Numerical simulation of HI thermal decomposer for a VHTR helium loop-based SI hydrogen production test facility

Youngjoon Shin*, Taehoon Lee, Juho Lee, Kiyoung Lee, Changkeun Jo, Minhwan Kim
Korea Atomic Energy Research Institute150 Dukjin-dong, Yuseong-gu, Daejeon, Republic of Korea 305-600
** Corresponding author: nyjshin@kaeri.re.kr*

1. Introduction

A VHTR-based method of generating hydrogen from the Sulfur-Iodine (SI) cycle is one of the promising approaches to produce massively hydrogen shown in Fig. 1 [1-4].

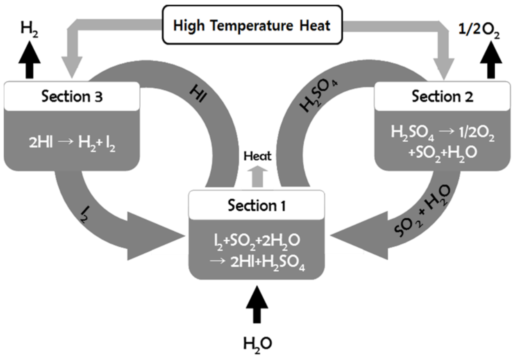

Fig. 1. SI hydrogen production cycle.

Based on the previous study on the experimental performance test of the catalyst-packed type HI thermal decomposer for a 50 NL-H_2/h SI test facility [5], which was directly heated using electrical heating chambers, a HI decomposer for the 1 Nm^3-H_2/h SI test facility coupled to an out-of-pile helium loop (SP-HITD-1000L) has been designed, and it was theoretically confirmed that the design specifications satisfy the hydrogen production capacity based on a Computational Fluid Dynamics (CFD) analysis. The effect of the overall heat transfer coefficient on the helium outlet temperature and decomposition percentage of the decomposer was identified. The HI decomposer proposed is capable of outlet helium temperatures of 383 ^{o}C for an overall heat transfer coefficient of 5 W/m^2-K, which satisfies the operating temperature condition of the out-of-pile helium loop. The average thermal decomposition percentage of the proposed decomposer is 22.4 % for hydrogen iodide. The decomposition percentage obtained from the numerical result is acceptable with a hydrogen production rate of 1 Nm^3-H_2/h.

2. Description of SP-HITD-1000L

The SP-HITD-1000L for the 1 Nm^3-H_2/h scale SI test facility was designed using a common type shell-and-tube heat exchanger shown in Fig. 2. Because the SP-HITD-1000L has a clogging problem owing to the solidification of iodine produced from HI decomposition, the common type shell-and-tube heat exchanger has an advantage over the bayonet-type decomposer in operation and maintenance aspects.

The single tube decomposer was manufactured with a Hastelloy C-276 Alloy tube (52.7 mm-ID, 60.5 mm-OD) consisting of preheating and decomposition regions. The hexagonal tube bundle consisted of 19 Hastelloy C-276 Alloy tubes with a triangular pitch of 75.6 mm. The height of the tube heating region is about 1,500 mm.

