High Temperature Heat Exchanger Project
Under Financial Assistance
DE-FC07-04ID14566

Awarded by the United States of America Acting Through the
United States Department of Energy

High Temperature Heat Exchanger Project

FINAL REPORT

Period Ending September 30, 2008

The UNLV Research Foundation
4505 Maryland Parkway
P. O. Box 452036
Las Vegas, NV 89154-2036

Anthony E. Hechanova, Ph.D.
Project Manager
(702) 895-1457
(702) 895-2354 (FAX)
hechanova@unlv.nevada.edu

High Temperature Heat Exchanger Project Phase I Completion Report

Table of Contents

1.0 HTHX Program 3

2.0 Metallic Materials Characterization and Testing at UNLV (Ajit Roy, Principal Investigator, Department of Mechanical Engineering) 4

3.0 Construction Materials Screening for the HI Decomposition Process at GA (Bunsen Wong, Principal Investigator, General Atomics) 11

4.0 Ceramic Heat Exchanger Development for Application to NHI at Ceramatec, Inc. (Merrill Wilson, Principal Investigator, Ceramatec, Inc.) 15

5.0 Materials, Design, and Modeling for C/SiC Ceramic Heat Exchangers at UC Berkeley (Per Peterson, Principal Investigator, UCB Nuclear Engineering Department) 18

6.0 Development of Self-Catalytic Materials at MIT (Ronald Ballinger, Principal Investigator, MIT Departments of Materials and Nuclear Engineering) 22

7.0 Heat Exchanger Design at UNLV (Yitung Chen, Principal Investigator, UNLV Mechanical Engineering Department) 24

8.0 Experimental Facility Design for the Testing of Prototype Components at UNLV (Samir Moujaes, Principal Investigator, UNLV Mechanical Engineering Department) 29

9.0 Analytical Studies of the Effects of Acid Exposure on Structure Materials (Allen Johnson, Principal Investigator, UNLV Harry Reid Center for Environmental Studies) 31

10.0 Efficiency Improvement and Cost Reduction of Solid Oxide Electrolysis Cells at UNLV (Clemens Heske, Principal Investigator, UNLV Department of Chemistry) 34

1.0 HTHX Program

1.1 Introduction

The UNLV Research Foundation (UNLVRF) assembled a research consortium for high temperature heat exchanger (HTHX) design and materials compatibility and performance comprised of university and private industry partners under the auspices of the U.S. Department of Energy Office of Nuclear Energy, Science & Technology (DOE-NE) Nuclear Hydrogen Initiative in October 2003. The objectives of the consortium were to conduct investigations of candidate materials for high temperature heat exchanger components in hydrogen production processes and design and perform prototypical testing of heat exchangers. The initial research of the consortium focused on the intermediate heat exchanger (located between the nuclear reactor and hydrogen production plant) and the components for the hydrogen iodine decomposition process and the sulfuric acid decomposition process. These heat exchanger components were deemed the most challenging from a materials performance and compatibility perspective.

1.2 Objectives

To assist DOE-NE in the development of hydrogen production from nuclear energy through:

- Identification and testing of candidate materials and coolants for heat exchanger components.
- Design of critical components in the interface and sulfur iodine thermochemical process.
- Fabrication and testing of prototypical components.
- Innovative materials development.

1.3 Approach

The research was divided into three technical areas: metallics, ceramics, and catalysts. Materials were identified through communications and consensus among the UNLVRF consortium members and researchers within the Nuclear Hydrogen Initiative materials group. Corrosion testing in hydrogen iodide was conducted at General Atomics. Materials evaluation and characterization were conducted at UNLV and General Atomics for metallic materials, UC Berkeley and Ceramatec, Inc. for ceramic materials, and the Massachusetts Institute of Technology (MIT) for catalytic materials. A variety of heat exchanger designs were also investigated for the NHI applications. The consortium met quarterly to update collaborators on research progress and exchange information.

2.0 Metallic Materials Characterization and Testing at UNLV (Ajit Roy, Principal Investigator, Department of Mechanical Engineering)

2.1 Introduction

The structural materials for high temperature heat exchangers to be used in nuclear hydrogen generation using the Sulfur-Iodine (S-I) cycle must possess significant corrosion resistance and excellent resistance to plastic deformation including the structural stability of the component. In view of these rationales, a few candidate materials were identified for evaluations of their metallurgical and corrosion properties under conditions relevant to the S-I cycle. These materials include Alloys C-22, C-276, 800H, Waspaloy and Zr705. While the metallurgical and corrosion testing involving these five alloys is in progress, additional materials namely, AL610, Alloy 617, Nb1Zr and Nb7.5Ta were added in FY05 to the list of S-I cycle candidate materials.

The tensile properties of all five alloys cited above were determined at temperatures ranging from ambient to 600°C using MTS/Instron testing equipment. Tests are in progress at temperatures above 600°C up to a maximum temperature of 1000°C. The susceptibility of the test materials to stress corrosion cracking (SCC) was determined under both constant load (CL) and slow-strain-rate (SSR) conditions.

The effect of controlled potentials (E_{cont}) on the SCC behavior of these alloys is also being studied. The general and localized corrosion behavior of these alloys is also under investigation at elevated temperatures (500°C) using the newly installed autoclave system. An electrochemical polarization technique is also being used to determine the critical potentials of these alloys in a simulated environment. Further, the metallographic and fractographic evaluations of all tested specimens are being performed by optical microscopy and scanning electron microscopy (SEM), respectively.

The impact resistance and the fracture toughness of the structural materials were determined using fracture mechanics concepts. In addition, the crack growth behavior of the candidate materials was studied. Transmission electron microscopy (TEM) was utilized to determine the characteristics of imperfections (voids/dislocations) in the tested specimens.

2.2 Research Highlights and Accomplishments

- The results of tensile testing indicate that Alloys C-22, C-276, 800H and Waspaloy are capable of maintaining metallurgical stability at elevated temperatures. However, Waspaloy outperformed the other three alloys in terms of tensile strength at elevated temperatures.
- The tensile parameters (the yield strength (YS), the ultimate tensile strength (UTS), the percent elongation (%El) and the percent reduction in area (%RA)) of Alloys C-22, C-276, 617 and 718 at temperatures ranging from ambient to 1000 C were completed.
- Failure strain were gradually reduced in a critical temperature range followed by its enhancement beyond this range. This phenomenon may be the result of dynamic strain ageing. For Waspaloy, a maximum dislocation density (ρ) was observed at 300ºC.

- Ductile failures were observed up to a temperature of 600ºC for all tested materials. However, intergranular failure was observed with Waspaloy at 800ºC possibly due to the precipitation of Boron at the grain boundaries.
- The reduced ductility in terms of %El and %RA within certain temperature regimes has been attributed to the occurrence of dynamic-strain-aging (DSA) behavior of susceptible materials related to the diffusion of solute elements and their accumulation near the grain boundaries. The reduced dislocation mobility through grain boundaries can lead to reduced plastic strain in terms of failure strain (ef) or %El.
- Transmission electron microscopy (TEM) was used to characterize dislocations using the resultant micrographs and superimposition of grids onto them. The results indicate that the magnitude of dislocation density (ρ) was highest at temperatures in the vicinity of 200 and 300 C (C-276), where reduced plastic strains were noted. The magnitude of ρ was relatively lower at 450 C due to enhanced dislocation mobility and ease of plastic deformation.
- Zr705, a candidate structural material for application in the HIx decomposition process showed reduced failure strain below 200°C possibly due to the dynamic strain ageing effect. A similar effect of temperature on failure strain has also been noted for the other four tested materials.
- None of the candidate materials showed failure in the simulated S-I environment under constant load at applied stresses corresponding to either 95 or 98% of the materials' yield strength values.
- The results of SSR testing in a similar solution showed a gradual reduction in true failure stress with increasing temperature. However, insignificant variations in the ductility parameters and time-to-failure (TTF) were noted in these tests.
- The results of localized corrosion studies by the cyclic potentiodynamic polarization (CPP) technique revealed more active (negative) critical potentials at elevated temperatures.
- The results of autoclave testing in a 150°C acidic solution indicate that Alloy C-276 experienced the lowest corrosion rate in terms of mils per year (mpy). The C-ring specimens of Zr705 did not exhibit cracking even after an exposure of four weeks.
- The fractographic evaluations of the tested specimens by SEM revealed dimpled microstructures indicating ductile failures.
- Evaluated metallographic evaluations and fractographic evaluations. A mechanistic understanding of DSA is now established based on activation energy and work hardening index.
- A fracture morphology was used to identify ductile or brittle failure as a function of temperature.
- The Scanning Electron Microscopy (SEM) micrographs of Alloy C-276 revealed dimpled microstructures in tensile testing at temperatures up to 600 C, indicating ductile failures. However, brittle failures were seen at temperatures of 700 and 800 C.
- Fractographic evaluations of the Alloy C-22 specimens by SEM revealed ductile failures up to 500 C, beyond which a combination of ductile and brittle failures was seen.
- The superimposition of the s-e diagrams obtained on Nb7.5Ta at temperatures ranging from ambient to 400 C also indicates the occurrence of DSA, showing the lowest failure strain (ef) at 300 C. The analysis of the TEM micrographs revealed an order of magnitude higher ρ value at 300 C compared to that at room temperature. The SEM micrographs of the specimens tested at different temperatures revealed predominantly ductile failure characterized by dimples.

