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Boiling heat transfer characteristics of a sulfuric-acid flow in thermochemical iodine–sulfur cycle

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ABSTRACT

The Japan Atomic Energy Agency has been conducting research and development on the thermo-chemical iodine–sulfur (IS) process, which is one of the most attractive water-splitting hydrogen production methods that uses nuclear thermal energy. The sulfuric acid decomposer is one of the key components of the IS process. The boiling heat transfer coefficients of sulfuric acid solutions are required to design the sulfuric acid decomposer. These coefficients were measured in aqueous solutions where the mole fraction of H_2O ranged from 0.17 to 0.37 (heat flux range from 16.9 kW/m^2 to 5.6 kW/m^2) and compared with the empirical correlations formulated for binary mixtures. A combination of the Stephan–Körner correlation, using the empirical constant $A_0 = 2.00$, and the Nishikawa–Fujita correlation was used to predict the experimental results with an accuracy of ±10%.

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Keywords: Hydrogen production; Iodine–sulfur process; Sulfuric acid; Boiling heat transfer; Binary mixture; Thermo-chemical

1. Introduction

Hydrogen is expected to serve as a clean alternative energy source because it can be produced from water and converted back to it after being used as a fuel. However, to realize a hydrogen energy system, the development of efficient, economical hydrogen production methods that can meet the immense energy demand in future is required. The iodine–sulfur (IS) process, which is a thermo-chemical water-splitting cycle, is a promising candidate for such a hydrogen production method. To date, extensive research on the IS process has been reported worldwide (Kubo et al., 2012; Moore et al., 2009; Park et al., 2013; Zhang et al., 2010; Le Duigou et al., 2007).

The Japan Atomic Energy Agency (JAEA) has been conducting research and development on the IS process for the production of hydrogen using the nuclear thermal energy from a high-temperature gas-cooled reactor (HTGR) as a heat source (Kubo et al., 2012). The IS process consists of three chemical reactions. In the process, the raw material, water, reacts with iodine and sulfur dioxide to produce hydrogen iodide (HI) and sulfuric acid (H_2SO_4). The reaction proceeds exothermically below 100 °C and is called the Bunsen reaction. Endothermic decomposition of the HI and H_2SO_4 at elevated temperature produces hydrogen and oxygen, respectively. The highest temperature reaction is SO_3 decomposition to produce SO_2 and oxygen at approximately 850 °C in the presence of the catalyst. The closed-cycle operation of the thermo-chemical reactions produces the free energy required for water splitting (Onuki et al., 1994).

Continuous hydrogen production was demonstrated for 1 week at a hydrogen production rate of approximately 0.03 N m^3/h using a bench-scale test apparatus made of glass at the JAEA (Kubo et al., 2004). As a next step in IS process development for the realization of future nuclear hydrogen production, JAEA has been conducting research and development on reactors made of engineering materials (Onuki et al., 2011). A sulfuric acid decomposer is one of the key components that is exposed to the severest corrosive environment in the IS process, including a high-temperature sulfuric acid boiling flow. Terada et al. (2006) proposed a block-type sulfuric acid decomposer made of SiC ceramic that has excellent corrosion resistance and mechanical strength performance.

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Nomenclature

A_0	empirical constant, dimensionless
C_1, C_2	constant number determined by physical properties, dimensionless
f_p	pressure factor (=1), dimensionless
f_ξ	nucleation factor (=1), dimensionless
h	heat transfer coefficient, W/m^2 K
P	pressure, bar
q	heat flux, W/m^2
T	temperature, °C
T_{bo}	boiling point, °C
T_{de}	dew point, °C
x	molar fraction in the liquid phase, dimensionless
x'	molar fraction in the liquid phase modified for an azeotropic mixture, dimensionless
y	molar fraction in the gas phase, dimensionless
ΔT_E	temperature difference between the dew point and the boiling point, K
ΔT_{id}	ideal wall superheat, which is the molar averaged wall superheat for the pure components, K
ΔT_{sat}	wall superheat

Subscripts

1	low-boiling point component, which is H_2O in this paper
2	high-boiling point component, which is H_2SO_4 in this paper
az	azeotropic concentration of sulfuric acid (0.08 molar fraction of H_2O)
H_2O	water
w	wall
L	liquid

A mock-up model of the proposed sulfuric acid decomposer was test-fabricated to confirm the fabricability and structural integrity (Terada et al., 2006; Noguchi et al., 2006). Meanwhile, a bayonet-type sulfuric acid decomposer made of SiC ceramics was proposed by Sandia National Laboratory and an experimental study using sulfuric acid was carried out (Gelbard et al., 2007; Moore et al., 2009). Nagarajan et al. (2008) performed thermal and chemical reaction analysis on a bayonet-type sulfuric acid decomposer using the FLUENT software package. Park et al. (2013) performed a process simulation and an experimental study of the sulfuric acid decomposition process, in which a bayonet-type sulfuric acid exchanger was used.