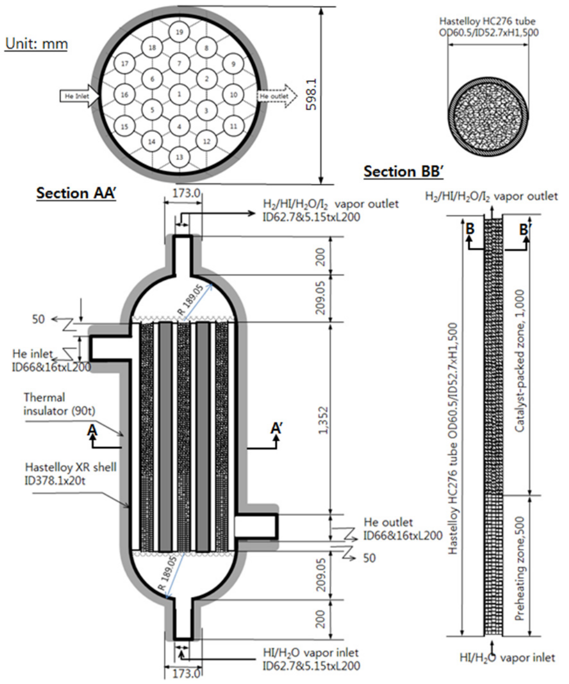

Fig. 2. Schematic of the HI decomposer (SP-HITD-1000L) for the 1 Nm^3-H_2/h scale SI test facility.

The 500 mm lower part is the preheating zone, which is packed with Al_2O_3 Raschig rings (2.2 mm-ID, 6.4 mm-OD, 6.4 mm-L), and the 1,000 mm upper part is the HI catalytic thermal decomposition zone packed with Pt-doped Al_2O_3 Raschig rings. Studies on kinetic modeling and experimental dissociation yields of HI have been done to provide efficient decomposition processes in the SI thermochemical process [6-8].

The hexagonal tube bundle is contained in an Hastelloy XR shell (378.1 mm-ID, 418.1 mm-OD, 1,500 mm-H not including the hemisphere part) with a thermal insulator to reduce heat loss from helium to the external air environment. The helium flow path was designed such that the high-temperature helium discharged from the sulfuric acid decomposer SP-SATD-1000L enters the top-left of the shell and provides thermal energy to the hexagonal tube bundle, and then flows down inside the shell to the outlet nozzle, which is connected at the bottom-right of the shell.

3. Simulation results

A numerical simulation was conducted using a specified helium inlet temperature of 734 ^{o}C, which is the helium outlet temperature discharged from the SP-SATD-1000L in the case of an outer shell of 5 W/m^2-K.

A parameter study was also performed in terms of the overall heat transfer coefficient change as one of the design specifications.

Table 1 summarizes the boundary conditions of the SP-HITD-1000L.

Table 1. Boundary conditions of SP-HITD-1000L

Condition	SP-HITD-1000L
Inlet flow rate of process stream	504 mol-HI/h 1,944 mol-H_2O/h
Outlet flow rate of process stream	414.72 mol-HI/h 44.64 mol-H_2/h 44.64 mol-I_2/h 1,944 mol-H_2O/h
Conversion	18 %
He molar flow rate	13,424.4 mol/h
Inlet temperature of process stream	195 °C
He inlet temperature	734 °C
Atmosphere temperature	25 °C
Operating pressure (tube side/shell side)	5 bar/7 bar

Fig. 3 shows the velocity contours of the cross-sectional and longitudinal planes inside the SP-HITD-1000L at 5 W/m^2-K. The helium velocity displays a maximum velocity of 17 m/s just after discharging from the left upper nozzle and reduced to around 1.5 m/s passing-down through the shell, and then increased again until 5 m/s just before the helium outlet nozzle at the right part of the bottom. The helium path inside the shell has a channeling flow from the upper-left part to the lower-right part, as shown in Fig. 3. In the case of a HI/H_2O gas mixture flow, a circulation region due to the vortex formation is also observed at the bottom disengagement section of the inlet part of the HI/H_2O gas mixture. The circulation phenomena of the HI/H_2O gas mixture provide the reason for an inhomogeneous flow into the tubes.

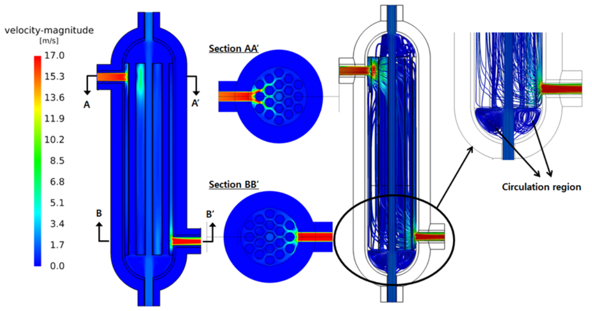

Fig. 3. Velocity contours of a cross-sectional and longitudinal planes inside the SP-HITD-1000L at 5 W/m^2-K.

