Development of C-SiC ceramic compact plate heat exchangers for high temperature heat transfer applications

Per F. Peterson, Haihua Zhao, Fenglei Niu, and Wensheng Huang*
Department of nuclear engineering, University of California, Berkeley

Jens Schmidt and Jan Schulte-Fischedick
Institute of Structures and Design and Institute of Technical Thermodynamics German Aerospace Center, Stuttgart

* Corresponding author

Abstract:

This paper investigates the use of polymer and liquid silicon infiltrated carbon/silicon-carbide composite (C-SiC) materials for the development of inexpensive compact heat exchangers, as part of efforts for thermochemical hydrogen production. These heat exchangers will be capable of operating in the temperature range of 500 to 1400°C with high-pressure helium, liquid fluoride salts (a potential intermediate heat transfer fluid), or other corrosive gases such as SO_3 and HI. C-SiC composites have several potentially attractive features, including ability to maintain nearly full mechanical strength to temperatures approaching 1400°C, inexpensive and commercially available fabrication materials, and the capability for simple forming, machining and joining of carbon-carbon performs, allowing the fabrication of highly complex component geometries. To meet cost goal, candidate materials must have relatively low bulk costs, and fabrication methods must extrapolate to low-cost mass manufacturing. Composite compact offset fin plate heat exchangers concept has been developed to meet the above functional and cost goals, which will serve as the intermediate heat exchanger (IHX) to transfer high temperature heat from a helium-cooled high temperature nuclear reactor to a liquid salt intermediate loop which couples to hydrogen production loops. The IHX uses offset fin structures with fin width and height at 1 mm scale. The detailed local and global thermal mechanical stress analyses show that the designed composite plate heat exchanger can tolerate pressure difference up to 9 MPa and large temperature difference from two fluid sides. Two potential low cost methods to fabricate C-SiC are liquid silicon (melt) infiltration (MI) and Polymer Infiltration and Pyrolization (PIP). Mechanical strength tests on MI coupons show above 200 MPa failure stress. Leak-tight pyrolytic carbon coatings have been successfully applied on MI C-SiC coupons and excellent helium hermeticity were obtained under high pressure and stress after coating. PIP plates with high-quality millimeter-scale fins formed using teflon molds have been successfully demonstrated. The teflon molds were proven to be reusable, so that the process could be extrapolated to inexpensive mass fabrication of compact ceramic heat exchangers. Prototype test heat exchangers are being fabricated basing on both MI and PIP methods.

I. Introduction

High temperature heat exchangers for nuclear hydrogen production

In support of the Nuclear Hydrogen Initiative, the UC Berkeley Thermal Hydraulics Laboratory has been studying the use of carbon and silicon carbide composite materials (C-

SiC) in manufacturing high temperature heat exchangers. These heat exchangers will be capable of operating in the temperature range of 500 to 1400°C with high-pressure helium, liquid fluoride salts (a potential intermediate heat transfer fluid), or other corrosive gases such as SO_3 and HI. C-SiC composites have several potentially attractive features, including ability to maintain nearly full mechanical strength to temperatures approaching 1400°C, inexpensive and commercially available fabrication materials, and the capability for simple forming, machining and joining of carbon-carbon performs, allowing the fabrication of highly complex component geometries. To meet cost goal, candidate materials must have relatively low bulk costs, and fabrication methods must extrapolate to low-cost mass manufacturing. Composite compact offset fin plate heat exchangers concept has been developed to meet the above functional and cost goals. One application for such design is the intermediate heat exchanger (IHX) which transfers high temperature heat from a helium-cooled high temperature nuclear reactor primary loop to a liquid salt intermediate loop which couples to hydrogen production loops.

The IHX operates at temperatures ranging from about 600°C to 1000°C and at pressure differences from 6 to 8 MPa. The liquid salt loop will operate at 1 MPa while the gas loop pressure depends on choice of reactor. For example, GT-MHR (Gas Turbine Modular Helium Reactor) operates at 7 MPa and PBMR (Pebble Bed Modular Reactor) operates at 9 MPa. IHX will be immerged in high pressure helium environment. The IHX uses offset fin structures with fin width and height at 1 mm scale. The proposed design provides a good candidate approach for constructing an IHX because high power density can be obtained with small amounts of material and small fluid inventories. Two potential low cost methods to fabricate C-SiC plates are liquid silicon (melt) infiltration (MI) and Polymer Infiltration and Pyrolization (PIP). Thin ceramic plates are manufactured with features like flow channels and fins on one side and a smooth surface on the other side. Plates are reaction-bonded to form a heat exchanger monolith.

