc analyses of the DRS data. Photoelectrochemistry experiments on CaCe<sub>2</sub>S<sub>4</sub> films showed *n*-type semiconductor behavior. Analyses of these data via the Butler-Gärtner model afforded a flat-band potential of -0.33 V (vs. Ag/AgCl/KCl 4 M) in ~pH 9 aqueous sulfite electrolyte. The potential and limitations of this material for solar water splitting and photocatalytic environmental remediation (e.g., dye photodegradation) are finally presented against the backdrop of its photoelectrochemical stability and surface hole transfer kinetics in aqueous electrolytes.

Graphical abstract

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