# Numerical simulation of HI thermal decomposer for a bench-scale nuclear hydrogen production test facility

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## 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.

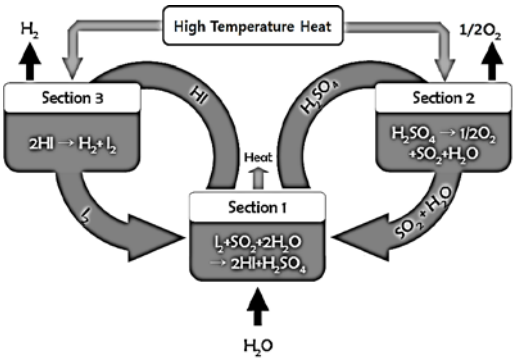


Fig. 1. SI hydrogen production cycle.

The SI cycle offers the potential of highly efficient production of hydrogen from water, but most studies have only been concerned with evaluating its thermal efficiency [1-4] and small scale atmospheric operation test of the SI process [5]. A study was done by Yamawaki et al. on application of nuclear energy to the SI cycle for hydrogen production [6].

The current technical issue of the SI process is to reach steady state operation and then achieve long-term closed-loop operation of the SI integrated test facility without any additional feeding of recycled reagents such as iodine and sulfur dioxide other than water. In this case, the molar ratio of oxygen and hydrogen generated from Section 2 and 3 in Fig. 1 is 0.5/1 which should be maintained. A study on the start-up and static behaviors of a $H_2SO_4$ thermal decomposer and HI thermal decomposer, which are key components in Sections 2 and 3, is thus important.

This study focused on the HI thermal decomposer (HITD-50L), which is the key component of the bench-scale SI test facility. The HITD-50L was designed by the Korea Institute of Energy Research (KIER). Based on the design specification and test operation results, numerical calculations for the HITD-50L were done to evaluate the start-up and static behaviors. The numerical simulation was performed at the operational condition to produce 50 NL/h of hydrogen, based on the mass balance of the SI integrated process. The results are presented to confirm and recommend practical design specifications and operating conditions for the component to achieve a hydrogen productivity of 50 NL/h.

## 2. Description of HITD-50L and operating condition

The HITD-50L for the 50 NL-$H_2$/h scale SI test facility was designed by KIER in Korea shown in Fig. 2. The decomposer, HITD-50L, was manufactured with a Hastelloy C-276 Alloy tube (ID 52.7 mm/OD 60.5 mm) consisting of preheating and decomposition regions. The height of the electrical heating region is about 1,500 mm. The front part of 480 mm is the preheating zone which is packed with $Al_2O_3$ Raschig rings, and the back part of 1,020 mm is the HI catalytic thermal decomposition zone packed with Pt-doped $Al_2O_3$ Raschig rings. The decomposer is uniformly heated by a 9 kW electrical heater, and a constant operation temperature is automatically maintained using the PID control mode with a temperature measuring element (TME) shown in Fig. 2. The HITD-50L is initially filled with nitrogen gas, which is thermally equilibrated with the heating compartment temperature under 5 bar. The start-up operation of the decomposer is initiated by opening a valve connected with the outlet-pipe line of the partial condenser of the $HI_x$ distillation column that is situated at the head of the HITD-50L in the SI process. Table 1 shows the design and operation parameters of the HITD-50L.

