VHTR-based Nuclear Hydrogen Plant Analysis for Hydrogen Production with SI, HyS, and HTSE Facilities

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1. Introduction

In this article, a VHTR-based method of generating hydrogen from Sulfur-Iodine (SI), Hybrid Sulfur (HyS), and High Temperature Steam Electrolysis (HTSE) processes [1-3] is evaluated. An analysis is performed for nuclear hydrogen plants with different hydrogen production methods including coupling technologies. VHTR-based hydrogen production plants constitute an attractive option for massive production of hydrogen without any greenhouse gas emission. Such processes allow recycling of chemicals internally within the process and only using water as a raw feeding material. The hydrogen production process requires process heat at high temperature and electricity to generate hydrogen. These are important parameters to decide hydrogen production efficiency and cost.

In this paper, analyses of material and heat balances on the SI, HyS, and HTSE processes coupled to a Very High Temperature gas-cooled Reactor (VHTR) were performed. The hydrogen production efficiency including the thermal to electric energy ratio demanded from each process is found and the normalized evaluation results obtained from three processes are compared to each other. The currently technological issues to maintain the long term continuous operation of each process will be discussed at the conference site.

2. Outline of VHTR-hydrogen coupling system

As shown in Fig. 1, nuclear hydrogen production facilities consist of the VHTR, intermediate heat exchanger (IHX), and hydrogen production plant (HPP).
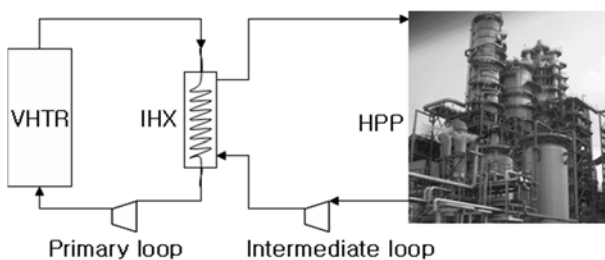

Fig. 1. Directly heating loop between IHX and a hydrogen production plant.

In order to prevent the propagation of the thermal disturbance owing to the abnormal operation of the hydrogen production process from the IHX to the VHTR, a cooling system for the secondary helium of the IHX is required [4]. Based on the recommended IHX secondary helium pathway, Fig. 2 shows the conceptual flow diagram of the IHX-SI coupling system. The IHX-SI coupling system consists of the IHX, helium cooling system, helium circulator, and SI process components such as SO_3 and H_2SO_4

decomposers, H_2SO_4 vaporizer, and HI decomposer including a HI pre-heater (HE301). The helium cooling system will only be operated at abnormal operation of the IHX-SI coupling system. Under normal operation, the secondary helium discharged from the IHX provides heat to the process components in a regular sequence, and the cooled helium passing through He12 is compressed by the helium circulator and recycled to the IHX for reheating.
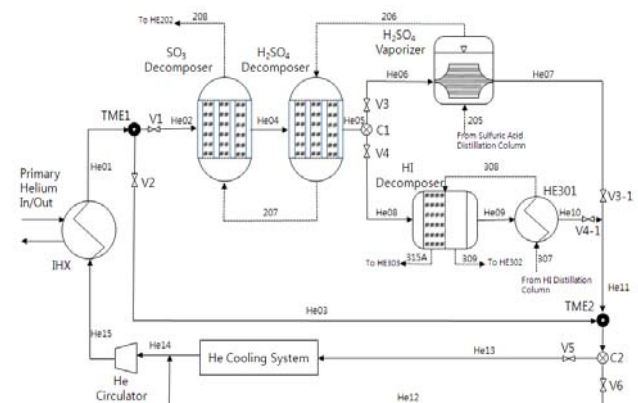

Fig. 2. Conceptual flow diagram of the IHX-SI coupling system.

The temperature fluctuation of the secondary helium due to the abnormal operation of the hydrogen production process is then calculated based on the proposed coupling system model. Finally, the conceptual design of the cooling system for the secondary helium of the IHX with a steam generator and forced-draft air-cooled heat exchanger to mitigate the thermal disturbance can be prepared as shown in Fig. 3.
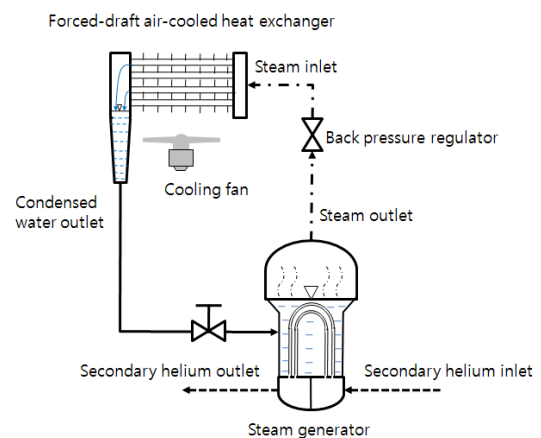

Fig. 3. Conceptual flow diagram of the helium cooling system.

