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Understanding the Material Thermodynamics of Two-Step Solar Thermochemical Water-Splitting Cycles[1] BRYCE MEREDIG, CHRIS WOLVERTON, Northwestern University Department of Materials Science and Engineering, SUNSHINE TO PETROL GRAND CHALLENGE AT SANDIA NATIONAL LABORATORIES TEAM — Metal oxide materials may be used in two-step solar thermochemical water-splitting cycles to renewably produce hydrogen: At high temperature, the oxide material is reduced, and at a lower temperature, the material re-oxidizes upon contact with water vapor producing hydrogen gas. Here, we present the first completely general analysis of the equilibrium thermodynamics of a two-step metal oxide water splitting cycle. We determine the temperature and pressure regimes in which both steps of the cycle are thermodynamically favorable in terms of the enthalpy and entropy of oxide reduction. Armed with this thermodynamic construct, we apply computational methods, such as density functional theory (DFT) and CALPHAD modeling, to assess many proposed oxide cycles. Using CALPHAD thermodynamic data, we survey a large number (more than 100) binary oxide redox couples, and show that none have both thermodynamically favorable reduction and oxidation steps. In an effort to find more thermodynamically favored redox couples, we investigate the utility of DFT calculations to screen materials' thermodynamics. In addition, we identify several driving forces which could enable more efficient two-step cycles, including a large positive solid-state entropy of reduction of the oxide.

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