- The results of SSR testing involving Alloy C-22 indicate that the magnitude of %El, %RA, time to failure (TTF) and true failure stress (σf) was reduced in the presence of acidic solution at 90 C. A similar trend was seen with Nb7.5Ta.

2.4 Project Profile

Start Date: August 2004

End Date: August 2007

Researchers: A. Roy (Principal Investigator), A. Ghosh, and C.K. Mukhopadhyay
Students: M. Hasan, A. Kaiparambil, R.A. Karamcheti, V. Kondur, R.S. Koripelli, N. Kothapalli, V. Marthandam, J. Pal, R. Santhanakrishnan, L. Savalia, A. Venkatesh, V. Virupaksha, and J. Yelavarthi

Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):

A. Roy, J. Pal, V. Marthandam, A. Venkatesh, R.S. Koripelli, and M. Hasan, "Crack Growth Rate of Structural Materials for Heat Exchanger Applications," Technical Report, UNLV Department of Mechanical Engineering, August 8, 2007.

A. Roy, J. Pal, V. Marthandam, and M. Hasan, "Fracture Toughness Measurements of Structural Materials for High Temperature Heat Exchanger Applications," Technical Report, UNLV Department of Mechanical Engineering, June 29, 2007.

A. Roy, R.S. Koripelli, and J. Pal, "Stress Corrosion Cracking Behavior of Structural Materials for Liquid Sulfuric Acid Applications," Technical Report, UNLV Department of Mechanical Engineering, June 4, 2007.

A. Roy, R.S. Koripelli, V. Marthandam, A. Venkatesh, A. Kaiparambil, V. Virupaksha, J. Yelavarthi, and J. Pal, "High-Temperature Heat-Exchanger Materials," Preliminary Technical Report, UNLV Department of Mechanical Engineering, March 30, 2006.

Theses Generated:

Rama S. Koripelli, "Tensile Deformation, Corrosion and Crack-Growth Characterization of a Nickel-Base Alloy," Ph.D. Thesis, Department of Mechanical Engineering, UNLV, December 2007.

Joydeep Pal, "Tensile Properties, Fracture Toughness and Crack Growth Study of Alloy C-276," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2006.

Venkateswarhu Kondur, "Characterization of Refractory Materials for HIx Decomposition Process," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2006.

Jagadesh Yelavarthi, "High-Temperature Deformation and Environment-Induced Degradation of Waspaloy," M.S. Thesis, Department of Mechanical Engineering, UNLV, August 2006.

Vinay Virupaksha, "Use of Alloy 800H for Applications in Hydrogen Generation using Nuclear Power," M.S. Thesis, Department of Mechanical Engineering, UNLV, May 2006.

Ancila Kaiparambil, "Characterization of Structural Materials for Applications in HIx Decomposition," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2005.

Raghunandan A. Karamcheti, "Corrosion and Metallurgical Behavior of Alloy-22 for High-Temperature Applications," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2005.

Radhakrishnan Santhanakrishnan, "Metallurgical and Corrosion Characterization of Structural Materials for S-I Cycle," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2005.

Lalit Savalia, "Performance of Waspaloy at Elevated Temperature for Heat Exchanger Applications," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2005.

Narendra Kothapalli, "Characterization of Alloy C-276 as a Structural Material for High-Temperature Heat Exchangers," M.S. Thesis, Department of Mechanical Engineering, UNLV, August 2005.

Conference Proceedings:

R. Koripelli and A. Roy, "Tensile Deformation of Alloy-22 at Ambient and Elevated Temperatures," PVP2007-26464, American Society of Mechanical Engineers, San Antonio, TX, July 2007.

V. Kondur, A. Ghosh, and A. Roy, "Tensile Behavior of Nb7.5Ta for Heat Exchanger Applications," PVP2007-26490, American Society of Mechanical Engineers, San Antonio, TX, July 2007.

V. Marthandam and A. Roy, "Tensile Deformation of Alloy 617 at Different Temperatures," PVP2007-26500, American Society of Mechanical Engineers, San Antonio, TX, July 2007.

J. Pal, C. Mukhopadhyay, and A. Roy, "Cracking of Alloy C-276 in an Acidic Environment," PVP2007-26429, American Society of Mechanical Engineers, San Antonio, TX, July 2007.

A. Venkatesh and A. Roy, "The Evaluation of the Cracking Susceptibility of Alloy 718 in an Acidic Solution," PVP2007-26493, American Society of Mechanical Engineers, San Antonio, TX, July 2007.

A.K. Roy, R.S. Koripelli, J. Pal, and A. Venkatesh, "Tensile Properties of Superalloys for Components in Hydrogen Generation Systems," MS&T Conference, Cincinnati, OH, October 2006.

A.K. Roy, A. Venkatesh, and J.K. Yelavarthi, "Structural Stability of Waspaloy at Elevated Temperatures," AMPT Conference, Las Vegas, NV, July-August 2006.

A.K. Roy and R.S. Koripelli, "Effect of Temperature on Tensile Properties of Alloy C-22," AMPT Conference, Las Vegas, NV, July-August 2006.

A.K. Roy and J. Pal, "Tensile Properties and Stress Corrosion Crack Growth Studies of Alloy C-276," AMPT Conference, Las Vegas, NV, July-August 2006.

A.K. Roy, J. Pal, R. Koripelli, A. Venkatesh, and J. Yelavarthi, "High-Temperature Tensile Properties of Nickel-base Alloys for Hydrogen Generation," SAMPE Conference, Long Beach, CA, April-May 2006.

A.K. Roy, R.S. Koripelli, and J. Pal, "The Corrosion Behavior of Nickel-base Austenitic Alloys for Nuclear Hydrogen Generation," MRS Spring Meeting, San Francisco, CA, April 2006.

A.K. Roy and V. Virupaksha "Performance of Alloy 800H at Elevated Temperature for Heat-Exchanger Application," TMS Conference, San Antonio, TX, March 2006.

A.K. Roy, A. Kaiparambil, and R. Santhanakrishnan, "Metallurgical and Corrosion Behavior of Refractory Materials for Hydrogen Generation," TMS Conference, San Antonio, TX, March 2006.

A.K. Roy and V. Virupaksha, "High-Temperature Deformation Characteristics and Corrosion Susceptibility of Alloy 800H," MRS Fall Meeting, Boston, MA, November 2005.

A.K. Roy, and A. Kaiparambil, "Use of Refractory Materials for Hydrogen Generation using Nuclear Power," MRS Fall Meeting, Boston, MA, November 2005.

A.K. Roy and V. Virupaksha, "Metallurgical and Corrosion Study of Alloy 800H," SAMPE Conference, Seattle, WA, November 2005.

A.K. Roy, A. Kaiparambil, and R. Santhanakrishnan, "Characterization of Materials for Hydrogen Generation by HIx Decomposition," SAMPE Conference, Seattle, WA, November 2005.

A.K. Roy, V. Virupaksha, J. Yelavarthi, "Use of Alloy 800H as a Heat-Exchanger Structural Material," AIChE Conference, Cincinnati, OH, November 2005.

A.K. Roy and J. Pal, "Tensile Properties and Corrosion Susceptibility of C-276 in S-I Environment," AIChE Conference, Cincinnati, OH, October 2005.

A.K. Roy, R. Santhanakrishnan, and A. Kaiparambil, “Characterization of Structural Materials for Nuclear Hydrogen Generation,” AIChE Conference, Cincinnati, OH, October 2005.

A.K. Roy, V. Virupaksha, and J. Yelavarthi, “Use of Alloy 800H as a Heat-Exchanger Material,” AIChE Conference, Cincinnati, OH, October 2005.

A.K. Roy, A. Kaiparambil, and R. Santhanakrishnan, “Characterization of Materials for Hydrogen Generation by HIx Decomposition,” SAMPE Conference, Seattle, WA, October 2005.

A.K. Roy and V. Virupaksha, “Metallurgical and Corrosion Study of Alloy 800H,” SAMPE Conference, Seattle, WA, October 2005.

A.K. Roy, R. Santhanakrishnan, A. Kaiparambil, B. Wong, G. Besenbruch and L. Brown, “Metallurgical and Corrosion Characterization of Structural Materials for the Nuclear Hydrogen Initiative,” MS&T 2005 Conference, Pittsburgh, PA, September 2005.

A.K. Roy and V. Virupaksha, “Characterization of Alloy 800H for Heat Exchanger Applications,” MS&T Conference, Pittsburgh, PA, September 2005.

A.K. Roy, L. Savalia, N. Kothapalli, and R. Karamcheti, “Mechanical Properties and Cracking Behavior of High-Temperature-Heat-Exchanger Materials,” ASME PVP 2005, Denver, CO, July 2005.