3. SI hydrogen production process

The flowsheet of an electrodialysis cell (EDC) and membrane reactor (MR)-embedded SI cycle is shown in Fig. 4 [5].
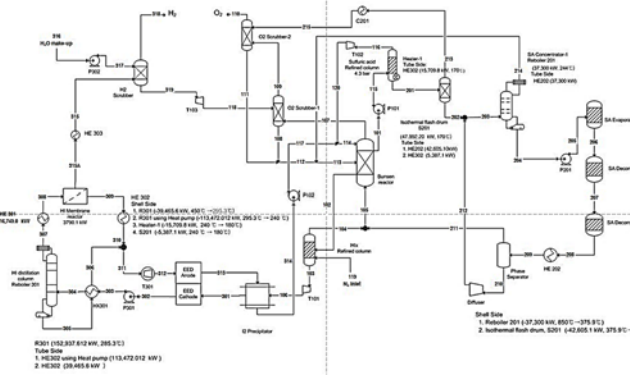

Fig. 4. Flowsheet of SI hydrogen production process.

The key components consisting of the flowsheet are as follows : a Bunsen reactor having a mutual separation function of sulfuric acid and hydriodic acid phases, a sulfuric acid-refined column for the purification of a sulfuric acid solution, a HI_x-refined column for the purification of a hydriodic acid solution, an isothermal drum coupled to a multi-stage distillation column to concentrate the sulfuric acid solution, a sulfuric acid evaporator, a sulfuric acid decomposer, a sulfur trioxide decomposer, a sulfuric acid recombination reactor, a condensed sulfuric acid solution and sulfur dioxide/oxygen gas mixture separator, a precipitator to recover excess iodine dissolved into the hydriodic acid solution, an electrodialysis cell to break through the azeotrope of a $HI/I_2/H_2O$ ternary aqueous solution, a multi-stage distillation column to generate the highly concentrated hydriodic acid vapor as a top product of the column, a membrane reactor to decompose hydrogen iodide and preferentially separate hydrogen, and a hydrogen scrubber. The material and energy balance on each component was established by a computer code simulation using Aspen Plus^{TM} . The optimized energy balance of the SI process to produce 300 mol/s is shown in Table 1.

Table 1. Energy balance of SI process

Unit No.	Description	Thermal energy (kW$_t$)	Electric energy (kW$_e$)
HE 301		16,749.000	
MR	HI membrane reactor	3790.10	
R301	HI distillation column	152,937.612	
HE302	heat transfer to R301	-39,465.600	
	heat transfer to R301 with heat pump	-113,472.012	
	heat transfer to HEATER-1	-15,709.800	
	heat transfer to S201	-5,387.100	
MP-MC	Multistage compressor net work	36,364.509	17,454.964
P301	pump	8,533.648	4,096.15
P302	pump	58.696	28.17
T301	turbine	-5,404.917	-2,594
EDC	Electrodialysis cell at 0.112 V	16,572.553	7,954.826
P201	pump	228.43	109.64
Diffuser	turbine	-59.08	-28.36
HEATER-1	Sulfuric acid refined column	15,709.800	
S201	Isothermal flash	47,992.200	
R201	Sulfuric acid concentrator	37299.90	
SA section		140,800.20	
HE202	Heat transfer to R201	-37,299.90	
	Heat transfer to S201	-42,605.10	
P101 &102	pump	773.14	371.11
T101&102&103	turbine	-487.26	-233.88
Total Heat duty		217,919.024	
Efficiency	39.4 % in the case of heat-to-electric conversion efficiency=48%		

The hydrogen production thermal efficiency for this heat network, based on the higher heating value of the produced hydrogen, is anticipated by 39.4 %.

