A High Pressure and High Temperature Sulfuric Acid Experimental System

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1. Introduction

The Very High Temperature Reactor (VHTR) heat is transferred to a thermo-chemical hydrogen production process through an intermediate loop. Since nuclear hydrogen coupled components such as a SO_3 decomposer and a sulfuric acid evaporator will operate at a at high pressure (~2.0MPa) and high temperature (~900^oC) and a large mechanical stress with a corrosive H_2SO_4 and SO_3 environment, the development of nuclear hydrogen coupled components is one of the current unsolved issues in nuclear hydrogen generation. Most of the sulfuric acid loops have an operational difficulty at a high pressure and high temperature condition because of the harsh corrosion characteristic of the high temperature sulfuric acid. Fragile quartz is an excellent component material at low pressure sulfuric acid loop but quartz is not allowed to use beyond the pressure of 0.3MPa at high temperature. We designed and constructed a small scale sulfuric acid experimental system which can simulate a part of the hydrogen production module of a Nuclear Hydrogen Development and Demonstration (NHDD) reactor. The main usage the loop is to screen and verify the candidates for the nuclear hydrogen coupled components. This system will be connected to a primary loop of a VHTR simulated small scale gas loop in the Korea Atomic Energy Research Institute [2].

2. Design analyses

2.1 Sulfuric acid loop

A sulfuric acid (H_2SO_4) loop is an open loop and consists of a H_2SO_4 storage tank, a H_2SO_4 feed pump, a pre-heater, a heat exchanger (evaporator), a PHE, a separator, a SO_2 trap, and a H_2SO_4 collector (Figure 1). Cold 98% H_2SO_4 is superheated to 500°C. In a superheating process, H_2SO_4 decomposes into H_2SO_4 and SO_3. In the PHE, some fraction of the SO_3 is dissolved into SO_3 and O_2. The toxic SO_3 is separated in the separator and the sulfur dioxide in the mixture gas is removed in the NaOH trapping system. The design condition of the secondary loop is as follows;

- o Working Fluid — Sulfuric acid
- o Design Temperature — 950 °C
- o Design Pressure — 1.0 MPa.
- o Design Flow — 1.0 kg/min

2.2 Sulfuric acid super-heating system

A Sulfuric acid super-heating system is composed of two components; a sulfuric acid pre-heater and sulfuric acid super-heater. The pre-heater composed of corrosion free Teflon lined inlet plenum and SiC tube flow path (Figure 2). But the material of outlet plenum is selected to Hastelloy C276 which has good corrosion resistance in SO_3 environments. The heat is induced by indirect heating of Kantal coil heater on the SiC tube surface.

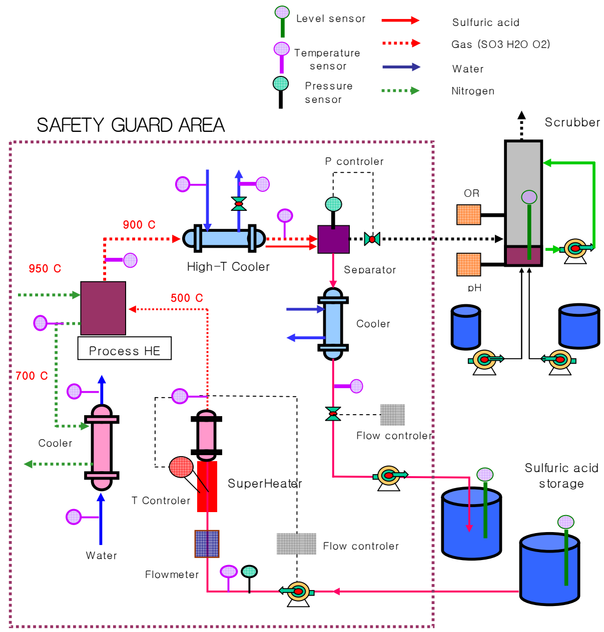


Figure 1 Schematics of a sulfuric acid loop

The SiC tube surface is electrically insulated by quartz tube. The heater has a maximum of the 8kW power and can withstand a fluid pressure up to 1.0MPa.The super-heater shape is same with pre-heater's except Hastelloy C276 inlet plenum and high-temperature sealing. The super-heater heats the mixture gas (sulfuric acid decomposed to water and SO_3) up to 500°C by using a maximum power of 8kW.

2.3 T/H analysis

A convection heat transfer in a tube has a relatively simple relation if the flow condition is laminar. For a low enriched sulfuric acid, the following heat-transfer coefficient is recommended [3].