Fig. 4 shows the flow percentage of HI/H_2O-mixed gas in each Hastelloy C-276 tube at 5 W/m^2-K due to the circulation phenomena of the HI/H_2O gas mixture at the bottom disengagement section of the inlet part. Forty-percent of the HI/H_2O-mixed gas enters through the tube "1" in Fig. 4, and the mole flow rate is 201.6 mol-HI/h. Forty-two percent of the HI/H_2O-mixed gas flows into the six tubes from "2" to "7" located at the second orbital, and the mole flow rate in each tube is 35.28 mol-HI/h. The remaining HI/H_2O-mixed gas is distributed to the 12 tubes from "8" to "19" located at the outside, and the mole flow rate in each tube is 5.04 or 10.08 mol-HI/h. The flow rate in the odd number tubes at the third orbital is 2 times higher than the even number tubes because of wall effect of the bottom disengagement section.

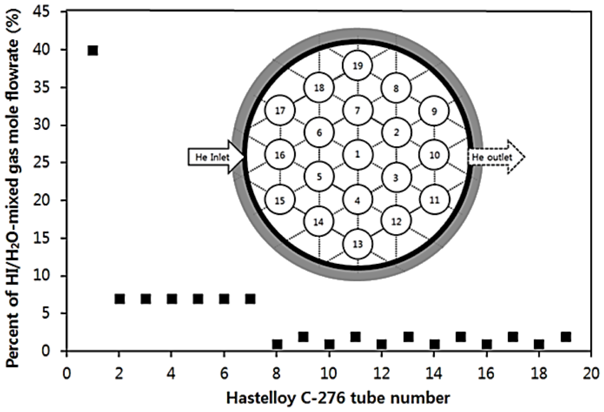

Fig. 4. Flow percentage of HI/H_2O-mixed gas in each tube at 5 W/m^2-K.

Fig. 5 shows the contours of static temperatures inside the SP-HITD-1000L with the overall heat transfer coefficients of the shell covered with a thermal insulator of 5 W/m^2-K and 10 W/m^2-K. The outlet temperatures of helium are 383 °C for the overall heat transfer coefficient of 5 W/m^2-K and 321 °C for the overall heat transfer coefficient of 10 W/m^2-K. When the outlet temperature is 321 °C, heat supply to the H_2SO_4 distillation column is impossible due to the low temperature considering the column reboiler temperature. Therefore, the heat transfer coefficient should be at least smaller than 10 W/m^2-K.

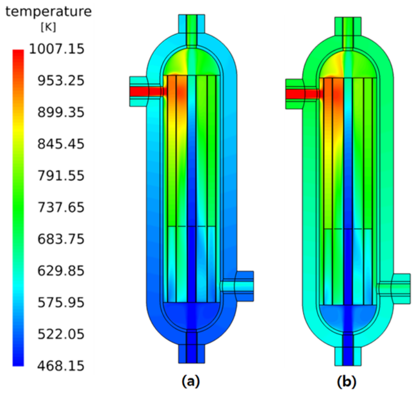

Fig. 5. Temperature contour lines of a longitudinal plane inside the SP-HITD-1000L: (a)10 W/m^2-K and (b) 5 W/m^2-K.