4. HyS hydrogen production process

The key components consisting of the HyS hydrogen production process shown in Fig. 5 are a SO_2-depolarized electrolyzer, two multi-stage distillation columns to concentrate a sulfuric acid solution, a sulfuric acid evaporator, a sulfuric acid decomposer, a sulfur trioxide decomposer, a sulfuric acid recombination reactor, a condensed sulfuric acid solution and sulfur dioxide/oxygen gas mixture separator, a SO_2 absorber, a SO_2 desorber, a SO_2 dissolver, and a hydrogen purifier [6].
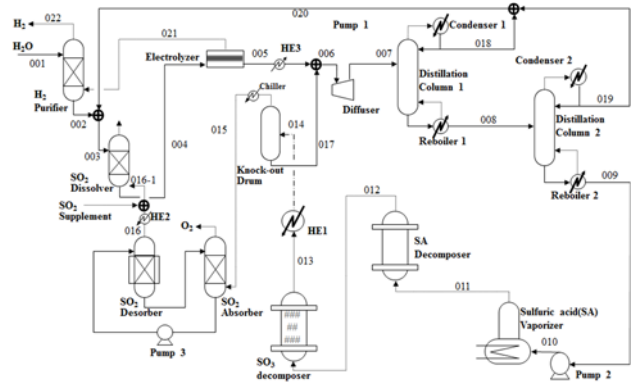

Fig. 5. Flowsheet of HyS hydrogen production process.

The material and energy balance of each component was established by using the KAERI DySCo computer code with Excel sheet calculations partly. The optimized energy balance to produce 300 mol/s is shown in Table 2. The maximum hydrogen production thermal efficiency is anticipated by 39 %.

Table 2. Energy balance of HyS process

Equipment	Electricity (kW$_e$)	Thermal (kW$_t$)	Remarks
Electrolyzer	34,618.1		$V_{applied}$=0.568VDC(I/A=200 mA/cm^2) & η_{acid_e}=0.95
Pump 1	16.2		
Pump 2	18.3		
Pump 3	3.5		Assumption Head; 5m
Diffuser	-26.7		Hydraulic turbine efficiency; 0.9
Reboiler 1		59,640.0	Tube side; HE1(by gas phase sensible heat, heat of formation of H_2SO_4, and latent heats of 98 wt% H_2SO_4 (49,646.9 kJ/s; 638.9°C→185°C) and by using a heat pump(9993.1kJ/s; 185°C→165°C(H_2O BP at 7 bar)), 86.55% of Line # 013 stream)
Heat pump	114.1		COP = 87.6
Reboiler 2		26,000.0	Tube side; HE1(Input 850°C/Output 638.9°C, 100% stream of Line # 013)
H_2SO_4 vaporizer		75,281.4	Heated by He
H_2SO_4 decomposer		38,015.3	Heated by He
SO_2 decomposer		34,881.8	Heated by He
SO_2 desorber		14,977.8	-Primary heating(1,189.2 kJ/s; 40°C→48.6°C) by the sensible heat of uncondensed gas stream from the Reboiler 1 tube side(Input 165°C/Output 94°C) -Secondary heating(13,788.6 kJ/s; 48.6°C→130°C) by the gas phase sensible heat and the latent heats of H_2SO_4 and H_2O of HE1 tube side stream(Input 638.9°C/Output 165°C(H_2O BP at 7 bar), 13.45% of Line # 013 stream)
HE1		-26,000.0 -59,640.0 -14,977.8	Shell side; Reboiler 2(SS Operation Temp 240°C) Shell side; Reboiler 1(SS Operation Temp 170°C) Shell side; SO_2 desorber(Heating from 40°C to 130°C)
HE2		-1,130.4	Shell side; HE3
HE3		1,130.4	Tube side; HE2
Total	34,743.5	148,178.5	η_{eff}=0.39 at $\eta_{electricity}$=0.48 η_{eff}=0.35 at $\eta_{electricity}$=0.35

5. HTSE hydrogen production process

The flowsheet of a HTSE process coupled to VHTR to produce hydrogen is shown in Fig. 6 and evaluated by using the Hyprotech's HYSYS computer code [7].

From the flowsheet evaluation of the HTSE process coupled with VHTR based on the hydrogen production rate of 300 mol/s, thermal neutral voltage has been 1.2454 ~ 1.4 V from the computational fluid dynamic (CFD) analysis of a steam electrolyzer based on the experimental result as well as the structure of SOEC proposed by INL in US. When each voltage of 1.4 V and 1.2454 V is applied, the average value of the hydrogen production with an electrolysis cell of 8 cm x 8 cm is approximately 4.28 NL/h and 2.89 NL/h, respectively.
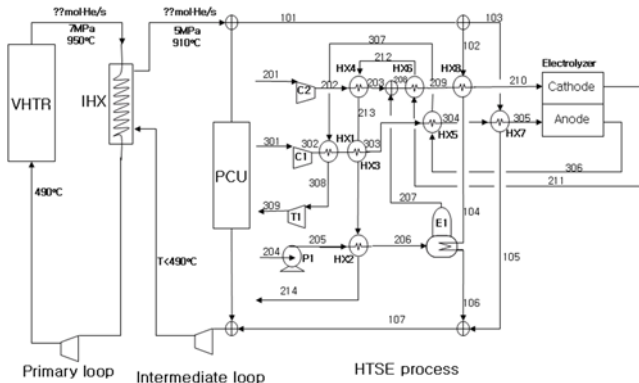

Fig. 6. Flowsheet of HTSE hydrogen production process.