A.K. Roy, R Karamcheti, L. Savalia and N. Kothapalli, “Metallurgical Stability and Corrosion Behavior of Structural Materials for Hydrogen Generation,” ASTM Conference, Reno, NV, May 2005.

A.K. Roy, R. Karamcheti, L. Savalia, and N. Kothapalli, “Tensile Properties and Corrosion Characteristics of Heat-Exchanger Materials,” SAMPE Conference, Long Beach, CA, May 2005.

A.K. Roy, L. Savalia, N. Kothapalli and R Karamcheti, “High-Temperature Properties and Corrosion Behavior of Nickel-Base Alloys,” Corrosion/2005, NACE International, Paper No. 05430, April 2005, Houston, TX.

A. Kaiparambil and R. Santhanakrishnan, “Corrosion Behavior of Candidate Materials for Hydrogen Generation,” ANS Student Conference, April 2005, Columbus, OH

L. Savalia, N. Kothapalli and R. Karamcheti, “Corrosion and Metallurgical Behavior of Ni-Base Alloys for Heat-Exchanger Application,” ANS Student Conference, April 2005, Columbus, OH

A.K. Roy, L. Savalia, R. Karamcheti, and N. Kothapalli, “Mechanical properties and Cracking of High-Temperature Heat Exchanger Materials,” TMS Annual Meeting, San Francisco, CA, February 2005.

A.K. Roy, L. Savalia, N. Kothapalli and R Karamcheti, "High Temperature Properties of Structural Material for Hydrogen Generation," INMM Conference, Orlando, FL, July 2004.

Journal Articles:

A.K. Roy, R.S. Koripelli, and J. Pal, "Tensile Deformation of a Nickel-base Superalloy for Application in Hydrogen Generation," *International Journal of Hydrogen Energy*, Elsevier Science, in press.

A.K. Roy, A. Venkatesh, V. Marthandam, and A. Ghosh, "Tensile Deformation of a Nickel-base Alloy at Elevated Temperatures," *Journal of Materials Engineering and Performance,* ASM International, in press.

A.K. Roy, J. Pal and C.K. Mukhopadhyay, "Dynamic Strain Ageing of an Austenitic Superalloy – Temperature and Strain Rate Effects," *Materials Science and Engineering A*, Elsevier Science, Vol. 474, pp 363-370, 2008.

A.K. Roy and V. Virupaksha, "Performance of Alloy 800H for High-Temperature Heat Exchanger Applications," *Materials Science and Engineering A,* Elsevier Science, Vol. 452-453, pp. 665-672, 2007.

A.K. Roy and A. V. Kaiparambil, "Tensile and Corrosion Behavior of Zr705 for Nuclear Hydrogen Generation," *Materials Science and Engineering A*, Elsevier Science, Vol. 427, pp 320-326, 2006.

3.0 Construction Materials Screening for the HI Decomposition Process at GA (Bunsen Wong, Principal Investigator, General Atomics)

3.1 Introduction

In order to identify the material of construction candidates for the HI decomposition process, immersion corrosion coupon tests in HI_x ($HI + I_2 + H_2O$) were used to screen potential candidates. Materials that performed well in this test were put through developmental testing which will gauge material processing effects on their corrosion and mechanical properties. Two test systems capable of handling HI_x at the reaction temperature were set up at General Atomics. 22 different material coupons were screened at both the feed (262°C) and boiler (310°C) conditions for up to 120 hours from January 2004 through June 2005. The corrosion rate of the different materials has been determined.

The Nb- and Ta-based alloys all performed well and will undergo further testing. Another refractory, Mo, did not exhibit adequate corrosion resistance as it lost weight during testing. Weight loss of MoRe was acceptable but it is still not as good as Ta and Nb alloys. Both zirconium coupons, Zr702 and Zr705 showed signs of corrosion pitting and some dissolution. The normally corrosion resistant superalloys such as Hastelloy C-276 and Haynes 188 showed severe weight loss through dissolution. Hence, these materials are not suitable for this application.

The SiC based ceramic materials demonstrated that they are also suitable for HI decomposition heat exchanger applications. Three different varieties of SiC were tested: two sintered samples and one fabricated by Chemical Vapor Deposition (CVD). They all performed well in the liquid HI_x environment and underwent long-term testing. The Si-infiltrated SiC coupons also are resistant to HI_x liquid but there is minor absorption of HI_x by the specimens during the test.

Although the extruded graphite rod that was tested HI_x did maintain its structural integrity, it absorbed a large amount of liquid and resulted in a final weight gain of 20%. The application of graphite will be extremely limited.

From the screening results, Ta-2.5W, Ta-10W and Nb-10Hf were qualified as development candidates. First, they underwent long term immersion coupon testing. In addition, there materials will be subjected to stress corrosion environment to determine their susceptibility to crack initiation and growth.

Test systems are set up to accommodate the stress corrosion specimens. C-ring, and tensile specimens made from Zr705 were tested demonstrating the functionality of the test system. The Ta-2.5W, Ta-10W and Nb-10Hf specimens were subsequently tested in the system.

3.2 Research Highlights and Accomplishments

- Ta-2.5W, Ta-10W and Nb-10Hf were determined to be suitable for high temperature use in HI_x.

- Stress corrosion studies showed the dependence of mechanical properties on environment.
- Material test system for three different environments within extractive distillation were completed.
- Ta-2.5W has the best performance in HI_x + H_3PO_4.
- A total of six test systems were constructed to help qualify materials for the different HI_x and H_3PO_4 environments.
- The HI decomposition materials test system replicates the HI gaseous decomposition environment.
- The acid (HI_x; HI_x + H_3PO_4) circulating system enabled materials testing in a dynamic environment.
- In FY05/06, a total of 25 materials were screened in HI_x at 310°C; Ta and Nb alloys met the criteria.
- In FY06/07, additional construction materials for the different environments were identified.
- 13 different materials were tested in the static Iodine Separation (HI_x+H_3PO_4) environment.
- Materials that can handle HIx at high temperature may not be suitable for Iodine Separation.
- 11 candidates were tested in boiling 95 wt% phosphoric acid.
- The addition of contaminants (I_2, HI) lead to unanticipated corrosion of materials.
- Hastelloys showed the best corrosion performance in the HI decomposition environment (HI + I_2 + H_2).
- Ta coated components can be an effective means to reduce equipment cost.
- Performance of Ta coated components such as fittings and valves were also evaluated.
- Ta, Ta-2.5W, Ta-10W and SiC have been qualified for use and further testing in the the liquid streams in Section III. Hastelloy B2, C22 and C276 all met the performance criteria for use in the HI decomposition environment.
- Stress corrosion studies of Hastelloy specimens in the HI gaseous decomposition environment have not shown any stress corrosion effect.
- It was demonstrated that Ta coated parts can be a viable means to manufacturing of heat exchanger and process components.
- Testing of PTFE and FEP rods in the boiling HI distillation mixture (185°C) was completed. There is iodine penetration in both specimens but no observable weight change and physical appearance has been measured. Hardness measurement did not show any noticeable changes in the samples.
- Testing of the PTFA specimen rod in iodine separation circulation bath (I_2 and HI + conc. phosphoric acid) was completed. Similar to what was observed in the specimens tested in the HI distillation liquid, penetration of iodine into the specimen has been observed after 1120 hours of testing. No physical change in dimension has been measured.

3.3 Project Profile

Start Date: October 2003

End Date: April 2008

Researchers: B. Wong (Principal Investigator), G.E. Besenbruch, J.M. Bolin, L.C. Brown, and M.P. LaBar

Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):

B. Wong, L.C. Brown, and G.E. Besenbruch, "Ta Cladding and Coating for Scale Up of the Sulfur Iodine Hydrogen Production Loop," Technical Report, GA-C25875, July 2007.

B. Wong, "Corrosion and Crack Growth Studies of Heat Exchanger Construction Materials for HI Decomposition in the Sulfur-Iodine Hydrogen Cycle," Poster, DOE Annual Review, PDP31, May 15, 2007.

B. Wong, L.C. Brown, and G.E. Besenbruch, "Corrosion Studies of Construction Materials in HI Decomposition Environment," FY2006 Year End Report, GA-C25728, April 2007.

B. Wong, L.C. Brown, and G.E. Besenbruch, "Effects of Chemical Contaminants, Chemical Flow, and Stress on the Corrosion Properties of Structural Materials Exposed to HI_x Environments," FY2006 Year End Report, GA-C25727, April 2007.

B. Wong, L.C. Brown, and G.E. Besenbruch, "Construction of a HI_X Circulation Loop for Testing of Process Components such as Valves and Pump," Milestone Completion Report, GA-C25628, October 2006.

"Interim Report on Stress Corrosion Effect of an HI_x Environment on Crack Growth," May 31, 2006.

G. Besenbruch, L. Brown, B. Buckingham, and B. Russ, "HI Decomposition Demonstration Experiments for the Sulfur Iodine Cycle," Draft Status Report, September 15, 2005.