Ceramic Composites fabrication via LSI and PIP process

Ceramic matrix composites (CMCs) fabricated by the Liquid Silicon Infiltration (LSI) process provide a potentially very attractive lightweight construction material for high-temperature heat exchangers, due to their ability to maintain nearly full mechanical strength to high temperatures (up to 1400°C), the simplicity of their fabrication, their low residual porosity, and their relatively low cost. The typical steps in fabricating CMCs, especially C/C-SiC, include the green manufacturing of CFRP preforms by molding short fibres or laminating carbon fabrics, carbonization (up to 1650 °C) and optionally graphitization (up to 2800 °C) and final pressureless silicon melt infiltration in the porous C/C preforms.

The German Aerospace Research Center (DLR) has developed high-temperature CMC bayonet heat exchanger components for use in Externally Fired Combined Cycles (EFCC) under corrosive flue gas atmospheres up to 1300 °C. The engineering of such CMC tubes with flanges as well as the application of a multilayer Environmental Barrier Coating (EBC) was successfully demonstrated (figure 1). The latter demonstrated improved properties under thermocycling, oxidative and corrosive loadings up to 1400 h under 1300 °C [1]. Since none of the tested C/C-SiC components showed sufficient gas-tightness, future work must be focused on substrates and coatings to reduce gas permeation of ceramic heat exchanger handling with high-pressure gas or liquids.

Figure 1 Cordierite coated open and end capped CMC tubes (Ø 50/40 mm, length up to 800 mm) with flanges (left) [1]; microstructure of BoraSiC® (SiC-B_4C-SiC) and cordierite coated C/C-SiC (right).

Starfire has already fabricated plate-type heat exchangers using PIP process (http://www.starfiresystems.com). PIP is the latest technique resulting from worldwide research to develop a process that enables the fabrication of advanced ceramics more efficiently than conventional processes. This technique involves soaking a fiber preform or powder compact with a liquid polymer precursor that converts to ceramic material upon pyrolysis. Advantages of PIP include simpler, less costly equipment, lower process temperatures, shorter cycle times, and capability to produce more complex parts. Also, polymers afford the potential to control materials chemistry at the molecular level. Starfire has fabricated carbon fiber reinforced SiC matrix heat exchanger with sub-millimeter flow channels. They used carbon felt preform material, cut the flow channels using a saw blade, bonded the plates together and then used multiple PIP processing steps to create a leak-tight part.

II. Heat exchanger design and thermal analysis

Plate-type ceramic heat exchangers with relatively small flow channels provide a good candidate approach for constructing an IHX because high power density can be obtained with small amounts of material and small fluid inventories. Figure 2 illustrates the offset geometry suggested for high temperature heat transfer between helium and liquid salt. Figure 3 shows a potential design for the plates that will make up such a heat exchanger (shown in right).

To perform stress analysis on a compact plate-type heat exchanger such as the IHX described above, the Pro/Engineer Wildfire finite-element stress analysis module Pro/Mechanica (Pro/M) was used [2]. Finite element analysis has a proven history of yielding accurate results for stresses over various kinds of geometries. However, for complex geometries the amount of time needed to process finite-element models can become prohibitively large. As the number of polygons and the order of the equations fitted increase, the number of simultaneous linear equations that must be solved increases exponentially. For example, finding the stresses accurate to 10% on a 600-polygon unit cell model with Pro/M can take up to 30 minutes on the latest personal computer. Finding detailed stress distribution in a complete heat exchanger using the finite element method (FEM) requires on the order of millions of hours in PC computation time. Therefore, it is not practical to analyze the entire heat exchanger design directly. Previously, plate-fin heat exchangers (PFHE) have been analyzed by dramatically simplifying their geometry. We propose an alternative approach, based on equivalent properties, that is more ambitious than the previous attempts at analyzing the stresses on a PFHE. This method, dubbed the unit cell method, gives more accurate results than previous approximate methods.