The electrolysis cell of the number of 5,652,912 and 8,365,868 is required to produce the hydrogen of 300 mol/s. And then, the electric consumption is 81,043 kJ/s and 72,094 kJ/s corresponding to the thermal energy of 177,726 kJ/s and 158,101 kJ/s with the PCU efficiency of 0.48 and AC/DC conversion efficiency of 0.95, shown in Table 3.

Based on the higher heating value (HHV) of hydrogen, the hydrogen production efficiencies of 45.2 % and 50.4 % are anticipated at 1.4 V and 1.2454 V applied at the single cell of 8x8 cm^2, respectively.

Table 3. Energy balance of HTSE process

Equip. #	Equipment	Energy duty(kJ/s)	Remarks
E1	Steam generator	9800.0	From He
HX1	Recuperator	263.0	-263.0 kJ/s from Line #307-308
HX2	Recuperator	5270.0	-5270.0 kJ/s from Line #213-214
HX4	Recuperator	63.4	-63.4 kJ/s from Line #212-213
HX5	Recuperator	1624.3	-1624.3 kJ/s from Line #306-307
HX6	Recuperator	3070.0	-3070.0 kJ/s from Line #211-212
HX7	Heat exchanger	79.0	From He
HX8	Heat exchanger	4620.0	From He
Total thermal energy		14499.0	
C1	Air compressor	502.4	Calculation base of η_{eff}=0.75
C2	Hydrogen compressor	85.5	Calculation base of η_{eff}=0.75
P1	Pump	30.0	Calculation base of η_{eff}=0.90
T1	Turbine	-3238.0	Calculation base of η_{eff}=0.65
Electrolyzer	Electrolyzer	158101 at 1.2454 V 177726 at 1.4 V	Calculation base of η_{eff}=0.48
Total electrical energy		155481 175106	
Total energy demanded		169980 189605	

6. Conclusions

VHTR-based nuclear hydrogen plant analysis for hydrogen production with SI, HyS, and HTSE facilities has been carried out to determine the thermal efficiency.

In the optimal flowsheet and operating condition of each process, the highest thermal efficiency of 45.2% is revealed by the VHTR-based HTSE hydrogen production facilities, when the electrical energy demanded from the process is supplied from the Brayton cycle of η_{elec}=0.48. However, when the electrical energy is supplied from the Rankin cycle of η_{elec}=0.35, the hydrogen production thermal efficiency is decreased until 33.5%, which is lower than the SI process of 35.9%. It is evident that the thermal to electrical energy ratio demanded from each hydrogen production process is an important parameter to select the adequate process for hydrogen production.

To improve the hydrogen production efficiency in the SI process coupled to the VHTR without electrical power generation, the demand of electrical energy in the SI process should be minimized by eliminating an electrodialysis step to break through the azeotrope of the HI/I$_2$/H$_2$O ternary aqueous solution.

REFERENCES

[1] J. H. Norman, 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] Westinghouse Electric Corporation, "A Study on the Electrolysis of Sulfur Dioxide and Water for the Sulfur Cycle Hydrogen Production Process", AESD-TIME-3043, Final report prepared for the JET Propulsion Laboratory Contract No. 955380; 1980.
[3] J. S. Herring, P. Lessing, J. E. O'Brien, C. Stoots, J. Hartvigsen, and S. Elangovan, "Hydrogen Production through High-Temperature Electrolysis in a Solid Oxide Cell", 2nd Information Exchange Meeting on Nuclear Production of Hydrogen, Argonne National Laboratory, Illinois, USA, 2 and 3 October 2003.
[4] Y. Shin, J. Chang, T. Lee, K. Lee, and Y. Kim, "A Cooling System for the Secondary Helium Loop in the VHTR-based SI Hydrogen Production Facilities", International Journal of Hydrogen Energy, Vol. 38, p. 6182, 2013.
[5] 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.
[6] Y. Shin, H. Shin, K. Lee, J. Chang, and Y. Kim, "A Hybrid-Sulfur Flowsheet Using an Ionic Liquid Absorbent to Separate Sulfur Dioxide from Oxygen", Energy Procedia, Vol. 29, p. 576, 2012.
[7] Y. Shin, W. Park, J. Chang, and J. Park, "Evaluation of the High Temperature Electrolysis of Steam to Produce Hydrogen", International Journal of Hydrogen Energy, Vol. 32, p. 1486, 2007.