Development of a Lab-Scale Hybrid Acid Cooler for in Very High-Temperature Process-Heat Conditions

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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 of H_2SO_4 decomposition process will operate at a at high pressure (~2.0MPa) and high temperature (~900^{o}C) 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[1]. 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. KAERI (Korea Atomic Energy Research Institute) designed and constructed a small scale sulfuric acid experimental system which simulates a H_2SO_4 decomposition part of the hydrogen production module of a Nuclear Hydrogen Development and Demonstration (NHDD) reactor [2]. The main usage of the loop is to screen and verify the candidates for the nuclear hydrogen coupled components. This experimental system needs a high-temperature sulfuric acid cooling system that is robust for a harsh environment owing to the corrosive chemicals with a very high temperature (>900^{o}C).

2. 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 (evaporator), a H_2SO_4 superheater (or H_2SO_4 decomposer), a PHE(or SO_3 decomposer), a high-temperature cooler, a separator, a SO_2 trap, a low-temperature cooler, and a H_2SO_4 collector (Figure 1).

Cold 96wt% H_2SO_4 is superheated up to 500°C. In a superheating process, H_2SO_4 steam starts decomposition into H_2O and SO_3, if it reaches the temperature of 400 ºC. In the PHE, some fraction of the SO_3 is decomposed into SO_3 and O_2. In the high-temperature cooler, SO_3 and O_2 are formed into H_2SO_4 gas and H_2SO_4 gas and water vapor are condensed into a weaker liquid H_2SO_4 than that at inlet of the loop. The toxic liquid H_2SO_4 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 Sulfuric acid 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

3. Design and validation

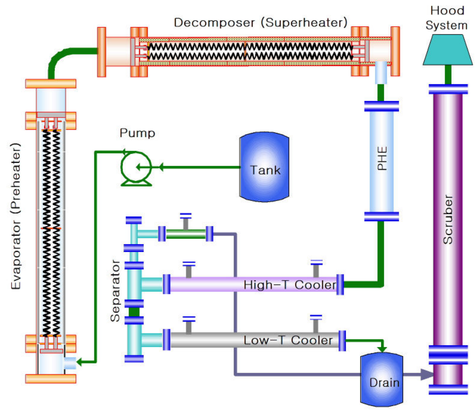

Figure 1 Schematics of a sulfuric acid loop

3.1 Hybrid acid cooler design

Harsh corrosion characteristic of sulfuric acid on metal limits the design of a high-pressure acid cooler. Ceramic or Quarts has excellent corrosion resistance, But they have many manufacturing problems. Quartz is not allowed to use beyond the pressure of 3.0bar due to its fragile characteristics. In the case of ceramics, it allowed at 5~20 bar. But these kind of brittle materials have many difficulties to connect or join between components because they are easy to break especially when subjected to thermal stress.

As described in the previous section, when high temperature H_2O and toxic SO_3 cool down, they react to each other and become sulfuric acid in the high-temperature cooler. We designed a hybrid acid cooler with two cooling zones; a air cooling zone for the hot side and a water cooling zone for the cold side (Figure 2). The sulfuric acid flow path composed of three parts in the cooler; a Teflon-lined outlet plenum, the SiC tubes and a Hastelloy C-276 inlet plenum which has good corrosion resistance in SO_3 environments.

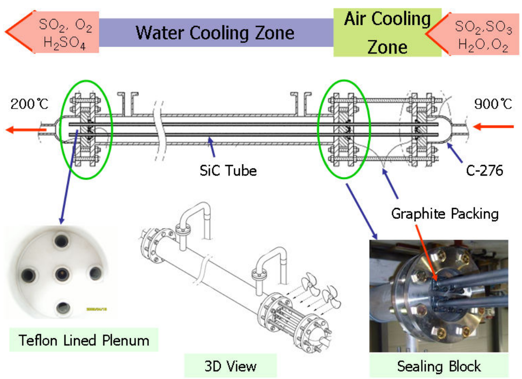

Figure 2. Hybrid acid cooler (Tin = 900°C, Tout = 200°C)

The design solutions are listed in Table 1 to overcome many design problems such as thermal stress, pressure sealing and metal corrosion. Because of the lack of thermodynamic and transport properties in the high-pressure and high-temperature region of sulfuric acid or mixture gas, clumsy calculation for sizing is inevitable and it lead to an excessive design margin of the cooler. The sizing of a 20kW acid cooler is provided by the Aspen plus chemical process simulator [3]. The design input is assumed that the pressure of 5bar, temperature of 150°C with 0.5 kg/min mass flowrate.