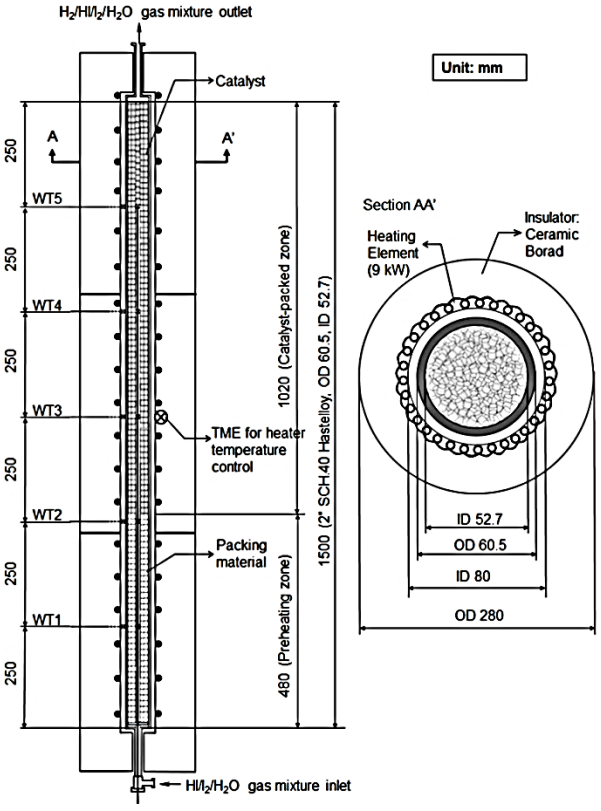


Fig. 2. Schematic of the HITD-50L.

Table 1. Design and operation parameters of HITD-50L

| | |
|---|---|
| ***Structure material*** | ***Hastelloy HC276 (ID52.7/OD60.5mm×L1500mm)*** |
| ***Catalyst*** | ***Pt-doped(1wt%) $Al_2O_3$ Raschig ring*** |
| ***Packing in preheating zone*** | ***$Al_2O_3$ Raschig ring(L/D=2)*** |
| ***Heating element*** | ***Kanthal A-1 (9kW)*** |
| ***Operation pressure*** | ***5 bar*** |
| ***Inlet vapor mole flow rate*** | ***97.2mol_$H_2O$/h, 25.2mol_HI/h*** |
| ***Inlet vapor temperature*** | ***195°C*** |

## 3. Simulation results

The start-up behavior of the HITD-50L for the 50 NL-$H_2$/h scale SI test facility was calculated by applying the decomposer design and operation parameters summarized in Table 1, with the temperature boundary conditions taken from the real measurements of the thermocouples of WT1 to WT5 in Fig. 2 at a constant operation temperature of the electrical heater. These outside wall temperatures of the HITD-50L measured from the 50 NL-$H_2$/h scale SI test facility at KIER are listed in Table 2.

Table 2. Outside wall temperatures of the HITD-50L measured by thermocouples

| ***Heater Temp.*** / ***Thermocouple location*** | ***450 °C*** | ***500 °C*** | ***550 °C*** |
|---|---|---|---|
| ***WT1*** | ***321 °C*** | ***367 °C*** | ***408 °C*** |
| ***WT2*** | ***343 °C*** | ***388 °C*** | ***436 °C*** |
| ***WT3*** | ***455 °C*** | ***504 °C*** | ***556 °C*** |
| ***WT4*** | ***432 °C*** | ***469 °C*** | ***508 °C*** |
| ***WT5*** | ***340 °C*** | ***390 °C*** | ***430 °C*** |

When the temperature boundary values were implemented in the start-up dynamic simulation of the HITD-50L, the polynomial curves along the height of the HITD-50L in Fig. 3 were applied.

As shown in Fig. 3, both end parts of the HITD-50L have some heat loss, and some effects of the radiation heat are observed around the middle position of the HITD-50L. As a result, the temperature of the end part of the HITD-50L is lower than the constant electrical heater temperature, and a higher temperature of 4 to 6 °C rather than the constant electrical heater temperature is measured at the middle position of the HITD-50L from the KIER experiment.

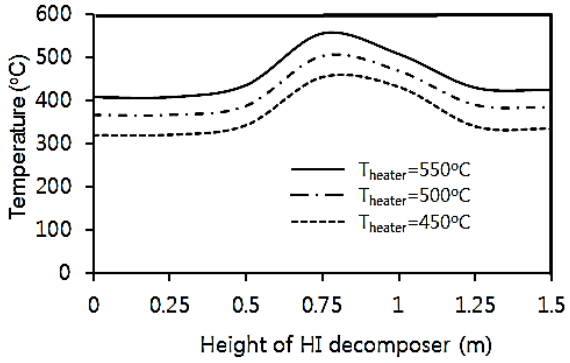


Fig. 3. Temperature boundary curves taken from real measurements of the thermocouples of WT1 to WT5 in the HITD-50L at various electrical heater temperatures.

Fig. 4 shows the temperature of the electrical heater (dotted line) and the accumulation value of the hydrogen discharged from the HITD-50L (solid line) as a function of the operation time obtained from the preliminary performance test of the HITD-50L by KIER.

