Development of Process Heat Exchanger (PHE) for Nuclear Hydrogen Production System

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1. Introduction

Process Heat Exchanger (PHE) consisting of He channels and sulfuric acid gas channels requires surface modification of metallic materials for corrosion protection and then a diffusion bonding of the machined and surface modified metallic sheets. Hastelly X was chosen as a material for PHE. For the surface modification Hastelloy X sheet materials subjected to an environment of the decomposed sulfuric acid were SiC-coated with an aid of ion beam mixing (IBM). The multiply conducted coating/IBM exhibited almost no corrosion in 80% sulfuric acid at 250℃ for up to 100hrs . A process for a good bonding between Hastelloy X sheets, which is essential for a good heat exchanger, was developed by diffusion bonding. The diffusion bonding was done by mechanically clamping the sheets with a heating above 1000 ˚C. When the clamping jig was made of materials having equal or less thermal expansion coefficient than the Hastelloy-X, a sound bonding was achieved. No He gas leak was detected up to 50 atm pressure.

2. Experiments and Results

2.1 Experiments

Surface modifications

Prior to a SiC deposition, a sputter cleaning of the sample (1cmx1cmx0.5cm) was carried with an N ion energy of ~10 keV and a current of 0.5 Ampere. Then, the deposition of the SiC was performed to 50 nm thickness, followed by a nitrogen ion beam mixing at 70keV with a dose of ~ $5x10^{16}$ ions/cm^2. A further SiC evaporative deposition up to a total of ~ 1 μm was then conducted with a deposition rate of ~3 Å/s produced by an electron beam current of ~0.15 A. It is the outermost surface of the samples that was again N-irradiated (hammered) at 70 keV with a dose of ~ $1x10^{17}$ ions /cm^2 to $4x10^{17}$ ions/cm^2 to increase their resistance to sublimation.

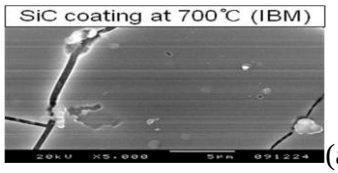
The samples were then placed in an alumina boat and annealed in a quartz tube vacuum furnace with a heating rate of ~7.5℃/min at temperature ranges of 550℃ to 950℃ for 2 hrs. This process was conducted multiply up to 4 times. Then the samples were placed in 80% sulfuric acid and the weight was measured periodically.

Diffusion bonding

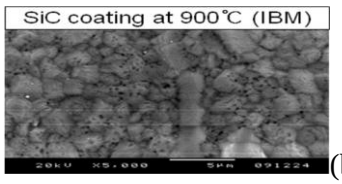
Hastelloy X work pieces were clamped mechanically with jigs made of SKD61 and the annealed SUS304 sheets in air were placed in between the Hastelly X work pieces, followed by a heating at 1000 ℃ in a vacuum for 5 hours. To ensure a leak protection, 30 ㎛ thick Ni foil was inserted at one occasion. The work pieces were tightly contacted by screwing the jig.

2.2 Results and Discussion

After heating at 700℃ in vacuum, SiC film was maintained as-coated on the Hastalloy-X surface, but cracked along the grain boundary of Hastelloy X substrate attributed to the thermal etching (Fig.1a). After annealing at 900℃, the coated layer was all crystallized exposing the coated surface as a result of the film island formation (Fig1b). In spite of a high difference in their CTEs, no film delamination occurred. This is attributed to IBM to produce a highly adherent coated layer and an interfacial reaction during annealing [1].



(a)



(b)

Fig. 1. The IBM coated SiC on Hastelloy X is cracked along the grain boundary of Hastelloy X substrate at ~700℃ along the grainboundary of the Hastelloy X substrate, while the coated layer is crystallized at 900℃.

Fig. 2 shows the corrosion test results of the as-received, as-coated and the one time, two times, three times and four times coating/IBM annealed samples. The as-received sample was corroded out completely after ~70 hours, however the IBM SiC coated samples were sustained for much longer duration. The one time IBM coated and annealed sample showed less weight retain rate than the as coated sample. This is attributable to the film island formation after annealing. The number of the process increases, the weight retain rate increases and finally almost no corrosion was achieved after 4 times processing.

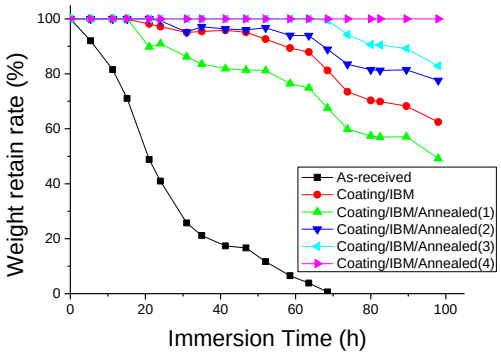


Fig. 2. 80% sulfuric acid corrosion test result: As the process of Coating/IBM/Coating/Annealing is multiply conducted, the weight retain rates are increased. The weight is not changed after 100 hrs immersion when the process repeated 4 times.

Fig. 3 shows a drawing of the diffusion bonded jig and the diffusion bonded interfaces. The direct diffusion bonded interface between Hastelloy X sheets (Fig. 3a) and the bonded interface with Ni foil insertion (Fig. 3b) are shown. Both methods do not show any incompletely bonded interface. However, we have found a gas leak protection is more ensured by the Ni foil insertion than the direct diffusion bonding. The small sized PHE produced by this method exhibited no leak up to 50 atm of He gas.

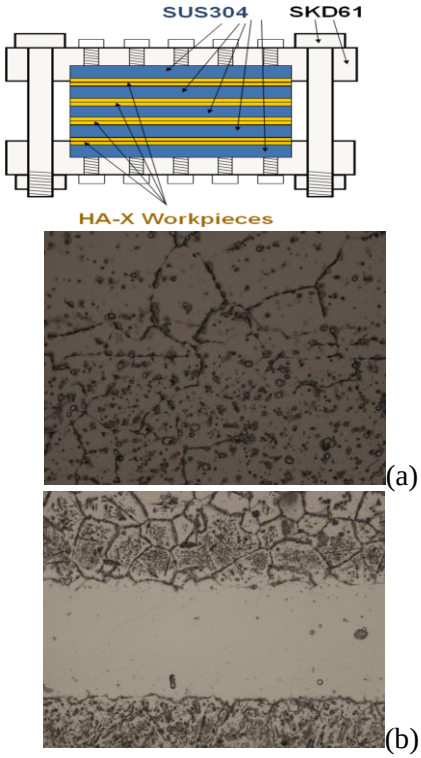


Figure 3. A drawing of diffusion bonded jig and the diffusion bonded interfaces: (a) The direct diffusion bonded interface between Hastelloy X sheets and (b) the bonded interface with Ni foil insertion.

3. Conclusions

Multiply processed IBM coated SiC on Hastelloy X showed almost no corrosion in 80%sulfuric acid up to 100 hrs, whereas the as-received is completed corroded out after 70 hours. The diffusion bonding by mechanically clamping Hastelloy X sheets with SUS304 was done with a heating above 1000 ˚C. When the clamping jig was made of materials having equal or less thermal expansion coefficient than the Hastelloy-X, a sound bonding was achieved with no He gas leak up to 50 atm pressure.

Acknowledgement

This study was supported by both Nuclear Hydrogen Development Project and Proton Engineering Frontier Project sponsored by Ministry of Education, Science and Technology, Republic of Korea

REFERENCES

[1] J. Park, Z. S. Khan, H. Kim, Y. Kim, Mater. Res. Symp. Proc., Vol. 1125, p65, 2009