B. Wong, L.C. Brown, and G.E. Besenbruch, "Effect of Long-term Immersion and Material Processing on the Corrosion Performance of Heat Exchanger Construction Materials for Hydrogen Iodide Decomposition," Progress Report, GA-C25223, June through September, 2005.

B. Wong, L.C. Brown, and G.E. Besenbruch, "Corrosion Screening of Construction Materials for Hydrogen Iodide Decomposition Heat Exchanger Fabrication," Milestone Completion Report, January 2004 through June 2005, GA-A25128, July 2005.

B. Wong, L.C. Brown, R. Buckingham, G.E. Besenbruch, G. Polansky, and P. Pickard, "Engineering Materials Requirements Assessment for the S-I Thermochemical Cycle," GA-A24902 and SAND2004-681, February 2005 and April 2005.

B. Wong, L. Brown, and G. Besenbruch, "Complete Installation of Test System for Corrosion Studies of Materials in HI Extractive Distillation Environment (Gaseous HI

Decomposition),” Draft Milestone Completion Report, March 30, 2005.

J.M. Bolin and M.P. LaBar, “High Temperature Heat Exchanger Study,” Final Report of work performed from contract beginning October 20, 2003 to contract end December 31, 2004, PC-000523, Rev 0, December 23, 2004.

Conference Proceedings:

B. Wong, L. Brown, R. Buckingham, A. Kaiparambil, R. Santhanakrishnan, B. Russ, A. Roy, and G. Besenbruch, “Construction Material Development in Sulfur-Iodine Thermochemical Water-Splitting Process for Hydrogen Production,” AIChE Spring National Meeting, Atlanta, GA, April 10-14, 2005.

B. Wong, B. Buckingham, L. Brown, J. Gomez, B. Russ, and G. Besenbruch, “Materials Challenges in Sulfur-Iodine Thermochemical Water Splitting process for Hydrogen Production,” ASM Conference, Columbus, OH, October 20, 2004.

Journal Articles:

B. Wong, R.T. Buckingham, L.C. Brown, B.E. Russ, G.E. Besenbruch, A. Kaiparambil, R. Santhanakrishnan, and Ajit Roy, “Construction materials development in sulfur–iodine thermochemical water-splitting process for hydrogen production,” *International Journal of Hydrogen Energy*, Vol 32 (4), pp 497-504, March 2007.

4.0 Ceramic Heat Exchanger Development for Application to NHI at Ceramatec, Inc. (Merrill Wilson, Principal Investigator, Ceramatec, Inc.)

4.1 Introduction

To meet cost targets, candidate ceramic materials must have relatively low bulk costs, and fabrication methods must extrapolate to low-cost mass manufacturing. Sintered silicon carbide materials have relatively low bulk costs and Ceramatec, Inc. has demonstrated a process for fabrication using a tape-casting, lamination and sintering methods that extrapolate to low-cost production. These fabrication methods are very scalable and have process flexibility with several candidate materials. This allows the heat exchanger designs to be optimized for cost and lifetime performance.

Construction Materials Screening

One of the most imperative obstacles facing materials selection for activities involving hydrogen production by thermochemical water splitting is that little is known about how materials will react to an environment containing sulfuric acid decomposition products. This lack of knowledge is an obstacle since this type of an environment must be endured and, more importantly, contained during a portion of the hydrogen production process. A few research studies located during a literature search displayed results that helped to narrow the groups of materials that would be appropriate candidates for further materials testing.

Heat Exchanger Designs

Within the Sulfur-Iodide process, the sulfuric acid is catalytically decomposed to SO_2, SO_3, O_2 and H_2O at temperatures between about 700°C and 900°C. These high temperatures and oxidizing conditions require corrosion resistant materials. The catalytic reactions require high surface to volume ratios and efficient heat transfer. This has lead to a shell and plate heat exchanger design (decomposer). These plates are fabricated with ceramic micro-channels, enabling efficient catalyst loading and highly integrated heat exchange.

Based on this conceptual design, the micro-channels and overall plate design were analyzed for performance optimization. The design variables included the micro-channel width, depth and pitch, the geometry of the internal flow distribution manifold, the thermal conductivity of the solid and the mass flow rate within the flow streams. From these analyses it was predicted that sulfuric acid feed stream could be sufficiently heated by the counter-flowing decomposed sulfuric acid stream and the hot helium stream. These results also indicated that the effectiveness of the heat exchanger is improved as the cross-sectional area is decreased or thermal conductivity is reduced; thus, minimizing the axial thermal dispersion – typical of micro-channel devices. Thermo-mechanical analyses are underway in order to determine the operational mechanical stresses in a heat exchanger plate.

4.2 Research Highlights and Research Accomplishments

- Based on material, mechanical and corrosion properties screening, candidate ceramic materials have been selected for sulfuric acid decomposition heat exchangers. The preferred materials are: silicon carbide, silicon nitride, sialon (non-oxide ceramics) and alumina (oxide ceramic).
- Preferred candidate materials were tested for 1000 hours in steam/H_2SO_4/air environments at 900°C.
- Mechanical Strength: Silicon Carbide & Silicon Nitride – all increased strength; Alumina – decreased strength.
- Weight Gain: Very slight weight gains < 0.1%/1000 hrs. Silicon Carbide & Silicon Nitride form stable "healing" SiO_2 layer. Alumina has corrosion products that are to be determined.
- Compact shell and plate heat exchanger was designed with high performance micro-channels for low volume and low cost.
- Flow/heat transfer coupons designed, fabricated, and tested.

4.3 Project Profile

Start Date: October 2003

End Date: October 2007

Researchers: M. Wilson (Principal Investigator), J. Cutts, and C. Lewinsohn

Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):

M. Wilson, "FY2007 Materials Development Report," Milestone/Deliverable Completion Report, Ceramatec, Inc., August 3, 2007.

M. Wilson, "FY2007 HX Scale-up Report," Milestone/Deliverable Completion Report, Ceramatec, Inc., August 3, 2007.

M. Wilson, "FY2007 Full-Size Wafer Sample," Milestone Completion Report, Ceramatec, Inc., August 3, 2007.

M. Wilson, "FY2006 Materials Characterization and Heat Exchanger Design Development for NHI Applications," FY2006 Annual Report, Ceramatec, Inc., October 23, 2006.

Conference Proceedings:

M. Wilson, C. Lewinsohn, J. Cutts, Y. Chen, and V. Ponyavin, "Design of a Ceramic Heat Exchanger for Sulfuric Acid Decomposition," *Proceedings*, ICONE15, 15th International Conference on Nuclear Engineering, ICONE15-10788, April 22-26, Nagoya, Japan, 2007.

M. Wilson, C. Lewinsohn, and J. Cutts, "Design Considerations for High Temperature, Ceramic Heat Exchangers," *Proceedings*, 2007 ASME-JSME Thermal Engineering Summer Heat Transfer Conference, HT2007-32229, Vancouver, Canada, July 8-12, 2007.

M. Wilson, C. Lewinsohn, J. Cutts, Y. Chen, and V. Ponyavin, "Optimization of Micro-channel Features in a Ceramic Heat Exchanger," *Proceedings*, 2007 ASME-JSME Thermal Engineering Summer Heat Transfer Conference, HT2007-3181, Vancouver, Canada, July 8-12, 2007.

5.0 Materials, Design, and Modeling for C/SiC Ceramic Heat Exchangers at UC Berkeley (Per Peterson, Principal Investigator, UCB Nuclear Engineering Department)

5.1 Introduction

To meet cost targets, candidate ceramic materials must have relatively low bulk costs, and fabrication methods must extrapolate to low-cost mass manufacturing. Researchers at the University of California, Berkeley surveyed candidate heat exchanger materials and fabrication methods, with a focus on low-cost chopped-carbon fiber reinforced silicon carbide, where the silicon carbide is infiltrated by either melt (MI) or polymer infiltration and pyrolysis, and where additional low-cost filler materials may be used.

Research conducted at UC Berkeley focused on inexpensive chopped carbon fiber based ceramic composite. Helium permeation testing and mechanical property testing were conducted. Young's moduli of carbon fiber reinforced SiSiC (FR-SiSiC), splint based SiSiC (SB-SiSiC) and pitch based carbon fiber reinforced SiSiC (BioKer) were tested by using a strain gauge.

<u>Heat Exchanger Designs</u>

Mechanical and initial thermal stress analyses were completed for an offset strip-fin hybrid plate type compact ceramic heat exchanger by UCB. The heat exchanger is made from liquid silicon impregnated carbon composite. The study was conducted with helium gas and molten salt as the working fluids with varied channel dimensions. Thermal and stress analysis were performed to identify optimal designs for core heat transfer unit, inlet and outlet manifolds, distribution channels, and channel dividers. Finding detailed stress distribution in a complete heat exchanger with direct FEM (Finite Element Method) requires an order of a billion FEM computation units and millions of hours computing time. Therefore, it is not practical to analyze the entire heat exchanger design directly. UCB proposes an alternative method to obtain approximate stresses that only requires several days to finish in a fast computer. The methods are composed of three steps. First, the heat exchanger is broken down into several regions. Unit cell models are built based on each region that captures all of the most important features of that region. The effective mechanical and thermal properties for each unit cell are then founded through FEM simulations. Second, average stress distribution in an overall model composed of various unit cell regions is computed by using the effective mechanical and thermal properties. Third, these average stress values are then applied to the unit cells to find localized points of high stresses. Pro/Mechanica module (Pro/M) in the Pro/E Wildfire Edition was used for FEM stress analysis.

