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Experiment and numerical analysis for sulfuric acid decomposition reaction for applying hydrogen by nuclear

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ABSTRACT

Nuclear hydrogen production is a large-scale process via direct thermochemical water splitting, using the high heat output of the very high temperature reactor (VHTR). However, transferring the high heat output of Helium (the gas-coolant for VHTR) to the SI (Sulfur–Iodine) thermochemical cycle involves the great challenge of developing a heat exchanger material capable of withstanding high temperatures and pressures. During the sulfuric acid decomposition reaction, which has the highest heat requirement in the SI thermochemical cycle, maintaining decomposition efficiency close to the equilibrium conversion rate, despite the elevated pressure, will afford greater flexibility in heat exchanger design. Herein, to determine the optimal pressure for VHTR-SI thermochemical hydrogen production, we performed *sulfuric acid decomposition* under atmospheric (0.4 kgf/cm²) and elevated (5.0 kgf/cm²) pressure conditions, and analyzed their respective decomposition efficiencies. Additionally, we performed a thermal hydraulic simulation to investigate the conditions inside and outside the reactor, which are difficult to verify experimentally, by simulating the interior of the reactor for the pressurized sulfuric acid decomposition system.

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Introduction

Nuclear hydrogen production is a large-scale hydrogen production process via direct thermochemical water splitting, using the high heat output of the very high temperature reactor (VHTR). A nuclear hydrogen system consists of a VHTR system, an intermediate heat exchange system, and a hydrogen production system [1,2].

The “Energy Depot” project [3] was carried out in the early 1960s with the purpose of producing fuel from air and water, which are both renewable resources that can be replenished from the environment. Fuel candidates include hydrogen, ammonia, and hydrazine. It was hypothesized that a portable high temperature reactor could be used as a heat source. Water electrolysis, whose thermal efficiency is hypothesized to be 24% [4], was taken as the comparison process. Since

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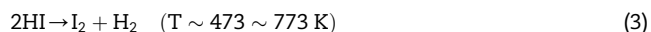
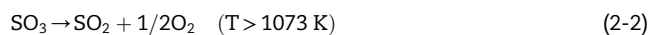
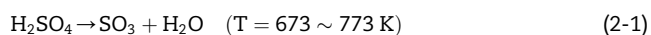
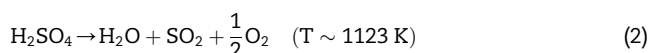
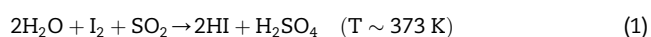
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then, water splitting technology induced by thermochemical cycles has been extensively studied. In particular, the “International Round Table on Direct Production of Hydrogen with Nuclear Heat”, held in Ispra, Italy, gave rise to theoretical discussions regarding hydrogen production in various thermochemical cycles [5–15]. For such thermochemical water-splitting technologies, solar energy and VHTR are regarded as the primary heat sources. Specifically, attempts at using the VHTR in thermochemical cycles requiring temperatures below 1273 K are underway. On the occasion of the successful continuous operation of a bench-scale closed cycle by the Japanese Atomic Energy Agency (JAEA), the sulfur–iodine (SI) thermochemical hydrogen production technology took center stage as one of the high-practical-potential hydrogen production technologies that could be coupled with the VHTR [16–20].

The SI process, wherein hydrogen is produced through thermochemical water splitting using the high heat output (~1223 K) of He, which is used as the coolant for the VHTR, was first studied by the American company General Atomics in the mid-1970s. According to reports published in 1982, the SI process consists of three types of chemical reactions [21,22]:



Equation (1) represents an exothermic reaction referred to as the Bunsen reaction. Equation (2) represents the sulfuric acid decomposition reaction, which is divided into two stages viz. thermolysis and subsequent catalyst decomposition, as expressed by Equations (2-1) and (2-2), respectively. Equation (3) represents the HI decomposition reaction, which is either a solid-state or a homogeneous catalytic reaction in the gas or liquid phase, respectively.

