Technology Development of an Advanced Small-scale Microchannel-type Process Heat Exchanger (PHE) for Hydrogen Production in Iodine-sulfur Cycle

Injin Sah^{*}, Chan Soo Kim, Yong Wan Kim, Jae-Won Park, Eung-Seon Kim, Min-Hwan Kim VHTR Technology Development Division, KAERI, Yuseong-gu, Daejeon 34057, Republic of Korea ^{*}Corresponding author: injin@kaeri.re.kr

1. Introduction

A massive production of hydrogen with electricity generation is expected in a Process Heat Exchanger (PHE) in a Very High Temperature gas-cooled Reactor (VHTR) system [1,2]. For the application of hydrogen production, a small-scale gas loop for feasibility testing of a laboratory-scale has constructed and operated in Korea Atomic Energy Research Institute (KAERI) as a precursor to an experimental- and a pilot-scale gas loops [3].

In this study, ongoing manufacturing processes of the components employed in an advanced small-scale microchannel-type PHE are presented. The components, such as mechanically machined microchannels and a diffusion-bonded stack are introduced. Also, preliminary studies on surface treatment techniques for improving corrosion resistance from the corrosive sulfuric environment will be covered.

2. Methods and Results

2.1 Advanced Small-scale Microchannel-type PHE

Figure 1 shows the schematic diagram of overall small-scale microchannel-type PHE consisting of the diffusion-bonded stack, plenums, and flanges. The plates for primary side (thickness of 2.5 mm, diameter of the channel 1.5 mm, and pitch of 4 mm) and secondary side (thickness of 2.5 mm, diameter of the channel 2.5 mm, and pitch of 4 mm) are stacked layer by layer by solid-state diffusion bonding process producing core dimension of $240 \times 160 \times 150$ mm³.

As expected, the heat is transferred from the primary side to the secondary side by counter-flow at the center



Fig. 1. An advanced small-scale microchannel-type PHE: (a) overall view, (b) plates with microchannel (primary side), and (c) plates with microchannel (secondary side)

of the core in parallelogram region and the cross-flow at both inlet and outlet region in the right-angled triangle region. Accordingly, the effective heat transfer region of the advanced small-scale PHE is $200 \times 120 \text{ mm}^2$.

2.2 Solid-state Diffusion Bonding

The chemical compositions of a commercial grade Ni-base alloy, Hastelloy-X, are summarized in Table I. Diffusion bonding is employed by a contractor (TNP

Hastelloy-X	Ni	Cr	Fe	Мо	Со	W	Mn	Р
Plate (2.5T)	Bal.	21.46	18.96	8.66	1.11	0.45	0.30	0.014
522797-02								
Hastelloy-X	S	Si	Al	Cu	Ti	В	С	Others
Plate (2.5T)	0.0001	0.42	0.21	0.09	0.01	0.003	0.06	-
522797-02								

Table I: Chemical compositions of the Hastelloy-X used in this study (wt.%)

Designation	Temperature	Pressure	Duration time	Surface condition
HX-A	1100 °C	14 MPa	70-80 min	Mechanical polishing
HX-B	1150 °C			(SiC #1000)
HX-C	1200 °C			



Fig. 2. Cross-sectional SEM micrographs of the diffusion-bonded Hastelloy-X: (a) HX-A, (b) HX-B, and (c) HX-C

Co.) following the diffusion bonding conditions suggested by the authors (Table II). As shown in the table, diffusion bonding is conducted in the temperature ranges of solution annealing with the uniaxial compressive stress applying a few percentage of plastic deformation after the diffusion bonding process.

Figure 2 shows the cross-sectional micrographs of the diffusion-bonded Hastelloy-X near the interface. While the precipitates are extensively present along the interface in HX-A condition, the precipitates are reduced in both HX-B and C conditions resulting in the



Fig. 3. Tensile properties of the diffusion-bonded Hastelloy-X: (a) UTS and (b) elongation

grain boundary migration across the interface while leaving the interface almost devoid of precipitates and barely visible.

Figure 3 shows the tensile properties of the diffusionbonded specimens compared with those of the asreceived. Up to the test temperature of 500 $^{\circ}$ C, the elongation of the diffusion-bonded reaches to that of the as-received in all condition. However, above the test temperature of 700 $^{\circ}$ C, the planar grain boundary and the presence of precipitates at the interface lead to the poor joint quality for both HX-A and B. In contrast, the tensile strength and elongation are similar to those of the as-received up to 900 $^{\circ}$ C for HX-C on account of extensively migrated grain boundaries.