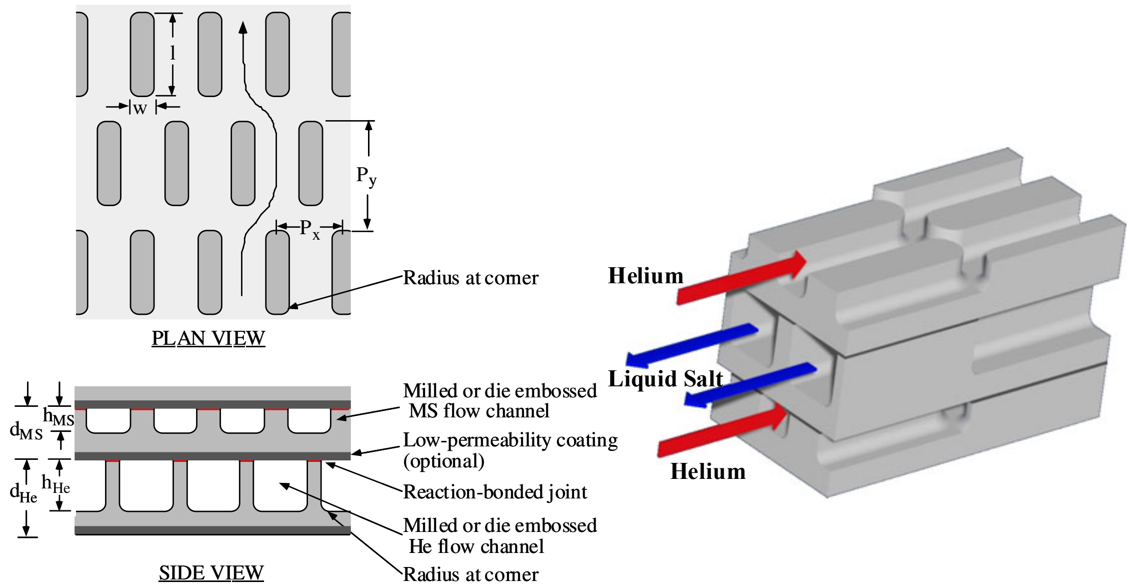


Figure 2 Cut-away view through a plate showing alternating liquid salt (top and bottom arrows) and helium (middle arrows) flow channels. Dark bands at the top of each fin indicate the location of reaction-bonded joints between each plate.

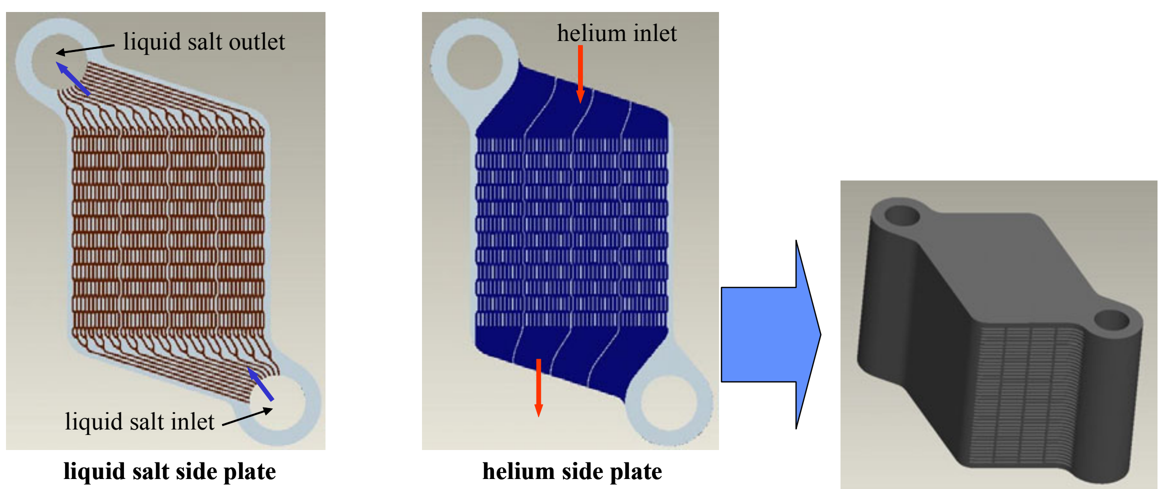


Figure 3 Liquid salt and helium plates, and complete HX module.