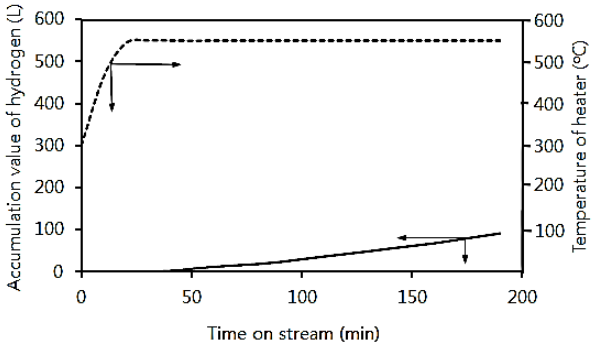


Fig. 4. Temperature of the electrical heater (dotted line) and accumulation value of the hydrogen production (solid line) measured through the preliminary performance test of the HITD-50L.

The test result reveals that the temperature of the electrical heater changed from the initial operation temperature of 300 °C to the steady state value of 550 °C within 30 minutes, where the initial operation temperature means that the temperature of the electrical heater was measured when opening a valve connected with the outlet pipe line of the partial condenser of the $HI_x$ distillation column, which is situated at the head of the HITD-50L in the SI process. The KIER experimental group measured the hydrogen productivity using a wet test meter every 10 seconds, and then plotted the accumulation value of hydrogen shown in Fig. 4. Based on these accumulation values, the hydrogen production rates can be obtained from the slope of the accumulation curve shown in Fig. 5. Although the experimental value of the hydrogen production rate has a fluctuating pattern due to the on/off actuation of the back-pressure regulator to maintain a constant operation pressure of 5 bar in the HITD-50L, steady state values of about 40 NL-$H_2$/h were approximately measured at the operation temperature of 550 °C by the electrical heater in the HITD-50L. The start-up behaviors of the HITD-50L with and without the induction period of HI decomposition were calculated.

As shown in Fig. 5, when an induction period of 12 minutes is used in the numerical simulation, the calculation results agree well with the experimental data when the pre-exponential factors of $A_{HI}$=2.0 and $A_{I2}$=8.1x$10^{-11}$ are applied. However, the simulation results calculated without the induction period in the HI decomposition have some deviation from the experimental values during the start-up operation of the HITD-50L.

Numerical simulations have also been done with the specified operation temperatures of the electrical heater.

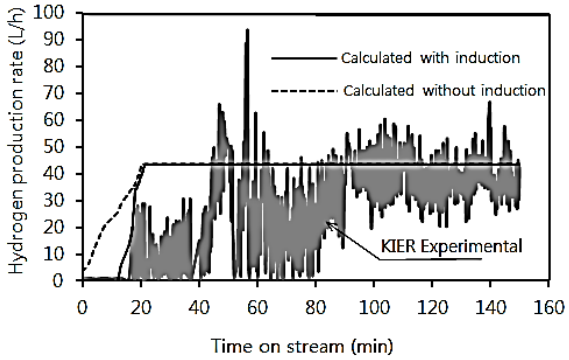


Fig. 5. Comparison of the experimental hydrogen production rate of the HITD-50L and start-up dynamic simulation results with and without the HI decomposition induction period at an electrical heater temperature of 550°C.

Fig. 6 shows the steady state temperature profile inside the HI decomposer along the column height at each constant operation temperature of 450 °C, 500 °C, and 550 °C, respectively. The temperature distribution is similar and increases along the operation temperature of the electrical heater.

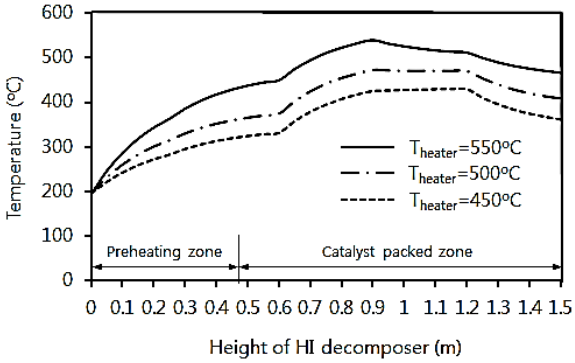


Fig. 6. Predicted steady state temperature profiles of the process streams in the HITD-50L at various electrical heater temperatures.