<u>Heat Exchanger Safety</u>

One of most important viability and safety issues to use liquid salt as an intermediate heat transfer fluid is chemical control. Ingress of sulfuric acid from hydrogen product loops into liquid salt results in strong corrosion of chromium from high-temperature metals and HF generation. UCB has checked various traditional approaches to salt chemistry control and concluded that improved methods could bring benefits. After examining aluminum and rare-earth chemistry in flinak, which has been extensively studied for the application to metallic aluminum production, new potential chemistry control approaches for liquid salt have been identified. These involve dissolving small amounts of aluminum or rare earths in the salt, to control oxygen potential, and contacting the salt with metallic sodium (and possibly small amounts of potassium) to control fluorine and sulfur potentials. The additives react with and consume sulfuric acid so that it is possible to permit the safe use of liquid salts for high-temperature heat transport without double-wall S-I process heat exchangers.

5.2 Research Highlights and Accomplishments

- High pressure helium permeation testing verified very good helium hermeticity of several types of melt infiltrated (MI) composite materials; Young's moduli of three types of MI materials were measured.
- Methods to make heat exchanger plates from chopped carbon fiber reinforced SiC composite material with polymer infiltration and pyrolysis (PIP) process were surveyed. Experimental trials were performed with die embossed methods.
- Thermal and stress analysis for the baseline ceramic compact off-set strip fin heat exchanger were performed to identify optimal designs for core heat transfer unit, inlet and outlet manifolds, distribution channels, and channel dividers. High stresses regions were found.
- An initial survey of liquid fluoride salt additives for chemical control was completed. The additives react with and consume sulfuric acid so that it is possible to permit the safe use of liquid salts for high-temperature heat transport without double-wall S-I process heat exchangers.
- It is possible to safely use liquid salts for high-temperature heat transport without double-wall S-I process heat exchangers with proper chemical control. Studied use of reactive metal control for flinak coolant to mitigate effects of process fluid ingress.
- Completed detailed design for ceramic compact helium-to-liquid-salt HX for thermal/mechanical analysis.
- Demonstrated fabrication of mm-scale fins with thru and blind (preferred) reusable teflon molds.

5.3 Project Profile

Start Date: October 2003

End Date: October 2007

Researchers: P.F. Peterson (Principal Investigator), P. Bardet, F. Niu, J. Schmidt, and H. Zhao

Students: K. Chen, D. Damba, E.U. Fernández, G. Fukuda, D. Huang, W. Huang, A. Jousse, B. Laurenty, K. Lee, T. Ma, A. Mai, A. Niquille, and E. Wendel

<u>Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):</u>

P.F. Peterson and E.U. Fernández, "Intermediate Heat Exchanger Dynamic Thermal Response Model," Project Report, University of California, Berkeley, CA, August 31, 2007.

P.F. Peterson, A. Niquille, and E.U. Fernández, "Intermediate Heat Exchanger Dynamic Thermal Response Model," Interim Report, Report UCBTH-07-004, University of California, Berkeley, CA, May 31, 2007.

P.F. Peterson, H. Zhao, E.U. Fernández, W. Huang, and K. Lee, "C/SiC Heat Exchanger Safety Analysis," Final Report, University of California, Berkeley, CA, May 29, 2007.

P.F. Peterson, "Capillary Tube and Shell Heat Exchanger Design for Helium to Liquid Salt Heat Transfer," Report UCBTH-07-003, University of California, Berkeley, CA, May 7, 2007.

P.F. Peterson, "Guidelines for Trade-Studies for Comparison of Liquid Salt and Helium Intermediate Coolants for the NGNP," Report UCBTH-07-002, Rev. A, University of California, Berkeley, CA, February 20, 2007.

B. Laurenty, "The LM-LS experiment: investigating corrosion control, in Liquid Fluoride Salts, by Liquid Alkali Metal," Report UCBTH-06-002, University of California, Berkeley, CA, January 2006.

J. Schmidt, "Potential Applications for Liquid Silicon Melt Infiltrated (LSI) Composites to the High-Temperature Sulfur-Iodine Process," Visiting Scholar Technical Report, UCBTH-06-004, University of California, Berkeley, CA, October – December 2006.

P.F. Peterson, H. Zhao, and G. Fukuda, "Functional Requirements Overview For a 50-MW(t) Liquid-Salt Intermediate Loop for NGNP," Report UCBTH-05-007, U.C. Berkeley, December 29, 2005.

P.F. Peterson, H. Zhao, F. Niu, G. Fukuda, W. Huang, A. Szweda, T. Easler, and R. Plunkett, "C/SiC Composite Fabrication Methods and Test Results," Deliverable Report, University of California, Berkeley, CA, October 20, 2005.

W. Huang, P.F. Peterson, and H. Zhao, "C/SiC HX Thermal and Mechanical Analysis," Deliverable Report, UCBTH-05-004, University of California, Berkeley, CA, September 30, 2005.

P.F. Peterson, H. Zhao, B. Laurenty, and G. Fukuda, "C/SiC Heat Exchanger Heat Transfer and Safety Analysis," Report UCBTH-05-006, U.C. Berkeley, September 30, 2005.

P.F. Peterson, H. Zhao, F. Niu, G. Fukuda, W. Huang, A. Szweda, T. Easler, and R. Plunkett, "Survey of Chopped Carbon Fiber and Matrix Materials for C/SiC Composite Heat Exchangers," Deliverable Report, UCBTH-05-003, University of California, Berkeley, CA, August 1, 2005.

P.F. Peterson, H. Zhao, and G. Fukuda, "Comparison of Molten Salt and High-Pressure Helium for the NGNP Intermediate Heat Transfer Fluid," Report UCBTH-03-004, U.C. Berkeley, December 5, 2003.

Theses Generated:

Alexandre Jousse, “Tritium Transport in Very High Temperature Reactors for Hydrogen Production,” M.S. degree requirement in Nuclear Engineering of the Grenoble National Engineering School for Physics, Report UCBTH07-005, University of California, Berkeley, Department of Nuclear Engineering, August 31, 2007.

Conference Proceedings:

W. Huang, H. Zhao, and P.F. Peterson, “Multi-scale Stress Analysis for Compact Plate Heat Exchangers,” 2006 International Congress on Advances in Nuclear Power Plants (ICAPP '06), Reno, NV, June 4-8, 2006.

P. Bardet and P.F. Peterson, “Design of Scaled Integral Experiments for High Temperature Liquid Salt and Helium Fluid Mechanics and Heat Transfer,” Eleventh International Topical Meeting on Nuclear Reactor Thermal Hydraulics, NURETH 11, Avignon, France, October 2-6, 2005.

B. Laurenty, G. Fukuda, D. Damba, and P. Peterson, “Inhibiting Corrosion By Molten Fluoride Salts: Investigations On Flinak,” AIChE, Cincinnati, OH, October, 2005. (Selected for AIChE Nuclear Engineering Division Student Award for Best Paper)

6.0 Development of Self-Catalytic Materials at MIT (Ronald Ballinger, Principal Investigator, MIT Departments of Materials and Nuclear Engineering)

6.1 Introduction

The purpose of the "catalyst" task in the project is to: (1) determine the viability of incorporating the catalyst and heat exchange (heat addition) functions into a single component for the thermal decomposition of SO_3 to SO_2 as a part of the SI hydrogen production process, (2) if viability is determined-design and fabricate a test "article" heat exchanger using the developed self catalytic materials, (3) evaluate the catalyst and heat exchange functions of the test article.

The approach taken is to add a catalytic function, through the addition of platinum, to two existing high strength, corrosion resistant materials that are already candidate materials for the high temperature SI heat exchanger. The two alloys chosen were Alloy 800 and Alloy 617. These materials are currently ASME code qualified for use under Section VIII of the code. Small amounts of platinum have been added to these base compositions and are being evaluated for their catalytic effectiveness. Pending the results of these tests, currently very positive, a test article heat exchanger will be designed and built using the final compositions that are decided upon. Extensive mechanical properties evaluation will also be conducted to verify adequate mechanical properties.

6.2 Research Highlights and Accomplishments

In the first year of the project several alloys based on the 800 and 617 systems were produced as small "button" heats for evaluation. To these alloys were added 2, 5, 15 and 30 wt% Pt. Characterization was begun during the first year of the project.

