

Activated Carbon Catalysts for the Production of Hydrogen via the Sulfur-Iodine Thermochemical Water Splitting Cycle

Daniel M. Ginosar^{1*}, Lucia M. Petkovic¹, Harry W. Rollins¹, Kyle C. Burch¹, Cristina Deiana²,

Hugo S. Silva², Maria F. Sardella², and Dolly Granados²

¹Idaho National Laboratory, Idaho Falls, ID, 83415-2208 (United States)

²Universidad Nacional de San Juan, San Juan 5400 (Argentina)

*Daniel.Ginosar@inl.gov

Introduction

Thermochemical cycles produce hydrogen through a series of chemical reactions where the net result is the production of hydrogen and oxygen from water at much lower temperatures than from direct thermal decomposition [1]. All chemicals within the cycle are recycled and the heat to drive the reactions, which tend to be endothermic, must be provided by a primary energy source. When the primary energy driver is nuclear or solar heat, hydrogen can be generated without producing greenhouse gasses, and can provide independence from our dwindling supplies of fossil fuels.

Among the high number of thermochemical water-splitting cycles proposed in the literature, the Sulfur-Iodine (S-I) cycle [2] has generated considerable interest. The S-I cycle consists of three simultaneous reactions; the decomposition of hydroiodic acid (HI) to produce hydrogen and generate iodine for recycle, the decomposition of sulfuric acid to produce oxygen and generate SO₂ for recycle, and a main reaction where water and the recycle chemicals react to regenerate HI and sulfuric acid.

Previous literature has reported that activated carbon is a desirable catalyst for the decomposition of hydroiodic acid [3]; however, the affect of carbon properties on reactivity has not been reported. In this presentation we will discuss the effect of activated carbon pore morphology surface properties on catalytic hydrogen production activity.

Materials and Methods

Activity measurements were performed on seven activated carbon samples whose surface areas (SA) ranged from approximately 200 to 2,000 m²/g. The experimental setup consisted of a high-temperature, continuous-flow testing system built of Teflon tubing, glass, and quartz. The catalyst sample was loaded in a quartz tube reactor and the temperature was increased to 250°C. HI acid at 55 wt% was then pumped into the system where the vaporized HI gas was reduced to H₂ and I₂ over the catalyst bed. Temperature was then increased to 500°C at a rate of 50°C/hr. Iodine and unreacted HI gas combined with H₂O in the colder sections downstream of the reactor and was collected as a liquid solution. The gaseous H₂ product was analyzed by gas chromatography.

Results and Discussion

Hydrogen yields at temperatures ranging from 250 to 500°C as a function of sample SA are shown in Figure 1. SA had a strong influence on hydrogen yield. A maximum hydrogen yield was observed using a sample surface area of 1030 m²/g with minimums in yield observed at the low and high extremes of sample surface area.

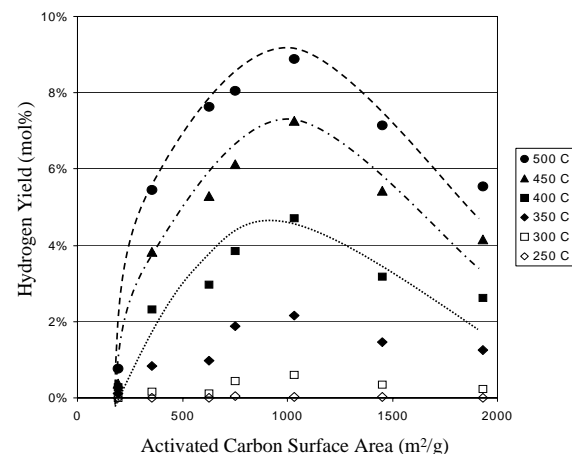


Figure 1. Hydrogen yields at temperatures from 250 to 500°C as a function of sample surface area.

Nitrogen physisorption analysis of the fresh activated carbon catalysts found that samples with surface areas above 1,000 m²/g had greater porosity in the micro-pore region than lower surface area samples and the fraction of micro-pores increase with increasing surface area.

Spent samples with fresh surface areas of approximately 300 and 2,000 m²/g, which provided similar hydrogen yields, were subjected to temperature programmed desorption (TPD). TPD produced a 28 % weight decrease from both samples, but most iodine species desorbed from the low SA sample at significantly lower temperatures than from the high SA sample. The higher temperatures required to desorb iodine species from high SA sample may explain lower activity despite its high surface area.

Significance

The S-I thermochemical water-splitting cycle can produce hydrogen without the generation of greenhouse gasses. For this cycle to become practical, minimum reactor size and catalyst loadings will be essential. This effort is developing an understanding of the affect of activated carbon properties on catalytic hydrogen production activity necessary to achieve that goal.

References

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