Table 1. Problems and solutions for design of an acid cooler

Problem	Key parts	Solution
Corrosion of the Sulfuric acid (or mixture gas) flow path	Materials Teflon, Ceramic, C-276	- Cold side: Teflon lined stainless steel Plenum - Cooling elements: SiC tubes - Hot side: C-276 Plenum + Ion Beam Mixing (periodic maintenances are required)
Thermal Stress of the cooler	SiC tube	- Soft connection of the SiC tubes by using a Graphite compression packing - Hot side: air cooling - Cold side: water cooling
Pressure Sealing	SiC tube Sealing block	- Multiple Graphite compression packing in a C-276 sealing block
Measurement	Thermo-couples	- Gold coating on the surface - Periodic maintenances

3.2 Validation test

We manufactured a Lab-scale (20kW) hybrid acid cooler and performed a pressure test up to 15bar (Figure 3). The overall performance test of the hybrid acid cooler is carried out in conjunction with two kinds of preliminary loop tests; a water (steam) test and a sulfuric acid test at atmosphere, respectively. The operating results of sulfuric acid loop are shown in Figure 4. Unfortunately the inlet temperature of the high-T cooler is very low for both tests. There is a need to raise the inlet temperature for a thermal performance test of the cooler. Teflon lined plenum and SiC tubes show very excellent results for corrosion. But the Hastelloy C-276 plenum, without coated by Ion-beam mixing, is observed a thin corrosion layer (Figure 3(b)

4. Conclusion

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

1. A control of the metal corrosion is a primary design concern for an acid cooler
2. Quartz material has a good corrosion resistivity but it is not suitable for a high pressure acid cooler application
3. Combination of a Teflon-lined plenum, the SiC tubes and a Hastelloy C276 plenum enables good corrosion resistance of a sulfuric acid cooler
4. We achieved a 15bar pressure test and accomplished both a high-temperature steam test and a sulfuric acid test for the present hybrid cooler

In the future, we will perform the validation tests of the hybrid cooler at higher temperature (> 100 C) condition and refine of the analytic design (thermal sizing) method for the high temperature sulfuric acid cooler with *Exothermic Reaction of SO_3 and H_2O and Condensation of H_2SO_4 and H_2O vapor.*

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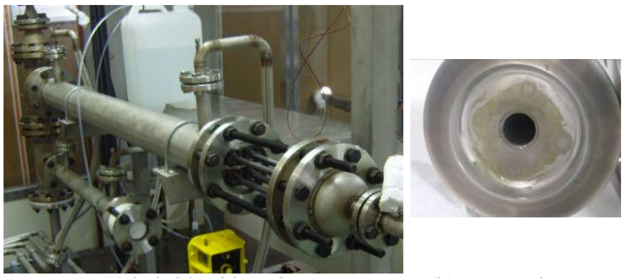

(a) hybrid acid cooler (b) C-276 plenum

Figure 3. Manufactured hybrid acid cooler and Hastelloy C-276 inlet plenum after 12 hours sulfuric acid test

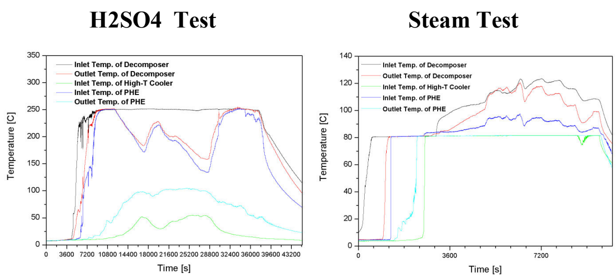

Figure 4. Operating results of a sulfuric acid test and a steam test