Fig. 7 shows the calculation results of the HI decomposition fraction along the height of the HI decomposer at each constant operation temperature of the electrical heater. The decomposition percentage of HI at the exit of the HITD-50L obtained from the numerical simulation is 8.5 % at 450 °C, 11.5 % at 500 °C, and 15.4 % at 550 °C. Compared with the equilibrium conversion, these values are lower than the equilibrium conversions and should be improved through parametric studies, such as a catalyst shape, void fraction, and HI vapor volumetric flow rate.

From the inlet HI mole flow rate condition shown in Table 1 and these decomposition percentages obtained by the numerical simulation, we can estimate the hydrogen production rate as a function of the constant operation temperature of the electrical heater, which is one of the key operational variables.

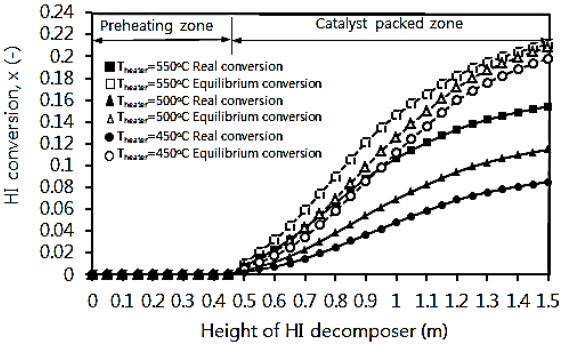


Fig. 7. Predicted steady state HI conversion profiles in the HITD-50L at various electrical heater temperatures.

The effect of the operation temperature of the electrical heater on the hydrogen production rate is shown in Fig. 8.

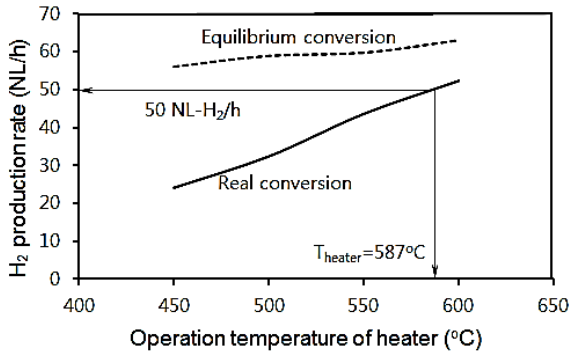


Fig. 8. Effect of the electrical heater temperature on the hydrogen production rate in the HITD-50L at feed flow rates of 97.2mol-$H_2O$/h and 25.2mol-HI/h.

As a result of the parametric study, it was anticipated that a hydrogen production rate of 50 NL-$H_2$/h in the HITD-50L can be obtained at a constant operation temperature of 587°C by the electrical heater.

## 4. Conclusions

Numerical calculations to investigate the start-up and static behaviors of the HI thermal decomposer (HITD-50L) for the 50 NL-$H_2$/h SI test facility were carried out. The simulations show that the design and operation parameters of the HITD-50L proposed in this study are reasonable to achieve a hydrogen production rate of 50 NL/h. Regarding the computer simulation of the HITD-50L, the present research covers the heat transfer and chemical decomposition of HI for an $Al_2O_3$ Raschig ring and Pt-doped $Al_2O_3$ Raschig ring-packed tubular type decomposer. The simulation results obtained from this research are consistent with the experimental results found by KIER with a hydrogen production rate of around 40 NL-$H_2$/h for an operation temperature of 550 °C. A parametric study was conducted with different operation temperatures for the electrical heater from 450 °C to 600 °C, and the decomposition percentages of HI were surveyed. From the parametric study results, it was found that an operation temperature

of 587ºC for the electrical heater satisfies a hydrogen productivity of 50 NL-$H_2$/h for the 50 NL-$H_2$/h SI test facility.

**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] Y. Shin, K. Lee, Y. Kim, J. Chang, W. Cho, and K. Bae, "A Sulfur-Iodine Flowsheet Using Precipitation, Electrodialysis, and Membrane Separation to Produce Hydrogen", International Journal of Hydrogen Energy, Vol. 37, p. 16604, 2012.

[5] 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.

[6] M. Yamawaki, T. Nishihara, Y. Inagaki, K. Minato, H. Oigawa, K. Onuki, R. Hino, and M. Ogawa, "Application of Nuclear Energy for Environmentally Friendly Hydrogen Generation. International Journal of Hydrogen Energy", Vol. 32, p. 2719, 2007.