Simulations of the irradiation and temperature dependence of the efficiency of tandem photoelectrochemical watersplitting systems

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The instantaneous efficiency of an operating photoelectrochemical solar-fuels-generator system is a complicated function of the tradeoffs between the light intensity and temperature-dependence of the photovoltage and photocurrent, as well as the losses associated with factors that include ohmic resistances, concentration overpotentials, kinetic overpotentials, and mass transport.

These tradeoffs were evaluated quantitatively using an advanced photoelectrochemical device model comprised of an analytical device physics model for the semiconducting light absorbers in combination with a multi-physics device model that solved for the governing conservation equations in the various other parts of the system. The model was used to evaluate the variation in system efficiency due to hourly and seasonal variations in solar irradiation as well as due to variation in the isothermal system temperature. The system performance characteristics were also evaluated as a function of the band gaps of the dual-absorber tandem component and its properties, as well as the device dimensions and the electrolyte conductivity.

The modeling indicated that the system efficiency varied significantly during the day and over a year, exhibiting local minima at midday and a global minimum at midyear when the solar irradiation is most intense, as depicted in figure 1. These variations can be reduced by a favorable choice of the system dimensions, by a reduction in the electrolyte ohmic resistances, and/or by utilization of very active electrocatalysts for the fuel-producing reactions. An increase in the system temperature decreased the annual average efficiency and led to less rapid ramp-up and ramp-down phases of the system, but reduced midday and midyear instantaneous efficiency variations. Careful choice of the system dimensions resulted in minimal change in the system efficiency in response to degradation in the quality of the light absorbing materials. The daily and annually averaged mass of hydrogen production for the optimized integrated system compared favorably to the daily and annually averaged mass of hydrogen that was produced by an optimized stand-alone tandem photovoltaic array connected electrically to a stand-alone electrolyzer system.

The model can be used to predict the performance of the system, to optimize the design of solar-driven water splitting devices, and to guide the development of components of the devices as well as of the system as a whole.

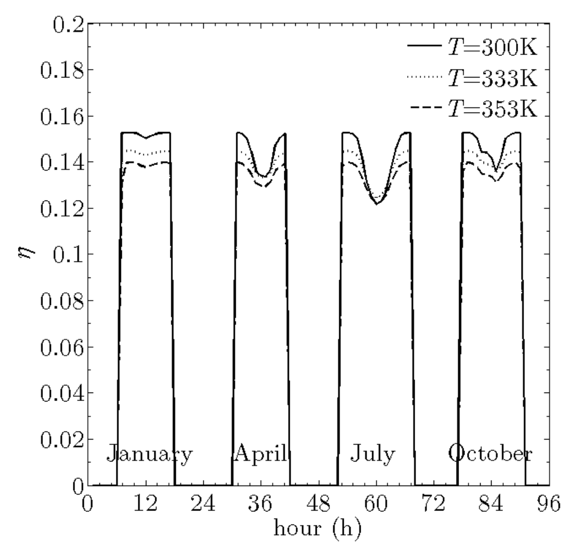

Figure 1. Efficiency of a PEC device for four typical seasonal days at three isothermal conditions (T = 300, 333, 353 K), for the Si/GaAs dual-absorber tandem cells.