A high efficiency solar conversion of CO_2 and water into formate and O_2 over a month using mixed oxide photoelectrodes

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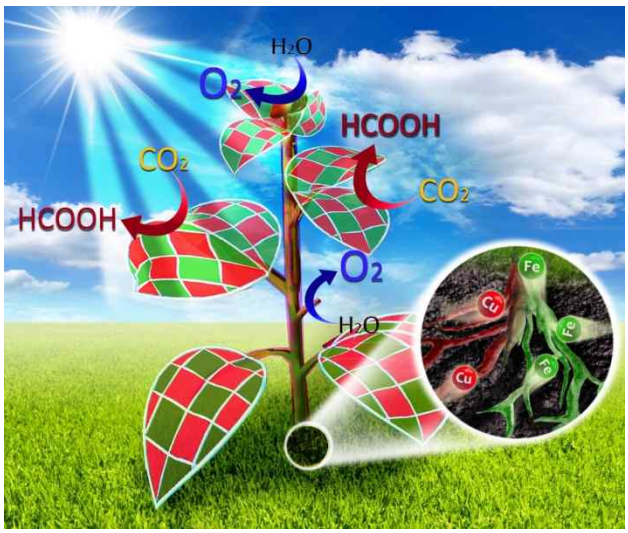

Solar conversion of carbon dioxide and water to value-added chemical fuels remains a challenge.[1-7] A number of solar-active materials have been reported but still require expensive components and complicated synthetic processes and operate mostly under chemical or electrical bias.[2,4] In addition, these materials suffer from low selectivity, poor energy efficiency, and instability and fail to drive simultaneous water oxidation. Herein, we report $CuFeO_2$ and CuO mixed p-type materials fabricated via a widely employed electroplating of earth-abundant cupric and ferric ions followed by annealing under atmospheric air.[8,9] Wired $CuO/CuFeO_2$ photocathode and Pt anode couples produced formate over 1 week at a solar-to-formate energy conversion efficiency of ~1% (selectivity > 90%) without any external bias while O_2 was evolved from water under simulated solar light (Air Mass 1.5, 100 $mW \cdot cm^{-2}$). Isotope and nuclear magnetic resonance analyses confirmed the simultaneous production of formate and O_2 at the stand-alone couples. An as-synthesized photocatalyst film with a three-dimensional, double-layer configuration further shows the continued production of formate for over 17 days. However, the crystalline structure and elemental state of the used photocatalysts undergo gradual chemical reduction. Such a deformation can be thermally healed by recycling the weekly used samples via oxidative annealing. Thus, a single photocatalyst sample produces formate continuously for 35 days. The photocatalyst components (Cu, Fe, and O) are earth-abundant, and the photocatalyst synthesis is straightforward, facile, environmentally benign, reproducible, and scalable. On achieving higher efficiencies in the future, the practical applicability of these photocatalysts will become enormous.

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