The methods we use to obtain approximate thermal and mechanical stresses are composed of three steps. First, divide full model into regions which are composed of unit cells, and find effective properties of unit cells; the effective mechanical and thermal properties for each unit cell are founded through various 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. For example, if the result of average stress analysis shows that region A has a maximum compressive stress of -15 MPa (all compressive stresses are written as negative numbers), an overall pressure of 15 MPa is applied across all six surfaces of unit cell A. The maximum compressive stress found in the unit cell A in this FEM simulation would be the max actual local compressive stress in region A. This also pinpoints locations where stresses are greatest. Similar method is employed to find the maximum tensile stress.

Figure 4 shows the region division and typical unit cells. Figure 5 shows the visual representation of the maximum principle stress distribution when unit cell A is under 2 MPa of average global pressure on all six boundary surfaces and the minimum principle stress distribution when unit cell A is under average global 11 MPa of pressure. From figure 5, we can see that the areas with the greatest tensile stresses are the roots and top corners of the fins on the helium plate, and the areas with the greatest compressive stresses are the roots and top corners of the fins on the liquid salt plate. This can be explained by the fact that the helium gas is under greater pressure than the liquid salt and will push against the walls of the helium channels. This effect causes the helium fins to become stretched and the liquid salt fins to become compressed. The max and min local principle stresses in region A were found to be 40 and -55 MPa. C-SiC materials have yield strengths ranging from 100 to 400 MPa depending on the manufacturing process. If we assume the final material to have a yield strength of around 300 MPa, the stress values we find were within the yield limit of C-SiC composite material by a factor of six to eight.

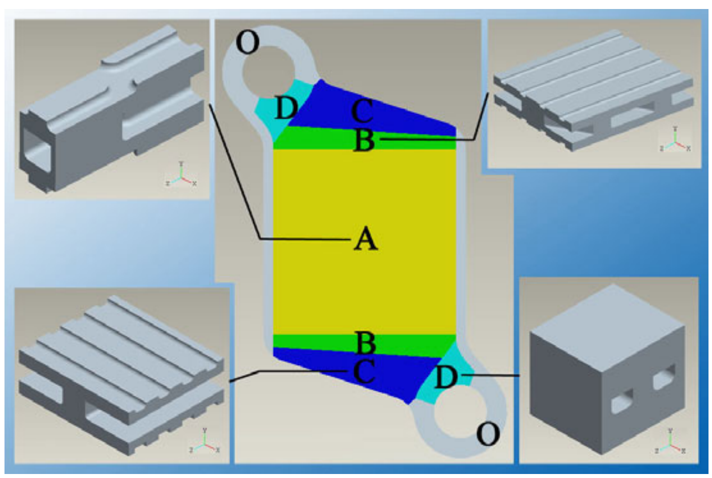


Figure 4 Region division and unit cells.

III. Concept for the low-cost fabrication of plate type HX assembly with biomorphic SiC ceramics

To overcome the problems of high gas permeability of CMCs, biomorphic SiC ceramics might be an alternative material. However, in this case the brittleness of monolithic ceramics has to be considered. Thus a design has to be developed exhibiting low stress under steady state as well as dynamic loadings. FEM calculations demonstrated that this can be achieved by using the OSF plate-fin heat exchanger also in the EFCC processes [3].

Currently there are different biomorphic SiC ceramic grades under investigation at DLR which differ in their phase distribution and therefore exhibit different properties [4]. Splint based (SB) SiSiC ceramics exhibit high strength due to the high amount of SiC whereas fibre reinforced (FR) SiSiC ceramics contain pitch based carbon fibres in the structure and are therefore less brittle. The basic materials for SB and FR ceramics are fine wood powders, phenolic resin and carbon additives which can be easily formed into wood based composite (WBC) plates by axial pressing with dies. As reference WBC material commercially available medium density fiber boards (MDF) can be used as green bodies. The further processing of the WBC is an adapted high temperature LSI processing. For the accurate machining of pyrolysed carbon plates into the final contour diamond milling tools have to be used. The joining of single plates into the final heat exchanger assembly can be performed by using carbon based glues. The final step is the pressureless Si-infiltration which combines the plates to the desired HX design.