Alloy Development

- The characterization of the button heats was completed with very positive results. The addition of Pt was successful. Based on the results of the button characterization, the base chemistries to be used for the remainder of the program were determined.
- A series of Alloy 800H plus Pt and Alloy 617 plus Pt alloys in button form were melted and characterized from a metallurgical standpoint.
- Five heats each of Alloy 800 H + Pt chemistry, and Alloy 617 + Pt chemistry were received from Special Metals Inc. Platinum additions were made to the base alloys in nominal amounts of 1 wt%, 2 wt%, and 5 wt% (5 wt% in Alloy 800H only).
- Based on the previous work, Alloy 800H + Pt chemistry materials were etched with a weak version of glyceregia. For Alloy 617 + Pt chemistry materials, stronger version of glyceregia was used. Each material was etched in 1 minute intervals interrupted with ultrasonic cleansing in an ethanol bath in order to periodically monitor microstructure development via optical magnification at 100 X.
- The microstructures in the as cast form were developed via optical microscopy
- When compared to the earlier heats of material, which contained an unexpectedly high carbon content, the new alloys are much cleaner and represent what would be achievable in commercial practice.

- The samples for SEM analysis were intentionally over-etched to facilitate SEM analysis so the exaggerated etching of some of the micrographs is an artifact of this process.

Catalyst Effectiveness System

- A system for the determination of the catalytic effectiveness of the Pt-added alloys was designed and constructed. Sulfuric acid was first decomposed and then supplied to the catalyst "bed" in a high temperature furnace. The effectiveness of the system for the promotion of the SO_3 to SO_2 reaction will be evaluated by analyzing the reaction products in a gas chromatograph system.
- The catalyst facility is in operation.
- The results of this task have shown proof of principle and have shown that the Pt-added alloys are catalytic.

Compact Heat Exchanger Design

A preliminary design for the compact heat exchanger was completed. The design choice is between a "compact" plate-fin design and a printed circuit design. The design must be able to fit into the existing testing facilities for the program. MIT and Sandia National Laboratory have testing facilities for catalytic effectiveness (MIT) and full heat exchanger effectiveness (Sandia).

6.3 Project Profile

Start Date: October 2003

End Date: December 2007

Research Staff: R.G. Ballinger (Principal Investigator) and J. Lim

Students: K. Miu and D.A. Rigual

Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):

R.G. Ballinger, "Overall Self Catalytic Alloy Development Topical Report," Milestone Completion Report, Massachusetts Institute of Technology, Cambridge, MA, July 17, 2008.

Theses Generated:

Kevin Miu, "The Development of Autocatalytic Structural Materials for Use in the Sulfur-Iodine Process for the Production of Hydrogen," B.S. Thesis, Department of Nuclear Engineering and Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, MA, May 2006.

David A. Rigual, "Metallurgical Characterization of Self Catalytic Structural Materials for Sulfuric Acid Decomposition," M.S. Thesis, Department of Nuclear Engineering, Massachusetts Institute of Technology, Cambridge, MA, May 2005.

7.0 Heat Exchanger Design at UNLV (Yitung Chen, Principal Investigator, UNLV Mechanical Engineering Department)

7.1 Introduction

The Nuclear Hydrogen Initiative Research and Development Plan identified the sulfur cycles as a promising cycles for use with nuclear heating. One of the most complicated parts of the cycles is sulfuric acid decomposition due to high temperature and corrosive environment. Although the functional requirements for the sulfuric acid decomposer are straightforward, engineering an efficient, long-lived and cost effective acid decomposition apparatus for this environment is challenging. The several design types of the decomposer have been proposed. This project designed and analyzed high temperature heat exchangers and chemical decomposers of different designs in order to assess the operating performance, reliability, technical risks and the "end-game" economics. The oxygen chiller will be used for cooling of oxygen as produced by high temperature electrolysis. This process includes two unit processes: a) water boiler and b) oxygen pre-cooler. Sulfuric acid boiler can be integrated into the intermediate labs scale (ILS) demonstration unit for the hydrogen production. Although the existent designs of the boiler may be effective, improvements to the design can eliminate near term (expensive) solutions for seals etc, such that the system would become more efficient and economic for commercial practice.

7.2 Research Highlights and Accomplishments

<u>Design of Offset Strip Fin</u>

The investigations on the effect of manufacturing on the geometry (such as fillets) and optimization studies for the offset strip fin heat exchanger design have been accomplished to study the effect of channel geometry on the pressure drop and thermal performance. The study was conducted with helium gas and liquid salt (LS) as the working fluids with varied channel dimensions. The analyses were performed for both curved and rectangular fin edge channel designs using an incompressible fluid flow model, constant physical properties, and k-omega turbulence model for the helium side wherever necessary.

<u>Design Including Manufacturing Geometrical Effects</u>

For the turbulent case the influence of the fillets is very strong for the pressure drop on the LS side (17%) and He side (26%). But the influence is almost negligible (less than 3%) for the thermal performance.

<u>Optimization Studies</u>

The baseline design parameters are kept as the reference values, and the values of the changed parameters were varied below and above the baseline parameter values for optimization studies. The studies were also performed in such a way that for all designs the overall dimensions such as the length, width, and the height are almost the same. Since these studies would require a lot of computational time all the simulations were carried out using a reduced overall length. The fin thickness, gap-length, and fin length values influenced the pressure drop performance very significantly, but were insignificant on their effect on thermal performance. The effects of varying channel heights and pitch in x-direction were very significant on both the pressure drop and thermal performance.

Stress Analysis

The dimensions of the investigated off-set strip fin high temperature heat exchanger are 0.9m×1m×1m. A few modules were used because of the symmetry in the computational domain. The material of fiber reinforced SiC has been used in the numerical model.

To perform the stress analysis, the needed boundary conditions for the temperature and pressure were obtained from CFD simulations of the heat exchanger. The pressure at the He side is 7 MPa and at the LS side is 0.1 MPa. The temperature distribution was chosen as uniform for both the He and LS sides and the temperature difference between both sides is 100K. The temperature at LS side was set to 900 K while the temperature at the He side was 1000K. Von Mises stresses were computed for both thermal and mechanical stresses.

Analysis of Sulfur Trioxide Decomposition Heat Exchanger

A cross-flow type of heat exchanger was chosen for the analysis of the decomposition of sulfur trioxide. Initial work has been carried out on a single reactor tube to study the different parameters affecting the decomposition of sulfur trioxide. A reactor tubes have a 12.7 mm diameter and 500 mm length. The computational mesh was created using GAMBIT 2.1 and numerical simulation was performed using FLUENT 6.2 on a two-dimensional numerical model after applying proper boundary conditions. Catalyst ALFA-4 properties were used instead of platinum because of unavailability of data related to platinum. Different simulation cases are studied to find the variation of decomposition of sulfur trioxide with the wall surface temperatures. It was observed that the percentage decomposition of sulfur trioxide is increasing with the increase of tube wall surface temperature. But the decomposition of sulfur trioxide is very low even at higher temperatures because there is a very high percentage of undecomposed sulfur trioxide remaining at the center of the reactor tube. The smaller diameter reactor tube would provide higher decomposition rates because of increased surface area at a specific given mass flow rate.

A total of 44 numerical cases were run from 500 to 1000^{o}C with an interval of 50^{o}C to study the variation in the percentage decomposition of sulfur trioxide for different reactor tube diameters and wall surface temperatures. The diameters selected for the analyses are 1, 2, 3 and 4 mm and the length of the reactor tube is kept constant at 500 mm. The decomposition of sulfur trioxide varies with the mole flow rate. The different simulation cases were also studied for the different mole flow rates of the sulfuric acid in the decomposer.

Idaho National Laboratory (INL) provided the necessary chemical and physical properties of platinum for the numerical catalytic reaction model. A reactor tube of diameter 1 mm and length 500 mm is used. The experimental results were also provided by at INL. Experimental work was compared with numerical results for Pt catalyst. It shows a fair agreement between experimental and numerical results.

7.3 Project Profile

Start Date: October 2003

End Date: November 2007

Research Staff: Y. Chen (Principal Investigator), R. Akberov, A.E. Hechanova, and M. Trabia

Students: C.R. De Losier, G. Kuchi, T. Mohamed, K. Muramalla, V. Nagarajan, V. Ponyavin, and S. Subramanian

<u>Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):</u>

S. Subramanian, V. Ponyavin, C.R. DeLosier, Y. Chen, and A.E. Hechanova "Design Considerations for Compact Ceramic Offset Strip-Fin High Temperature Heat Exchangers," *Advances in Compact Heat Exchangers*, Editors: B. Sunden, R.K. Shah. R.T. Edwards, Inc., Philadelphia, 2007

A. Hechanova, Y. Chen, V. Ponyavin, and V. Nagarajan, "Computational Analyses of the Sandia Bayonet Design for the Sulfuric Acid Decomposer," Milestone Completion Report, University of Nevada, Las Vegas, NV, May 14, 2007.

<u>Theses Generated:</u>

Kiran Kumar Muramalla, "Simulation of Decomposition of Sulfur Trioxide Gas on a Self-Catalytic Material," M.S. Thesis, Department of Mechanical Engineering, UNLV, December 2005.

Sundaresan Subramanian, "CFD Modeling of Compact Offset Strip-fin High Temperature Heat Exchanger," M.S. Thesis, Department of Mechanical Engineering, UNLV, August 2005.