Fig. 6 shows the maximum temperature in the process stream flow path inside the tube at the top in all of the tubes. The HI/H_2O gas mixture in each tube is heated from 195 ^oC to 363 - 678 ^oC at 5 W/m^2-K and 353 - 664 ^oC at 10 W/m^2-K, and flows up to the top of the tube. The helium is cooled from 734 ^oC to 383 ^oC at 5 W/m^2-K and 321 ^oC at 10 W/m^2-K, and flows down to the bottom of the shell. The minimum temperature differences between shell and tube sides are observed just below the top disengagement section. The lowest temperature differences of 15 ^oC at 5 W/m^2-K and 10 ^oC at 10 W/m^2-K between helium and HI stream are revealed at the top of the tube number 10. The maximum-temperature tube in both cases is tube number "16", which is located just in front of the helium inlet nozzle, and that the minimum-temperature tube in both cases is tube number "1", owing to the highest flow rate of the HI/H_2O gas mixture and the endothermic heat of HI dissociation.

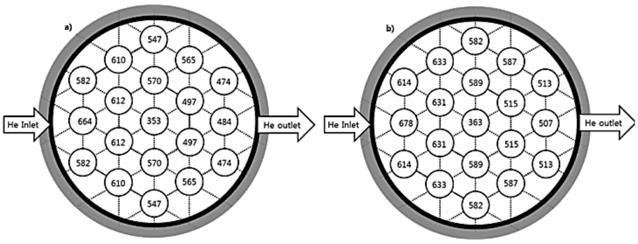

Fig. 6. Maximum temperature (^oC) at the top of each tube of the SP-HITD-1000L: (a) 10 W/m^2-K and (b) 5 W/m^2-K.

In Fig. 7, the equilibrium mole percentage of HI thermal decomposition in each tube was found to be 19.5 to 26.0 % for an overall heat transfer coefficient of 5 W/m^2-K, and 19.1 to 26.0 % for an overall heat transfer coefficient of 10 W/m^2-K. The reason for the difference in the maximum temperature and HI conversion according to the overall heat transfer coefficient is the heat loss from the reactor to the surrounding air through the shell wall.

The average HI thermal decomposition percentages of H_2 and I_2 are 22.4 % for an overall heat transfer coefficient of 5 W/m^2-K, and 22.0 % for an overall heat transfer coefficient of 10 W/m^2-K. These values are equivalent to hydrogen production rates of 1.3 Nm^3-H_2/h and 1.2 Nm^3-H_2/h, respectively. For the long-term closed-loop steady operation of the SI process, the hydrogen production rate of 1.2 to 1.3 Nm^3-H_2/h is unmatched with the H_2SO_4 decomposition rate of 0.9 to 1.0 Nm^3-H_2/h. This problem can be solved by decreasing the feed temperature of the HI/H_2O gas mixture into the SP-HITD-1000L. To achieve a hydrogen productivity of 1 Nm^3-H_2/h between the H_2SO_4 and HI decomposers, an additional cooling system in the HI/H_2O inlet stream is required to decrease the feed temperature of the HI/H_2O gas mixture into the SP-HITD-1000L for example. The inhomogeneous flow as in Fig. 4 is the main reason to bring about the lower decomposition yield of HI compared with equilibrium conversion of 22.4 % and 22.0 % owing to an inefficient heat transfer from helium to HI/H_2O-mixed gas. When improving the inhomogeneous flow rate of the HI/H_2O gas mixture in the 19 tubes to a homogeneous flow rate, the average decomposition percentage of the HI in the SP-HITD-1000L can be increased to 24.1 % and 23.7 %, respectively.

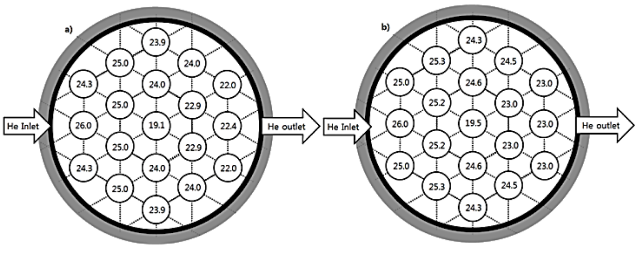

Fig. 7. Equilibrium conversion (%) of hydrogen iodide decomposition in each tube of the SP-HITD-1000L: (a) 10 W/m^2-K and (b) 5 W/m^2-K.