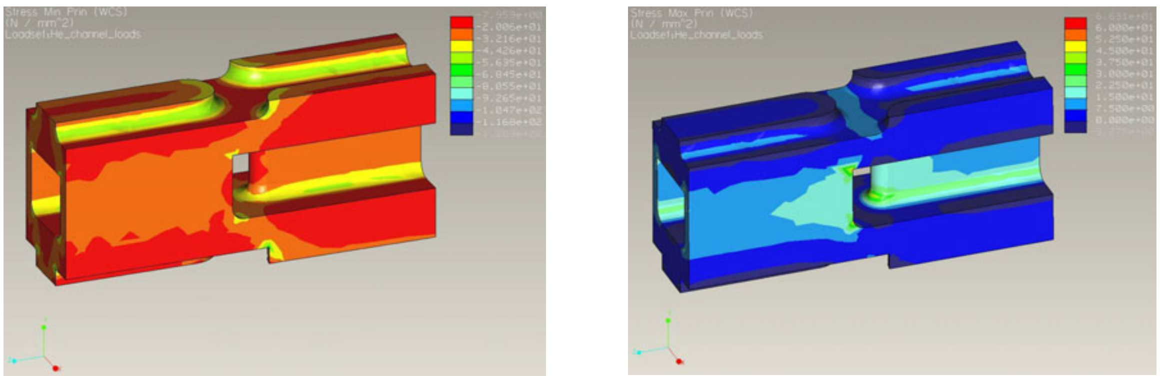


Figure 5 Max principle stress for unit cell A at 2 MPa pressure (left, scale: 0 to 60 MPa) and min principle stress for unit cell A at 11 MPa pressure (right, scale: -20 to -117 MPa).

If an oxidative or corrosive protection coating is required a layer of SiC, carbon or cordierite must be applied on the SiC ceramic surface (after Si-infiltration) via chemical vapor deposition (CVD), vacuum plasma spraying (VPS) or other deposition techniques. In this case the coated surfaces must be finally sintered together to obtain the gas-tight HX stack.

First prototypes of HX plates in OSF design with flow channels have been machined from FR bulk material in the smooth carbon stage (figure 6 left). Single fins with a length of 10 mm, a height of 4 mm and a width of 2 mm could be obtained (figure 6 right). With these plates prototypical heat exchanger modules shall be manufactured and tested in DLR's micro gas turbine laboratory. Preferably, future HX plates should be formed in inexpensive net shape design by axial warm pressing. The pressing is currently under investigation at DLR. The composition of the basic composites must be adapted due to the small dimensions of the flow channels. To maintain a good shaping during pressing and low shrinkage during pyrolysis short carbon fibres with a length < 1 mm are preferably used as additives.

IV. Material characterization: fabrication and tests

MI coupon testing for key material properties

DLR provided melt-infiltrated composites testing coupons, including uncoated splint based SiSiC samples, uncoated C/C-SiC samples, cordierite coated splint based SiSiC samples, and cordierite coated fiber reinforced SiSiC coupons. All of these coupons have approximate 50 mm diameter and 3 mm thickness. Hyper-Therm HTC, Inc. did CVD carbon coatings on C/C-SiC coupons with similar method as ORNL fuel cell plate coating process [5]. The main purpose of CVD carbon coating is to seal the surface to be hermetic since the fiber-reinforced samples have interconnected porosity to allow helium to leak through.