With respect to the boiling heat transfer coefficients of sulfuric acid solutions, the Nishikawa–Fujita correlation (Nishikawa and Fujita, 1977), an empirical correlation for single component systems, was used in the first stage of the conceptual design of the sulfuric acid decomposer in JAEA. Through follow-on boiling heat transfer experiments conducted by Noguchi et al. (Noguchi et al., 2007), it was found that the boiling heat transfer coefficients for sulfuric acid solutions are lower than the values estimated using the Nishikawa–Fujita correlation. It was unclear what was related to the decrease in the boiling heat transfer coefficients of the sulfuric acid solutions. This paper presents the experimental results of boiling heat transfer coefficients for sulfuric acid solutions based on newly obtained data in the H_2O molar fraction range from 0.17 to 0.37 (H_2SO_4 mass concentration from 96.5 wt% to 90.4 wt%) at atmospheric pressure as a first step towards determining the boiling heat transfer characteristics for sulfuric acid solutions. The experimental results were compared with empirical correlations for estimation of the boiling heat transfer coefficients for binary mixtures. We then investigated a predictable correlation for the boiling heat transfer coefficient of sulfuric acid solutions in the range of this experiment, and considered factors related to the decrease in the boiling heat transfer coefficients.

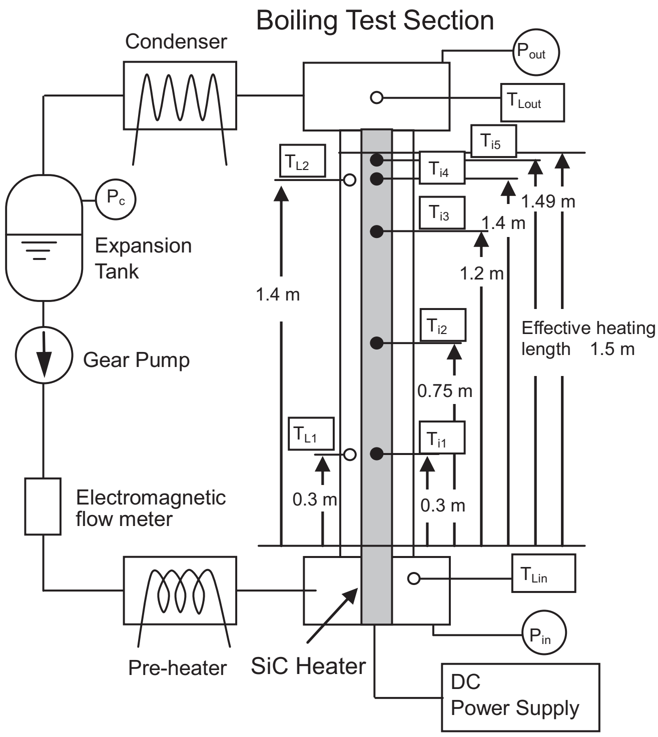

Fig. 1 – Schematic diagram of the experimental apparatus.

2. Experimental apparatus and method

2.1. *Experimental apparatus*

Fig. 1 shows a schematic diagram of the experimental apparatus, which consists of a boiling test section, a pre-heater, a water-cooled condenser, a Teflon coated expansion tank (39 L), a gear pump, and an electromagnetic flow meter. The boiling test section is an annular flow channel composed of a quartz glass tube (φ24 mm in inner diameter) for observing the boiling flow as an outer tube and an electric heater with a SiC sheath (φ9 mm in outer diameter) as an inner tube. The maximum heater power is 3.75 kW, and the effective heating length of the SiC sheath heater is 1.5 m. To prevent heater burn-up due to the rapid temperature rise caused by dryout, the heater power is set up in two steps in the length direction: 80% of the heating power is supplied to the heating length from the inlet of the test section to 1 m in length, and 20% of the heating power is supplied from 1 m to 1.5 m. The heater power is controlled by a DC power source (accuracy ±0.5%). The heater temperature was measured using K-type sheath thermocouples (φ0.5 mm in diameter, accuracy ±1.5 °C) installed inside of the SiC sheath ($T_{i1} - T_{i5}$) in order to estimate the heating surface temperature. The fluid temperature was measured using K-type thermocouples (φ1.6 mm in diameter, accuracy ±1.5 °C) coated with gold (T_{Lin}, T_{Lout}, T_{L1}, and T_{L2}), which has

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