Argonne investigates alternate hydrogen-production technique

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BMW's Hydrogen 7 is among the first hydrogen-drive luxury performance automobiles for everyday use. Being built in a limited series, it is equipped with a 260-hp internal combustion engine capable of running either on hydrogen or on gasoline and is based on the BMW 7 Series. The car's hydrogen storage tank holds approximately 17.6 lb of liquid hydrogen. Engine power and torque remain identical regardless of the fuel used. *(photo courtesy of BMW)*

The "hydrogen economy" aims to reduce the USA's reliance on imported fossil fuels along with the emission of greenhouse gases. Simulation is helping find the most effective path towards that goal.

A major US-based research initiative is looking at ways to make hydrogen a major transportation fuel that could largely replace fossil fuels. A first step is to find the most cost-efficient way to generate this hydrogen from domestic feedstocks without creating unnecessary greenhouse gases and yet be economically viable. Thus the US Department of Energy is investigating the use of nuclear energy for hydrogen production through the Nuclear Hydrogen Initiative. Although hundreds of possible thermochemical cycles have been identified, only a few seem to promise technical feasibility.

A team at Argonne is examining the potential of the calcium-bromine (Ca-Br) water-splitting cycle, where that material reacts endothermically with water to create calcium oxide (CaO) and hydrogen bromide (HBr). The latter is then converted through electrolysis or through a plasma decomposing process into bromine (Br_2) and hydrogen (H_2).

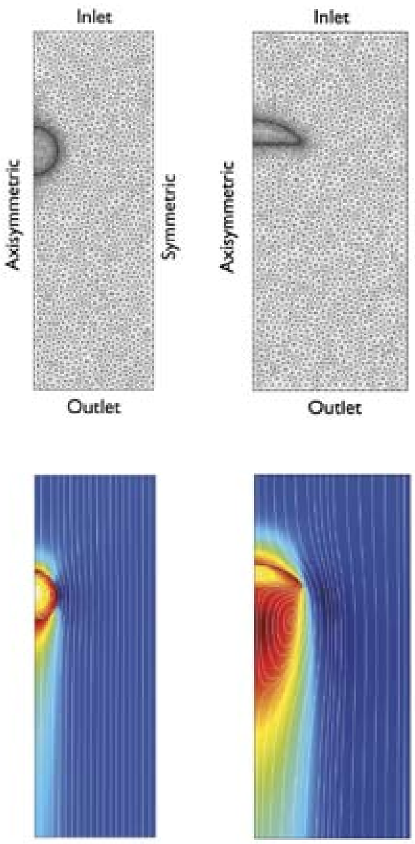

Figure 1. Mesh plot and results from the cases of a steam spherical bubble and steam spherical-cap bubble flowing through molten $CaBr_2$. The results show the velocity magnitude in the vertical direction (color plots), and vector (streamline plots) in both the bubbles and fluid flowing around the bubbles. The spherical bubble exhibits no recirculation in its wake, whereas the spherical-cap bubble does.

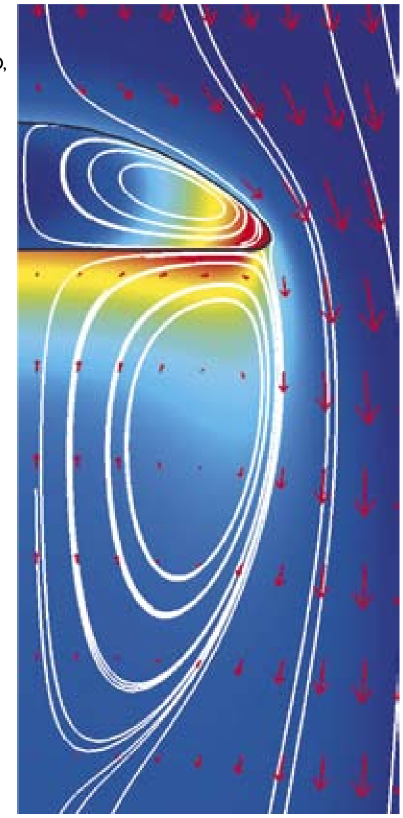

Figure 2: Concentration of the product, HBr, in the bubble and the product, CaO, in the molten $CaBr_2$. Here you can see the extent that CaO accumulates in the recirculation zone in the wake of the spherical cap bubble.

This cycle is particularly attractive because although it is highly endothermic, nearly half of the required thermodynamic energy for water splitting is delivered as nuclear heat.

Methods of $CaBr_2$ hydrolysis

Two options are under consideration for bringing the $CaBr_2$ (which carries much of the heat required for reaction) and steam together in a continuous process: either spraying molten $CaBr_2$ into a steam environment, or sparging bubbles of steam through a pool of molten $CaBr_2$. The COMSOL Chemical Engineering Module provides an environment wherein models for these complex interacting phenomena can be built and studied.

For the spraying method, the researchers ran simulations with droplet diameters and steam inflow velocities over two orders of magnitude. They found that for larger Reynolds numbers, a recirculation zone develops in the droplet wake into which reaction products tend to be swept rather than moving efficiently away from the region of reaction. They also discovered that the latent heat from solidifying $CaBr_2$ droplets supplies only a small fraction of the heat required for reaction of the entire droplet.

Can sparging steam do better?

What about the molten-pool method? The low viscosity of molten $CaBr_2$ opens up the possibility of using it as a heat reservoir, sparging steam into the pool as a bubble column. The modeling first studied the two limiting cases—the spherical and spherical-cap bubble re gimes. With the former, the ratio of the reacting surface to volume is quite large, but producing small bubbles in a high-temperature molten salt in the laboratory is challenging. In contrast, the faster flow with spherical-cap bubbles induces turbulence and a wake where chemical reaction product species might accumulate and slow down the reaction at the interface behind the bubble (Figure 1).

Simulation results showed that only negligible amounts of the product, CaO, are drawn into the wake of the spherical-cap bubble, precluding any possible reverse reaction and reduction of conversion (Figure 2). Furthermore, steam diffusion and induced recirculation within the bubble itself bring steam to the bubble surface at a rate sufficient to keep up with the reaction.

Funding for alternative cycles is limited, so modeling identifies designs that are not likely to be successful. With COMSOL we are able to quickly get rough answers to many questions that helped our decision-making. ■

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