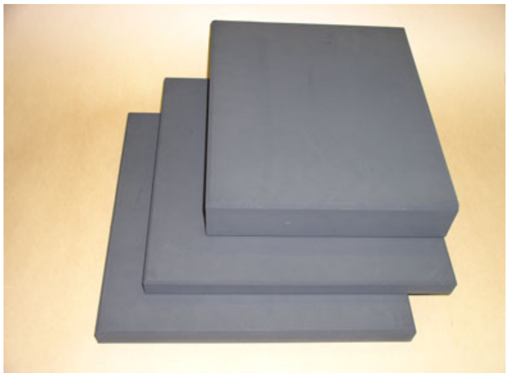
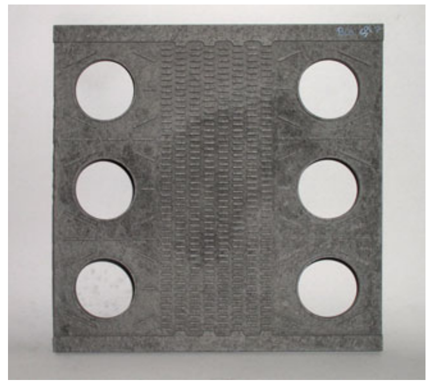



Figure 6 Carbon plates (dim. 300x300 mm², thickness 20-50 mm) derived from wood based composites after pyrolysis at 1650 °C (left). Plate type heat exchanger (dim. 300x300x5 mm³) in OSF design with flow channels after machining from such carbon bulk material (right)

Helium permeation tests were performed on uncoated splint based SiSiC coupons, uncoated fiber reinforced SiSiC coupons, both cordierite coated and CVD carbon coated fiber reinforced SiSiC using bubbles observing method. The schematic of test design is shown in Figure 7. A test coupon is clamped between two O-rings. High pressure helium is connected at the bottom side. The top side is covered with water. If there is any helium leakage through the test coupon, gas bubble will be observed. Before formal test, a copper disc was used to test hermeticity of the fixture. Under 0.5 MPa pressure difference, no visible bubble was detected after 10 minutes. This test verified that there was no baseline leak through O-rings. Different types of coupons were tested for hermeticity. Initially low pressure was applied. If no bubble observed, the helium pressure will be increased until that bubbles are observed. The experiments show that uncoated fiber reinforced SiSiC has considerable permeability. The threshold pressure difference for detection of leaks is as low as 3.4 kPa. No visible bubbles appear for both coated and uncoated splint based SiSiC coupons. The permeation test on the CVD SiC and Pyrolytic Carbon coated fiber sample shows that the coating layer kept good hermeticity under high pressure up to 5.5 MPa. No helium bubbles were observed on the sample surface, even though some baseline leaks started to be detected from the fittings of the test fixture. The sample, however, failed about five minutes later. Subsequent calculation showed the sample was experiencing a tensile stress of 276 MPa before breaking, which is much higher than 40 MPa maximum tensile stress (under 9 MPa He) typically found from the stress analysis. This test directly verified that CVD carbon coated SiSiC material could keep helium hermeticity under high pressure and stress well beyond the working pressure difference and stress expected for NGNP.

With the same test fixture for helium hermeticity testing, Young's moduli and failure stress of carbon fiber reinforced SiSiC (FR-SiSiC), splint based SiSiC (SB-SiSiC) and pitch based carbon fiber reinforced SiSiC (BioKer) were measured using strain gauge. Table 1 summarizes some typical MI material properties.

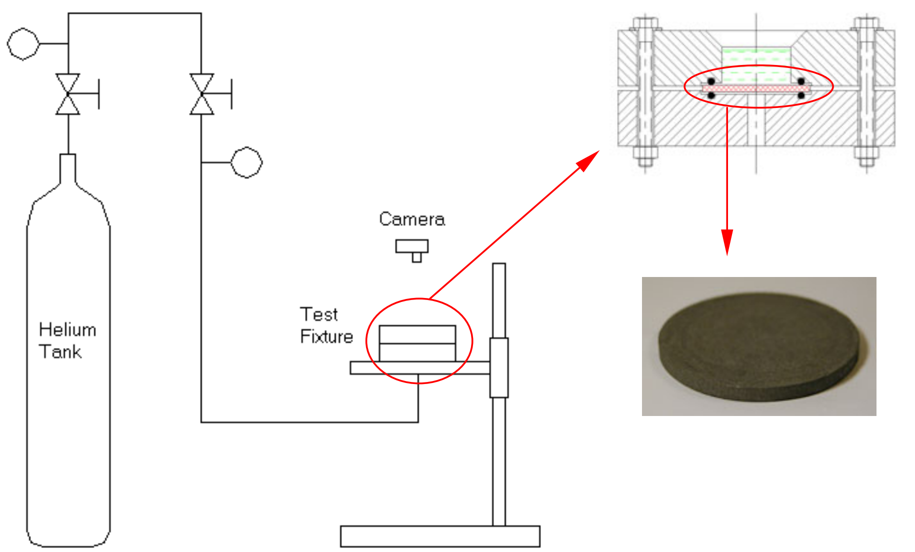


Figure 7 Helium permeation test using water bubble observing method.