2.3 Surface Treatment

In the iodine-sulfur cycle for the hydrogen production, metals/alloys are not suggested for the candidate materials of a PHE which is exposed to He/N_2 gas in the primary side and decomposed sulfuric acid gas (SO_2/SO_3) in the secondary side at high temperature. In contrast, ceramics, such as SiC and Al_2O_3 , have shown good corrosion resistance in such environment [4,5]. In this sense, a ceramic coating is applied on top of substrate to protect the alloy from the environment.

To enhance the adhesion at the interface (SiC/Hastelloy-X), ion beam mixing (IBM) method is employed by the following experimental procedure. Hastelloy-X plates with microchannels (thickness of 2.5 mm, diameter of channel 2.5 mm, and pitch of 4 mm) are loaded in a vacuum chamber and sputtered for about 10 min with nitrogen ions to eliminate the surface containments before film pre-deposition. Then, electron beam physical vapor deposition (EB-PVD) of SiC is applied on the surface up to the thickness of 50 nm. IBM process is followed to enhance the adhesion of the interface with the accelerating voltage of 70 kV and current of 5 mA with nitrogen ions $(5 \times 10^{16} - 1 \times 10^{17})$ $\#/cm^2$). Further deposition of SiC is performed up to 1 µm by EB-PVD on the pre-deposited SiC and then ion beam is again bombarded on the deposited SiC layer for about 10 min. The coated specimens are subjected to heat treatment (950 °C/2 h) in a vacuum chamber to release the stress developed during coating process. The procedure above is repeated for up to four times.

To verify the improvement of the corrosion resistance through the IBM process, corrosion testing is in progress for both the as-received and surface-treated specimens in boiling aqueous sulfuric acid (H_2SO_4) environment.

3. Conclusions

Ongoing manufacturing process for an advanced small-size microchannel-type PHE in KAERI is presented. Through the preliminary studies for optimizing diffusion bonding condition of Hastelloy-X, a diffusion-bonded stack, consisting of primary and secondary side layer by layer, is scheduled to be fabricated in a few months. Also, surface treatment for enhancing the corrosion resistance from the sulfuric acid environment is in progress for the plates with microchannels.

ACKNOWLEDGEMENT

This study was supported by the Nuclear Research and Development Program of the National Research Foundation of Korea (NRF) grant funded by the Ministry of Science, ICT and Future Planning (Grant Code: NRF-2012M2A8A2025682).

REFERENCES

[1] J. Chang, Y.-W. Kim, K.-Y. Lee, Y.-W. Lee, W.J. Lee, J.-M. Noh, M.-H. Kim, H.-S. Lim, Y.-J. Shin, K.-K. Bae, and K.-D. Jung, A Study of a Nuclear Hydrogen Production Demonstration Plant, Nuclear Engineering and Technology, Vol.39, No.2, p.111, 2007.

[2] S. Kubo, H. Nakajima, S. Kasahara, S. Higashi, T. Masaki, H. Abe, and K. Onuki, A Demonstration Study on a Closedcycle Hydrogen Production by the Thermochemical Watersplitting Iodine-sulfur Process, Nuclear Engineering and Design, Vol.233, p.347, 2004.

[3] C.S. Kim, S.-D. Hong, Y.-W. Kim, J.-H. Kim, W.J. Lee, and J. Chang, Thermal Design of a Laboratory-scale SO₃ Decomposer for Nuclear Hydrogen Production, International Journal of Hydrogen Energy, Vol.33, p.3688, 2008.

[4] S. Kubo, M. Futakawa, I. Ioka, K. Onuki, and A. Yamaguchi, Corrosion Resistance of Structural Materials in High-temperature Aqueous Sulfuric Acids in Thermochemical Water-splitting Iodine-sulfur Process, International Journal of Hydrogen Energy, Vol.38, p.6577, 2013.

[5] J.-W. Park, Y. Chun, and J. Chang, Effects of Ion Beam Mixing of Silicon Carbide Film Deposited onto Metallic Materials for Application to Nuclear Hydrogen Production, Journal of Nuclear Materials, Vol.362, p.268, 2007.