$$h_{SK} = \frac{h_{ID}}{B}$$

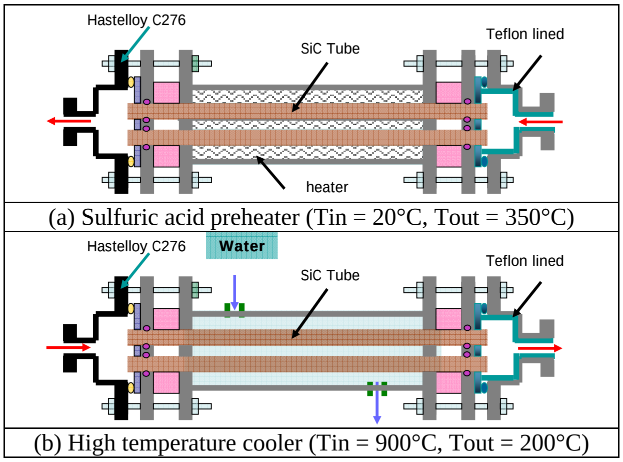
Where, $h_{ID} = 1/\left(x_1/h_1 + x_2/h_2\right)$,

$$B = \left(1 + A_0\left|y_1 - x_1\right|0.88 + 0.12P\right)$$

x_1, y_1: mole fraction of liquid water and steam
x_2: mole fraction of liquid H_2SO_4
h_1: HT coef. of water, P: Pressure

2.4 Design requirements

The maximum operating temperature of the SiC heater elements is over 1500°C, but we limited the maximum operating temperature of the heaters in the components design to lower than 1200°C by considering a sufficient enough operation



(a) Sulfuric acid preheater (Tin = 20°C, Tout = 350°C)
(b) High temperature cooler (Tin = 900°C, Tout = 200°C)

Figure 2. Layout of the main components

margin. Super-alloy C276 is adopted for the vessel of the super-heater because it has a good corrosion resistivity up to 120°C in sulfuric acid. The vessel thickness should be chosen at least one level higher than the normal pressure degree by considering corrosion loss of the vessel's internal surface. We designed a schedule 40 grade seamless pipe. All the components are remote controlled in a control room.

3. Simulation of a test loop

The simulation flow-sheet for the secondary system has been developed with unit operation models provided by the Aspen plus chemical process simulator [4]. The streams are labeled in Figure 3 and the temperature, pressure and mass flow rate are given in Table 1. In the simulation, it was assumed that secondary system was operating at 0.7MPa, 850°C with 0.5 kg/min flowrate of sulfuric acid.

Table 1. Material Balance for Secondary System

Substream	1	2	3	4	5	6
	LIQUID	LIQUID	VAPOR	VAPOR	VAPOR	MIXED
Mole Flow kmol/hr						
H2O	0.031055	0.031055	0.20575	0.329971	0.329971	0.215695
H2SO4	0.30017	0.30017	0.12547	1.25E-03	1.25E-03	0.11553
SO3	0	0	0.1747	0.298916	0.114275	0
SO2	0	0	0	0	0.18464	0.18464
N2	0	0	0	0	0	0
O2	0	0	0	0	0.09232	0.09232
Mole Frac						
H2O	0.093758	0.093758	0.40669	0.523646	0.456731	0.354654
H2SO4	0.906242	0.906242	0.248	1.99E-03	1.74E-03	0.189959
SO3	0	0	0.34531	0.474363	0.158175	0
SO2	0	0	0	0	0.255571	0.303592
N2	0	0	0	0	0	0
O2	0	0	0	0	0.127786	0.151796
Total Flow kmol/hr	0.331225	0.331225	0.50592	0.630141	0.722461	0.608185
Total Flow kg/hr	30	30	30	30	30	30
Temperature C	25	25.32105	429.49	750	850	119.85
Pressure atm	1	6.997286	6.99729	6.997286	6.997286	6.997286

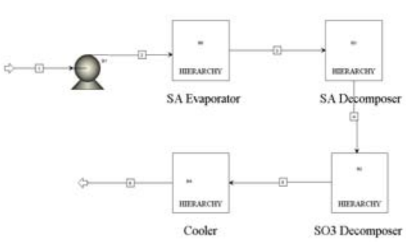


Figure 3. The simulation flow-sheet for secondary system

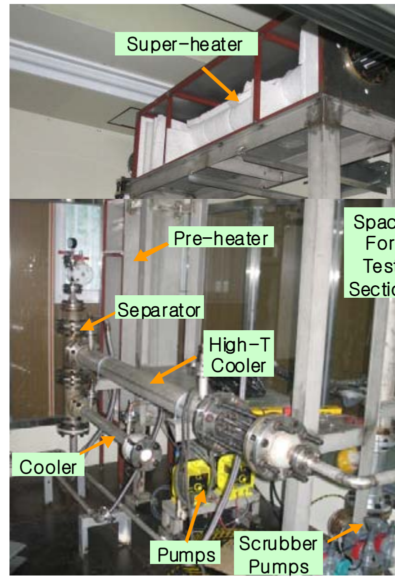


Figure 4. Assembled sulfuric acid facility

3. Conclusion

We designed and assembled a corrosion proof high-pressure and high-temperature sulfuric acid experimental system (Figure 4). We can draw the following conclusions for designing the system;

1. The primary concern of the system is to prevent a sulfuric acid corrosion
2. Quartz heater has a good corrosion resistivity but it is not suitable for a high pressure application
3. Sulfuric acid experimental system is accomplished a high-temperature (500℃) steam experimental condition by a combination of Teflon-lined parts, SiC tubes and Hastelloy C276 connections
4. Sulfuric acid experimental system is achieved 10bar pressure test at room temperature

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