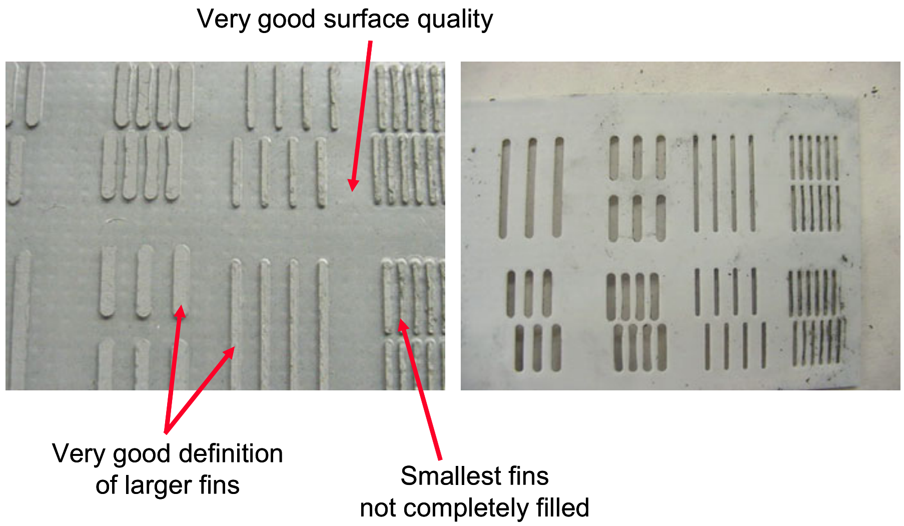
Table 1 Measured mechanical properties for some typical MI SiSiC materials

MI Material	Density kg/m^3	Young's modulus GPa	Failure stress MPa
SiSiC (MDF) with coating	2523	325	270
Splint based (SB) SiSiC	2932	450	224
Pitch carbon fiber reinforced (FR) SiSiC	2600	298	200

PIP coupon fabrication methods

For polymer-infiltrated composites, this work is being performed in collaboration with a U.S. composites vendor, COI Ceramics, Inc (COIC). In this collaboration, UCB provides plate design and fabrication ideas, and manufactures molds for COIC. COIC develops and optimizes detailed compositions of mixtures and fabrication processes, and fabricates test plates. Green plates with heat transfer channels are fabricated with Teflon molds. The green plate are cured at higher temperature within mold and then separated from mold. The cured plate is subject to multiple pyrolysis and polymer-infiltration under high temperature. The PIP plates are laminated together to form heat exchanger monolith.

Successive process improvements resulted in the successful fabrication of 1 mm and 1.5 mm high fins with high dimensional quality. The plates were cleanly released from Teflon molds. Figure 8 shows PIP C-SiC composite plate fabricated using 1mm through Teflon mold. Figure 9 shows the pyrolyzed plates with 1.5 mm fin height. A noteworthy result of the COIC work was the demonstration that the teflon molds can be used multiple times without noticeable degradation. This shows that teflon molds can be used for mass manufacturing of ceramic heat exchanger plates. Lamination and pyrolization of plates has been successfully demonstrated in a cross-flow HX geometry as shown in Fig. 10. This success demonstrated viability of PIP fabrication method for complex C-SiC plate heat exchangers. Further works are being performed to refine the whole processes.



Fabricated plate after cure **1mm through Teflon mold**

Figure 8 PIP C-SiC composite plate fabricated using 1mm through Teflon mold (COIC).