4. Conclusions

A creative semi-pilot scale HI thermal decomposer (SP-HITD-1000L) for a 1 Nm^3-H_2/h SI test facility incorporating a heat transfer device through coupling with the out-of-pile helium loop was newly designed. A numerical analysis was carried out using Computational Fluid Dynamics (CFD) software to simulate and analyze the helium flow pattern, temperature distribution, and thermal decomposition percentages of HI. The work highlighted the effect of the overall heat transfer coefficient on the helium outlet temperatures, maximum temperature in the reactor tubes, and the decomposition percentages. Outlet helium temperatures of the HI decomposer was 383 ^oC for an overall heat transfer coefficient of 5 W/m^2-K, which satisfy the operating temperature conditions of the out-of-pile helium loop. The average thermal decomposition percentage of the proposed decomposer was shown to be 22 % for HI in the case of an overall heat transfer coefficient of 5 W/m^2-K. In terms of the HI decomposition performance excluding the integrated operation of the SI process, when improving the inhomogeneous flow rate of the HI/H_2O gas mixture in the 19 Hastelloy C-276 tubes to a homogeneous flow rate, it was suggested that the average decomposition percentage of HI is increased until 24 % at the same temperature profile as the inhomogeneous flow case., which is equivalent to the hydrogen productivity of 1.4 Nm^3-H_2/h. It is expected that the novel design and CFD results of the SP-HITD-1000L including the out-of-pile helium loop investigated in this study will be provided to construct a semi-pilot scale SI test facility in Korea. To improve the performance of the decomposer, a minimization of heat loss into the atmosphere and optimization of component design are also needed.

Acknowledgments

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REFERENCES

[1] G. E. Besenbruch, L. C. Brown, D. R. O'Keefe, and C. L. Allen, "Thermochemical Water-splitting Cycle, Bench-scale Investigations, and Process Engineering", DOE/ET/26225-1, General Atomic Company; 1982.
[2] S. Kasahara, S. Kubo, K. Onuki, and M. Nomura, "Thermal Efficiency Evaluation of HI Synthesis/ Concentration Procedures in the Thermochemical Water Splitting IS Process", International Journal of Hydrogen Energy, Vol. 29, p. 579, 2004.
[3] S. Goldstein, J. M. Borgard, and X. Vitart, "Upper Bound and Best Estimate of the Efficiency of the Iodine Sulfur Cycle", International Journal of Hydrogen Energy, Vol. 30, p. 619, 2005.
[4] P. Zhang, S. Z. Chen, L. J. Wang, T. Y. Yao, and J. M. Xu, "Study on a Lab-scale Hydrogen Production by Closed Cycle Thermo-chemical Iodine-sulfur Process", International Journal of Hydrogen Energy, Vol. 35, p. 10166, 2010.
[5] K. Kang, C. Kim, J. Kim, W. Cho, S. Jeong, and C. Park, "Hydrogen Production by SI Process, with Electrolysis Stack Embedded in HI Decomposition Section", International Journal of Hydrogen Energy, Vol. 41, p. 4560, 2016.
[6] T. Nguyen, Y. Gho, W. Cho, K. Kang, S. Jeong, and C. Kim, "Kinetics and Medeling of Hydrogen Iodide Decomposition for a Bench-scale Sulfur-iodine Cycle", Applied Energy, Vol. 115, p. 531, 2014.
[7] G. Hwang and K. Onuki, "Simulation Study on the Catalytic Decomposition of Hydrogen Iodide in a Membrane Reactor with a Silica Membrane for the Thermochemical Water-splitting IS Pprocess", Journal of Membrane Science, Vol. 194, p. 207, 2001.
[8] N. Goswami, K. Singh, S. Kar, R. Bindal, and P. Tewari, "Numerical Simulations of HI Decomposition in Packed Bed Membrane Reactors", International Journal of Hydrogen Energy, Vol. 39, p. 18182, 2014.