<u>Conference Proceedings:</u>

Nagarajan, V., V. Ponyavin, Y. Chen, M. Vernon, P. Pickard, and A.E. Hechanova, "Numerical Study of Sulfur Trioxide Decomposition in a Bayonet Type Heat Exchanger and Chemical Decomposer," *Proceedings*, 2007 AIChE Annual Meeting, Salt Lake City, Utah, November 4-9, 2007.

G. Kuchi, V. Ponyavin, Y. Chen, S. Sherman, and A.E. Hechanova, "Modeling of High Temperature Shell and Tube Heat Exchanger and Decomposer for Hydrogen Production," *Proceedings*, 2007 AIChE Annual Meeting, Salt Lake City, Utah, November 4-9, 2007.

V. Nagarajan, V. Ponyavin, Y. Chen, M. Vernon, P. Pickard, and A.E. Hechanova, "CFD Modeling of Bayonet Type High Temperature Heat Exchanger and Chemical Decomposer with Different Packed Bed Designs," *Proceedings,* 2007 ASME International Mechanical

Engineering Congress and Exposition (IMECE2007), November 11-15, Seattle, Washington, USA, 2007.

G. Kuchi, V. Ponyavin, Y. Chen, S. Sherman, and A.E. Hechanova, "Flow Distribution on the Tube Side of a High Temperature Heat Exchanger and Chemical Decomposer," *Proceedings*, 2007 ASME International Mechanical Engineering Congress and Exposition (IMECE2007), November 11-15, Seattle, WA, 2007.

V. Ponyavin, Y. Chen, A.E. Hechanova, and M. Wilson, "Fluid/Thermal Analysis of High Temperature Heat Exchanger and Chemical Decomposer for Hydrogen Production," *Proceedings*, 2007 ASME-JSME Thermal Engineering Summer Heat Transfer Conference, July 8-12, Vancouver, Canada, 2007.

S. Subramanian, V Ponyavin, C.R. DeLosier, Y.T. Chen, A.E. Hechanova, and P.F. Peterson, "The Effect of Fin Geometry on design of Compact Offset Strip-Fin High Temperature Heat Exchanger," *Proceedings*, ASME International Mechanical Engineering Congress and Exposition 2005, Orlando, FL, November 2005

V. Ponyavin, S. Subramanian, C.R. DeLosier, Y.T. Chen, A.E. Hechanova, and P.F. Peterson, "Stress Analysis of an High temperature Heat Exchanger Used in an Advanced Nuclear Reactor," *Proceedings*, ASME International Mechanical Engineering Congress and Exposition 2005, Orlando, FL, November 2005

K.K. Muramalla, Y.T. Chen and A.E. Hechanova, "Simulation and Optimization of Homogeneous Decomposition of Sulfur Trioxide Gas on a Catalytic Surface," *Proceedings*, ASME International Mechanical Engineering Congress and Exposition 2005, Orlando, FL, November 2005

K.K. Muramalla, Y.T. Chen, and A.E. Hechanova, "Simulation of Decomposition of Sulfur Trioxide Gas on Self-Catalytic Metallic Material," AIChE Annual Meeting 2005, Cincinnati Convention Center, Cincinnati, OH, October 30 - November 4, 2005

S. Subramanian, V. Ponyavin, C.R. DeLosier, Y.T. Chen, and A.E. Hechanova, "Design Considerations for Compact Ceramic Offset Strip-Fin High Temperature Heat Exchangers," *Proceedings*, Fifth international Conference of Enhanced Compact and Ultra-Compact Heat Exchangers: Science, Engineering and Technology, Whistler, British Columbia, Canada, September 2005

V. Ponyavin, S. Subramanian, C.R. DeLosier, Y.T. Chen, A.E. Hechanova and P.F. Peterson, "Flow Calculations in High Temperature Heat Exchanger with Manufacturing Geometrical Effects," *Proceedings*, HT-2005 ASME Summer Heat Transfer Conference 2005, San Francisco, CA, July 2005

S. Subramanian, V. Ponyavin, C.R. DeLosier, Y.T. Chen, A.E. Hechanova and P.F. Peterson, "Design Considerations for Compact Ceramic Offset Strip-Fin High Temperature Heat Exchangers," *Proceedings*, GT2005 ASME Turbo Expo 2005, Reno, NV, June 2005

S. Subramanian and C.R. DeLosier, "CFD Simulation of an Offset Strip-Fin Heat exchanger," ANS Students Conference 2005, Columbus, OH, April 2005

K.K. Muramalla, "Simulation of Homogeneous Decomposition of Sulfur Trioxide Gas in a Wall Catalyzed Reactor Tube," ANS Students Conference 2005, Columbus, OH, April 2005

Journal Articles:

V. Ponyavin, Y. Chen, T. Mohamed, M. Trabia, A.E. Hechanova, and M. Wilson, "Parametric Study of Sulfuric Acid Decomposer for Hydrogen Production," *Progress in Nuclear Energy*, in press.

DeLosier, C.R., S. Subramanian, V. Ponyavin, Y. Chen, A.E. Hechanova, and P. Peterson, "The Parametric Study of an Innovative Offset Strip-fin Heat Exchanger," *Journal of Heat* Transfer, in press.

8.0 Experimental Facility Design for the Testing of Prototype Components at UNLV (Samir Moujaes, Principal Investigator, UNLV Mechanical Engineering Department)

8.1 Introduction

This study focused on the pressure drop (friction factor) and heat transfer (Nusselt number) across the heat exchanger core. These are important parameters with regards to the performance of the heat exchanger. For a given flow rate, a larger friction factor will means a larger pressure drop which increases the required pump power larger and operating cost; and a larger Nusselt number will means larger heat transfer coefficient which enhances the convection heat transfer. The enhanced heat transfer in compact heat exchangers is usually accompanied by increased pressure losses.

During the 1.5 year experimental period, ten test sections were designed, built, and tested with several testing fluid media to simulate as closely as possible the proposed high pressure helium and the molten salt for the proposed IHX unit as proposed by UC Berkley. More than 800 hours were used to construct and set up the testing loops. More than 300 hours' were employed in performing the tests. A large number of data were collected and processed to evaluate the hydraulic and thermal performance of the modeled IHX. CFD simulations with COSMOL and FLUENT were performed to compare and verify the experimental results.

To simulate the prototype IHX properly, prototype and experimental model dimension ratio was set to be 1:3or 1:2 to have reasonable flow velocities and minimize the natural convection effect. The testing media were room air to simulate pressurized helium gas and silicone oils of 2 cSt and 5 cSt to simulate molten salts. The flow rates of the testing fluids were controlled in a way to cover the prototype IHX Reynolds number ranges. The construction materials used were Alloy 6061 and acrylic sheets because of their ease of machining and their availability. There were single-chamber test sections for each testing fluid, and one double-chamber heating test section with DI (deionized) water and room air. A closer match of the Re was made in these simulated tests to the fluid flow conditions than the heating capacity as it was somewhat difficult to match both simultaneously. Isothermal hydraulic tests with the single-chamber test sections for the friction factors using the silicone oils and room air were initially performed followed by thermal-hydraulic tests for room air. Nusselt numbers were calculated from the experiments performed in the single-chamber test sections for the room air and 5 cSt silicone oil with electric heating pads, and the double-chamber test section with the heated DI water being circulated using a constant temperature water bath.

8.2 Research Highlights and Accomplishments

- Modeling was completed and prototype (1:3 scale for air and 1:2 for silicon oil) test sections were built with Alloy 6061 and Acrylic sheets (with square-edge-fins and round-edge-fins).
- Air to simulate He and silicon oil to simulate liquid salt were the testing fluids.
- Isothermal and heated tests with air were performed to measure friction factors. Friction factors were calculated and compared with that of the square-edge-fins. The volumetric

flow rate was varied and corresponded to a range of Reynolds numbers between 1800 and 2600. The results show that the friction factors of round-edged fins are 40% less than that of the square-edged fins.

- The hydrodynamic tests in an isothermal single-chamber test section with 2 cSt and 5 cSt silicon oils were completed. The experiments were performed using Reynolds numbers variations from 50 to 250 to simulate the prototype heat exchanger flow.

8.3 Project Profile

Start Date: October 2003

End Date: June 30, 2007

Researchers: S. Moujaes (Principal Investigator) and A. Wang

Students: S. Aekula and J. Patil

Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):

S. Moujaes, R. Boehm, A. Wang, S. Aekula, and J. Patil, "Design of Bench Scale HTHX," FY05 Deliverable Report, University of Nevada, Las Vegas, NV, September 29, 2005.

S. Moujaes, R. Boehm, A. Wang, S. Aekula, and J. Patil, "Thermal Property Tests," FY05 Deliverable Report, University of Nevada, Las Vegas, NV, September 29, 2005.

Conference Proceedings:

A. Wang, S. Moujaes, Y. Chen, and V. Ponyavin, "Experimental and Numerical Analyses of Friction Factors in Offset Strip Fin Heat Exchangers," *Proceedings*, FEDSM2007: 5th Joint ASME/JSME Fluids Engineering Conference, San Diego, CA, July 30 - August 2, 2007.