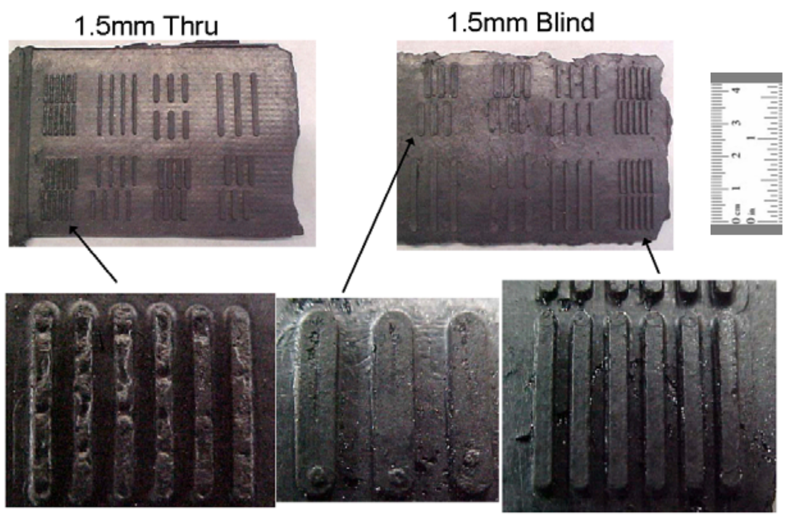


Figure 9 Pyrolyzed Plates with 1.5 mm fin height

PIP test plate stack to be laminated

PIP test plate stack after lamination and pyrolization processes

Figure 10 PIP plates lamination.

V. Conclusions

PIP and MI C-SiC materials are potential advanced materials for the development of inexpensive compact heat exchangers for thermochemical hydrogen production. Composite compact offset fin plate heat exchanger concept has been developed to meet the functional and cost goals, which will serve as the IHX to transfer high temperature heat from a helium-cooled high temperature nuclear reactor to a liquid salt intermediate loop which couples to hydrogen production loops. The IHX uses offset fin structures with fin width and height at 1 mm scale. The detailed local and global thermal mechanical stress analyses show that the designed ceramic plate heat exchanger can tolerate pressure difference up to 9 MPa and large temperature difference from two fluid sides. Mechanical strength tests on MI coupons show above 200 MPa failure stress. Leak-tight pyrolytic carbon coatings have been successfully applied on MI C-SiC coupons and excellent helium hermeticity were obtained under high pressure and stress after coating. PIP plates with high-quality millimeter-scale fins formed using teflon molds have been successfully demonstrated. The teflon molds were proven to be reusable, so that the process could be extrapolated to inexpensive mass fabrication of compact ceramic heat exchangers.

Acknowledgement

This work was funded by the U.S. Department of Energy under the contract DE-FC07-04ID14566 through Nuclear Hydrogen Initiative UNLV High Temperature Heat Exchanger Project.

Reference:

1. J. Schmidt; J. Schulte-Fischedick; E. Cordano, C. Mao, V. Liedtke, R. Fordham: *CMC tubes based on C/C-SiC with high oxidation and corrosion resistance*, Proceedings of the 5th international conference on high temperature ceramic matrix composites, ed. by M. Singh, R. Kerans, E. Lara-Curzio, R. Naslain, published by the American Ceramic Society, p. 531-536, 2004.
2. Huang, W., Zhao, H., and Peterson, P.F., "Multi-Scale Stress Analysis for Compact Plate Heat Exchangers," Proc., 2006 International Congress on Advances in Nuclear Power Plants (ICAPP '06), Embedded International Topical Meeting 2006 American Nuclear Society Annual Meeting, Reno, NV, USA, June 4-6, 2006.
3. J. Schulte-Fischedick, V. Dreißigacker, R. Tamme: An innovative ceramic high temperature plate- fin heat exchanger for EFCC processes, Applied Thermal Engineering, in press.
4. M. Gahr; J. Schmidt; A. Hofenauer; O. Treusch: *Dense SiSiC ceramics derived from different wood-based composites: processing, microstructure and properties*, Proceedings of the 5th international conference on high temperature ceramic matrix composites, ed. by M. Singh, R. Kerans, E. Lara-Curzio, R. Naslain, published by the American Ceramic Society, p. 425-430, **2004**
5. T.M. Besmann, J.W. Klett, J.J. Henry, Jr., and E. Lara-Curzio, "Carbon/Carbon Composite Bipolar Plate for Proton Exchange Membrane Fuel Cells," Journal of the Electrochemical Society, 147 (11) 4083-4086, 2000.