A Design of Sulfuric-Acid Decomposer for Nuclear Hydrogen Production System Utilizing Very High Temperature Gas Cooled Reactor

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ABSTRACT

A design of a sulfuric-acid decomposer for a nuclear hydrogen production system is presented in this paper. The heat exchanger is a hybrid type to meet the design requirements of the sulfuric oxides gas decomposer. A high temperature super alloy is used for the heat exchanger material due to its strength requirements at an elevated temperature. The Ni-based super alloy is coated with a silicon carbide to enhance the corrosion resistance. The coated layer and the base material have been mixed by using of the ion beam to release the stepwise change of material properties at the interface. The strength of the plate-fin type flow channel was investigated based on ASME design code at the elevated temperature. The stress field at the coated surface of the decomposer was also investigated with a simplified method.

INTRODUCTION

Very high temperature gas cooled technology and IS cycle technology are being developed in KAERI for the nuclear hydrogen production system [1~3]. The outlet temperature of the high temperature gas cooled reactor developed so far ranges 750 to 900°C. However, to produce hydrogen with economical efficiency, the coolant outlet temperature of VHTR should exceed 950°C. As shown in Fig. 1, there are three important loops, the 1st loop transfers the heat generated from the core to the intermediate heat exchanger, the 2nd loop is an intermediate loop which connects the 1st loop and the 3rd loop of the IS process. The sulfuric-acid decomposer is a key component which transfers the heat of the 2nd loop to the 3rd loop. This heat exchanger will suffer the extreme environments of high corrosion, high temperature and high differential pressure. A metallic heat exchanger has a short lifetime due to the complex interaction between stress and corrosion. A ceramic heat exchanger with a strong corrosion resistance has difficulties in the manufacturing and the thermal shock resistance because of its low toughness [4].

In this paper, a heat exchanger is being developed to overcome both the short lifetime and the difficulty in manufacturing. Base material of this process heat exchanger is a high temperature super alloy for easy manufacturing possibility than ceramic material. The presented design concept for the sulfuric-acid decomposer which shall be used is efficient at the elevated temperature and the sulfia acid conditions. The heat exchanger is a hybrid type exchanger in which the plate-fin type and printed circuit type heat exchanger concepts are combined. In order to enhance corrosion resistance, the heat exchanger is designed of Ni-based material which is coated with silicon carbide. In addition to this, ion beam mixing technology has been developed to reduce possible delamination between the SiC layer and the base material by releasing the stress gradient [5,6]. The shape design of the heat exchanger and surface modification technology are introduced. The effect of each design
feature on the structural integrity is also discussed.

DESIGN STUDY

Type of Heat Exchanger

The sulfuric-acid gas decomposer should maintain structural integrity at the operation temperature more than 900. Also, it should withstand pressure difference between intermediate loop and I-S loop. The sulfuric gas channel of the heat exchanger is exposed to a highly corrosive environment. There should be enough space to locate the catalyst which is necessary to decompose the sulfuric oxides gas. Based upon these design requirements, a hybrid heat exchanger shown in Fig. 2 is developed [7]. The heat exchanger exposed to helium gas is designed of a plate type heat exchanger where flow channel is small enough to withstand the mechanical loading. However, the flow channel of the sulfuric gas channel is designed of a plate-fin type heat exchanger in order to provide catalyst space. Also, the plate-fin type heat exchanger provides sufficient space for the catalyst maintenance. Comparing to a plate-fin type heat exchanger, this hybrid heat exchanger is more efficient to withstand the pressure difference between loops. Layout of the heat exchanger is a counter flow type since the decomposition of sulfuric oxides gas is an endothermic reaction. The thermal sizing methodology for this hybrid type sulfuric oxide gas decomposer including the endothermic reaction has been developed [8]. The size of the heat exchanger has been determined to meet small gas loop which has 10kw thermal power at the test section. All of the structural elements of the heat exchanger are designed of Hastelloy-X.
Implementation of Ion Beam Mixing Technology

The surface modification consists of coating and ion beam mixing. Silicon carbide (SiC) was chosen as the coating materials in this study. The silicon carbide coating is known to improve the lifetime or the performance of metallic substrates when exposed to aggressive environments. The high resistance of SiC to the corrosion could be due to the very strong covalent bonding between silicon and carbon and its tetrahedral coordination. Hastelloy X has been selected as a metallic substrate because the thermal expansion coefficient of Hastelloy X is more similar to that of SiC than any other Ni-based alloys and its corrosion resistance in the SO$_3$/SO$_2$ gases has been known to be better than the other Ni based alloys.