S.F. Moujaes and S.Aekula, "Experimental Preparation and CFD Simulation of Offset Fin High Temperature Heat Exchanger Model," WORLDCOMP'07 Conferences, International Conference on Scientific Computing, Las Vegas, NV, June 25-28, 2007.

S.F. Moujaes and S.Aekula, "CFD Simulation of Offset Fin High Temperature Heat Exchanger Model," 2007 STAR America's Conference, in Detroit, MI, June 25-26, 2007.

9.0 Analytical Studies of the Effects of Acid Exposure on Structure Materials (Allen Johnson, Principal Investigator, UNLV Harry Reid Center for Environmental Studies)

9.1 Introduction

A fundamental understanding of the behavior of the surface layer is necessary to understand the corrosion process. Corrosion occurs at the interface between a substrate (in these studies typically noble metals such as tantalum and niobium) and the corrosive agent (in these studies typically boiling or gaseous HI). A change in last layer of atoms at the surface (~1 microgram/cm^2 of material) can fundamentally change the character of the corrosion. These changes can occur due to minor alloy constituents, contamination of the corrosive environment, and the dynamics of the corrosion itself. Thus, to have confidence in the extensibility of model system studies to the general case, one needs to monitor the composition and structure of the corrosion layer, to make sure that untoward changes are not taking place.

The surface/corrosion layer thickness and structure were measured in this project. The thickness and composition of the corrosion layer, combined with weight changes, yield the corrosion rate. The corrosion layer composition was measured as a function of depth - this will monitor the chemical changes induced by the corrosion and monitor the corrosion environment for unexpected phenomena due to either fundamental mechanistic features or unexpected compositional variation. The surface characteristics of the starting materials were measured. Starting material can have significant changes in the surface layer depending on the details of surface preparation. Effects of surface preparation was studied on corrosion rate. Examining starting materials will also provide important baseline data as well as validating the consistency of the samples under test. Variations in composition of the underlying alloy as a function of position were measured. This included grain size variations, inclusions, intergranular corrosion, etc.

9.2 Research Highlights and Accomplishments

- Corroded samples from the HI decomposition loop chemistry (supplied by General Atomics) and from the SO_3 decomposition loop chemistry (supplied by Ceramatec) were evaluated. Both sample sets were exposed to boiling liquid and vapors at appropriate temperatures in test stands at the source facilities.
- The GA samples came in two sets. The first set was a powder and four samples of three alloys (Ta2.5W, Nb10Hf, Nb1Zr). The Nb1Zr sample had two specimens – the later one had exhibited accelerated corrosion. The powder was removed from the HIx liquid and was found to be Nb oxide – most likely Nb_2O_5 by XPS. All of the alloys show the formation of an oxidized surface – Ta2.5W shows W enhancement, whereas the others show depletions of the alloying component. The failed Nb1Zr sample was notable in that is shows small amounts of lead on the surface and more Zr in the oxide – doubling the sputter time removed the lead, showing that it was on the surface of the oxide.
- Samples exposed to HIx and phosphoric acid show incorporation of phosphorus oxides into the oxide layer. There was a loose powdery residue on some of the metals – they were very water soluble and so not analyzed.

- It was observed in general that the Ta based alloys had the lowest total corrosion and the best resistance to a wide range of chemical environments.
- The ceramics supplied by Ceramatec were in two sets. The first set was composed of exposed and unexposed samples of SiC and Si_3N_4.
- The SiC oxidized in the SO_3 decomposition media to form silicon dioxide and residual carbon was observed by XPS. A similar oxidized layer is seen in Si_3N_4, and no nitrogenous compounds are left behind. This oxide layer had more obvious grains in the oxide layer.

- Using XPS 6 Ceramatec samples were analyzed: CoorsTek Alumina 4, CoorsTek Alumina 6, Ceradyne gas pressure sintered Si_3N_4 G12, Ceradyne hot pressure sintered Si_3N_4 H12, Ceramatec laminated SiC L8 and Morgan SiC M10. They all (except CoorsTek Alumina 4 used as a baseline sample) were exposed to acid and air at 900°C for 1000 hrs.
- Samples were analyzed at different depths and data sets: survey as received, survey after sputtering10s with Ar, then a sputter depth profile (about 1,400nm into the surface), survey after sputter depth profile (SDP).
- The atom percentage of S ~1,400nm from the surface of Alumina 4, Alumina 6, Si_3N_4 G12, Si_3N_4 H12, SiC L8 and SiC M10 are 0; 0.164; 0.817; 0.537; 0.733 and 0.369 respectively. This indicates that acid gas infiltration of the ceramics occurred for all of the ceramics, but Alumina 7 and SiC M10 have the lowest infiltration.
- Except for Alumina 4 (as a base line sample) and Alumina 6, depth profiles of all samples (in the range of about 1,400 nm) showed changes in composition (generally oxidation). The ratios between component elements are far from their expected composition. From the survey and depth profile data, Alumina 6 and SiC M10 seem most stable and thus promising for further study.

In overall summary, the alloys and ceramics both showed the formation of oxidized surface layers in both the HIx and the SO_3 chemistries at temperature. It is expected then that the mechanical and chemical stability of the oxide layer will control the final stability of the material in the S-I thermochemical hydrogen cycle. The literature shows that in other high temperature system (e.g. SiC natural gas burners) the stability of the glassy outer layer was adversely affected by metallic contamination – metallic species in the S-I process fluids should therefore be avoided. The observation of modified metal abundances and the appearance of lead on the surface of the catastrophically failed part indicates that this may also be important for the alloys.

9.3 Project Profile

Start Date: October 2005

End Date: October 2007

Researcher: A. Johnson

Student: T. Ho

Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):

A. Johnson, "HTHX Chemical Support," Final Report, University of Nevada, Las Vegas, NV, June 15, 2007.

10.0 Efficiency Improvement and Cost Reduction of Solid Oxide Electrolysis Cells at UNLV (Clemens Heske, Principal Investigator, UNLV Department of Chemistry)

10.1 Introduction

In close collaboration with Ceramatec and ANL, UNLV investigated the chemical structure and morphology of surfaces that are relevant to solid oxide electrolysis cells (SOECs). Composition, structure, and chemical properties of samples provided by Ceramatec and ANL are analyzed at UNLV with a set of spectroscopic and microscopic surface and interface techniques. The experimental methods include X-ray and UV photoelectron spectroscopy, X-ray emission and absorption spectroscopy, and atomic force microscopy.

The work in particular focused on a comparison of the above-mentioned properties before and after testing at Ceramatec or ANL. This testing included electrochemical conditioning, high-temperature stress testing, and long-term operation. The results of the UNLV experiments are shared with the partners at Ceramatec and/or ANL, in order to give immediate feedback for the chemical surface processes occurring during testing and to assist in the development of more long-term stable devices.

10.2 Research Highlights and Accomplishments

- Successfully installed new electron spectrometer, scanning probe microscope, and sample insertion glove box.
- Completed spectral library for commercially available reference compounds.
- X-ray absorption and emission spectroscopy of $La_x Sr_y MnO_3$ and $La_x Sr_y CoO_3$ samples received from Argonne National Lab were taken at the Advanced Light Source at LBNL.
- Performed stoichiometry analysis of YSZ thin films deposited by Atomic Layer Deposition (ALD)
- Lax Cay MnO_3 (LCM) and Y-stabilized ZrO (YSZ) electrodes prepared by Pulsed Laser Deposition (PLD) were studied with X-ray absorption, emission spectroscopy, Atomic Force Microscopy, and Scanning Probe Microscopy to determine surface composition and chemical structure.
- X-ray absorption was found to be a very sensitive tool to investigate the actual compound and oxidation state of the samples.
- As expected, all samples show characteristic signatures of La, Ca, Mn, and O core levels and Auger transitions. Furthermore, significant C and O contaminations were found on the surface. This is not unexpected, since the surfaces were exposed to air after preparation and during additional tests at ANL.
- The samples from Ceramatec, Inc. consisted of one tested and one untested “button cell” (2.5cm x 2.5cm) and two tested cells (10cm x 10cm) from a standard cell stack (“10INL468”). The cells were tested in a fuel-cell configuration. Photoemission experiments were conducted on the tested cell. The photoemission experiments at UNLV focused on gathering information about the degradation mechanisms in the fuel cell.

10.3 Project Profile

Start Date: July 2005

End Date: September 2007

Researchers: C. Heske (Principal Investigator), M. Bar, and L. Weinhardt

Students: R. Felix, T. K. George, Hofmann, and J. Peiser

<u>Reports Generated (in addition to monthly, quarterly, semi-annual, and annual review reports):</u>

T. Hofmann, L. Weinhardt, M. Bar, and C. Heske, "Spectroscopic Examination of Solid Oxide Electrodes and Electrolytes," Milestone Completion Report, University of Nevada, Las Vegas, NV, May 1, 2007.