In the SI thermochemical cycle, the sulfuric acid and iodine cycles are connected by the Bunsen reaction. As shown in Equation (1), in the Bunsen reaction, H_2SO_4 and HI are produced from SO_2 and H_2O supplied from the sulfuric acid cycle and I_2 supplied from the iodine cycle; subsequently, phase-separated H_2SO_4 and HI are fed back to each cycle. In the sulfuric acid cycle, expressed by Equation (2), H_2SO_4 is decomposed into SO_2 , O_2 , and H_2O , whereby a half-loop cycle is formed. In the iodine cycle, expressed by Equation (3), HI is decomposed into I_2 and H_2 , wherein H_2 is separated and discharged and I_2 is recovered back to the Bunsen reaction process, thus constituting a half cycle.

The sulfuric acid decomposition reaction (Equation (2)), which is a key part of the SI thermochemical cycle, proceeds at high temperatures of 1073 K or even above 1173 K in the presence of a catalyst. As such, it has the highest thermal energy requirement in the SI thermochemical cycle. Consequently, it is the first to be supplied with the high heat output from He.

However, the coolant (He) has a high circulation pressure of 50–70 kgf/cm². Hence, the heat exchanger that transfers the high heat output of He to the thermochemical SI cycle producing hydrogen, especially the sulfuric acid decomposition reaction, should withstand not only high temperatures, but also the pressure gap within the heat exchanger, i.e. between the high-pressure side involving He and the relatively low-pressure side involving the sulfuric acid decomposition reaction. As no material that can simultaneously withstand temperatures exceeding 1173 K and overcome a pressure gap of 70 kgf/cm² currently exists, research efforts revolve around the reduction of the pressure gap between the two sides of the heat exchanger.

As shown in Equation (2), the total moles of gaseous products in the sulfuric acid decomposition reaction increase as the reaction advances. Consequently, as per Le Chatelier's law [23], the reaction is hindered on increasing the pressure. However, as the gas is compressed in proportion to the increase in pressure, it is likely to stay longer in the reactor. The longer the gas stays in the reactor, because of the reduced rate of the sulfuric acid decomposition reaction, the closer to the equilibrium conversion rate the reaction speed becomes.

Drawing on this fact, we performed *pressurized sulfuric acid decomposition* reaction experiments under atmospheric (0.4 kgf/cm²) and elevated (5.0 kgf/cm²) pressure conditions, and analyzed their respective decomposition efficiencies for application to the VHTR-SI thermochemical cycle producing hydrogen. To investigate the reaction conditions both inside and outside the reactor, which are difficult to verify experimentally, a thermal hydraulic simulation was performed, along with the pressure experiments, by simulating the interior of the unit reactor for the pressurized sulfuric acid decomposition system.

Experimental and simulation

Silicon carbide (SiC)-based pressurized sulfuric acid decomposition reaction system

Previous studies [24] verified that a Pt-lined reactor could be used for a high-pressure sulfuric acid decomposition system. However, fabricating large-scale equipment using a precious metal such as Pt is not practical. Therefore, materials having ceramic properties, such as SiC or Fe_xSi , have been proposed as candidate materials because of their corrosion resistance to sulfuric acid and ability to withstand high pressure [25–28].

Fig. 1 illustrates the unit reactor of the pressurized sulfuric acid decomposition (PSAD) reaction system, consisting of reactor and heater. A SiC-based PSAD reactor was designed as a tube-shaped bayonet-type one-piece reactor so that all reactions, namely, evaporation of sulfuric acid solution, thermolysis, and catalytic decomposition, would occur within the reactor. The outer and inner diameters of the reactor, and its height; were 31.5, 25, and 1400 mm, respectively. A SiC tube with a hemispherical head was used as the top part of the reactor, the high-temperature region where heat exchange with the outside occurs. The manifold into which the sulfuric acid solution is injected was fabricated with a PTFE (i.e. Teflon[®]) material so that the sulfuric acid solution supplied

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