Fig. 3 shows the optical microscopic observation of the surfaces etched electrolytically. The depth of erosion of the uncoated part of the sample was about 35 µm as estimated by measuring the moving distance of the lens during focusing the image. The sample without the ion bombardment shows flakes at the edge of the film (Fig. 3a) after electrolytic etching, implying that the corrosion initiates from the uncoated substrate to the film and then penetrates underneath the film. As the erosion propagates, the film is flaked-off from the edge. However, such corrosion is not found in the ion-bombarded sample after etching in the same conditions (Fig.
This means that the substrate under the film was reinforced to be corrosion-protective by ion beam mixing [5].

![Without ion beam mixing](image1)

![With ion beam mixing](image2)

**Fig.3.** Optical-microscopic observation of the surfaces etched electrolytically; as-deposited and ion beam mixed samples.

On the basis of this coupon test result, it has been expected that ion beam coating with ion beam mixing technology is one of the powerful solution for SO₃ decomposer heat exchanger operating at a high temperature corrosive environment.

### SIMPLIFIED STRESS ANALYSIS

**Stress Analysis for Plate-Fin Flow Channel**

Unit section of sulfuric-acid decomposer is shown in Fig. 4. ABAQUS has been used for the stress analysis of the heat exchanger. Two-dimensional solid element was used for the finite element analysis. Temperature dependent material properties were used in the analysis. CPE4 element was used for the analysis. In this modeling, coating at the surface was not considered since the thickness of the coating is so thin that it does not have an influence on the global structural behavior of the heat exchanger. The thickness of the plate is around 1mm but the thickness of the coating is several μm or less than 1 μm. Therefore, it is nearly impossible to model the coating zone and the mixing zone in the global stress analysis with a conventional method. Symmetric boundary condition is applied in one side and in the other side horizontal displacement was coupled as shown in Fig. 4. Most of the global pressure loading was supported by the fin as was shown in the stress contour. Stress analysis based on ASME was shown in Table 1 and maximum allowable pressure was estimated. The plate-fin
Type heat exchanger can withstand around 10MPa pressure at 950°C due to creep deformation.

Fig. 4 Boundary condition and stress contour at the plate-fin channel.

Table 1 Allowable pressure loading based on ASME.

<table>
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Simplified Local Stress Analysis at Coated Region

Stress fields for this coated side is modeled as three different layers which are coated layer, mixed layer and base material. The thickness of the coated layer is thin enough to consider that it has a negligible influence on the global structural behavior of the heat exchanger. Stepwise stress change at the interface before ion beam mixing and smooth change after ion beam mixing, which are calculated from one-dimension calculation for various coating material, is shown in Fig. 5.

Fig 5. Section view of heat exchanger and surface stress distribution.
A high stress field is expected at the surface due to thermal expansion coefficient difference between base material and coated material. A simplified shear lag analysis was used to estimate the stress field of the coated layer [9]. The displacement fields can be assumed when the plate is subject to a constant strain caused by thermal loading. The model and notations used for the calculation is given in Fig. 6. Detailed derivation of the equation was described in this work. The transverse shear stress component, which is expressed as displacement difference between coated layer and base material, is dominant in the mixing zone. Mixing zone is assumed to be the shear lag layer in the calculation.

Fig. 6 Simple model for local stress analysis.

It means that the coated layer is sound when the transverse crack spacing \( l \) becomes infinite. As the temperature elevates, the spacing of the transverse crack is decreased. That is, the number of cracks is increased. The multiplication of the crack can be predicted by the average stress concept. For the SiC coating on the Ni based alloy, the stress distribution at the coated layer is shown in Fig. 7 for various coating thickness.

Fig. 7 Effect of coating thickness and mixing zone size on the stress distributions.

The stress distribution is also influenced by the thickness of the mixing zone thickness as was shown in Fig. 7. However, the Young’s modulus difference between the base material and the coating material does not have much influence on the stress distribution.
SUMMARY AND CONCLUSION

A hybrid heat exchanger with ion beam coated and mixed plate is a promising candidate of the sulfuric-acid decomposer for nuclear hydrogen production system. It adapts super alloy for base material which is easy to fabricate comparing to the silicon carbide. The base material is coated with the silicon carbides to protect super alloy from the corrosive environment. Also, it is a hybrid heat exchanger with the plate type and the plate-fin type in order to withstand the high pressure of the helium side. Thermal expansion coefficient difference between the coated material and the base material and the mixing depth are important to the structural integrity of coating layer. However, those have negligible influence on the overall structural behavior of the heat exchanger. The developed SO$_3$ decomposer is being manufactured and it will be tested at a gas loop in 2008.

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REFERENCE