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Heat Exchangers for the Next Generation of Nuclear Reactors

Xiuqing Li, Renaud Le Pierres and Stephen John Dewson
Heatric Division of Meggitt (UK) Ltd.,
46 Holton Road, Holton Heath, Poole ,Dorset BH16 6LT, England
Phone: +44(0) 1202 627000, Fax: +44(0) 1202 632299, Email: xiuqing.li @heatric.com

Abstract – *The realisation that fossil fuel resources are finite, the associated rising price and a growing concern about greenhouse gas emissions, has resulted in renewed interest in nuclear energy. Generation IV and other programmes are looking at a variety of new reactors. These reactors vary in type from Very High Temperature Gas Cooled Reactors (VHTR) to Liquid Metal Fast Reactors (LFR and SFR) with cooling mediums that include:*

- *Helium*
- *Supercritical carbon dioxide*
- *Sodium*
- *Lead*
- *Molten salts.*

In addition interest is not just focused on production of electrical power with an efficiency greater than that associated with the Rankine Cycle (typically 30 -35%); there is now genuine interest in nuclear energy as a heat source for hydrogen production, via the Sulphur Iodine Process (SI) or high temperature electrolysis.

The production of electrical power at higher efficiency via a Brayton Cycle, and hydrogen production requires both heat at higher temperatures, up to 1000°C and high effectiveness heat exchange to transfer the heat to either the power or process cycle.

This presents new challenges for the heat exchangers. If plant efficiencies are to be improved there is a need for:

- *High effectiveness heat exchange at minimal pressure drop.*
- *Compact heat exchange to improve safety and economics.*
- *An ability to build coded heat exchangers in a variety of nickel based alloys, oxide dispersion strengthened alloys (ODS) and ceramic materials to address the temperature, life and corrosion issues associated with these demanding duties.*

Heatric has already given consideration to many of these challenges. Their Print Circuit Heat Exchanger (PCHE) and Formed Plate Heat Exchanger (FPHE) technology which are commercially available today, will fulfil all of the duties up to temperatures of 950°C. In addition products currently under development will further increase the temperature and pressure range, while offering greater corrosion resistance and operational life.

This paper outlines the challenges for the heat exchangers and the development required, with particular attention given to material selection. It is further the objective of this study to demonstrate that heat exchangers such as PCHE and FPHE are able to meet the above challenges.

I. INTRODUCTION

Growing concerns over energy resource availability, climate change, air quality and energy security, have resulted in a number of national and international programmes to develop advanced nuclear reactors such as "Generation IV" reactors.

The next generation of reactors need to target higher efficiency, improved economics and increased safety. In addition to traditional large scale electricity production, consideration will need to be given to small scale plant for remote locations and providing high grade heat for hydrogen production and similar.

Depending on their intended scale and application, a variety of reactor types are being considered using fluids such as:

- Helium
- Supercritical carbon dioxide (S CO_2)
- Nitrogen and helium nitrogen mixes
- Liquid metals
- Molten salts.

These reactors will need to operate at higher temperatures than today's reactors[1]. In particular, the very high temperature gas cooled reactor (VHTR), a leading candidate reactor for producing both hydrogen and electricity, could have outlet temperatures up to 1000°C (Table I). For supercritical carbon dioxide cycles, pressures could be as high as 200 bar. Nuclear hydrogen production using advanced methods such as high temperature electrolysis or thermo-chemical cycles (e.g. sulphur iodine cycle) not only require high temperature (up to 950°C) but also necessitate the use of corrosive intermediate fluids.

TABLE I

The Next Generation of Nuclear Reactors

Reactor Type	T^{max} (°C)	Pressure (MPa)	Coolant
GFR	850	7	gas
LFR	800	0.1	liquid metal
MSR	950	0.1	molten salt
SFR	550	0.1	liquid metal
VHTR	1000	7	gas
S CO_2 Reactor	550	20	S CO_2
PBMR/HTR-10	600	9	gas

This paper outlines the requirements for the heat exchangers characteristics, the need for compact technology and the need for high integrity construction. The paper then gives consideration to material selection. As ASME III needs to be extended before it can address many of the Generation IV issues, initial selection assumes ASME VIII (Boiler and Pressure Vessel Code). However, restricting material selection to ASME VIII approved materials may compromise exchanger temperature, pressure and life.

II. HEAT EXCHANGER

Irrespective of reactor type and the resultant coolant the key to efficiency is high effectiveness heat exchange. Process economics require an intermediate heat exchanger or recuperator with effectiveness greater than 90%, and for some applications under consideration as high as 98%. The combination of temperature, pressure and application demand high integrity construction, this rules out many exchanger types and joining techniques such as brazing. The need for containment (and associated vessels) reduced fluid inventory and economics. (The need to minimise the use of exotic alloys associated with high temperature pressure vessels) result in a need for compact heat exchange. The success of the next generation of nuclear reactor will depend in part upon the correct selection of a high effectiveness, high integrity compact heat exchanger.

Without doubt the next generation of nuclear reactor will be dependent upon compact heat exchanger technology. For the reactor types described in Table I there is significant benefit (often enabling benefit) associated with the use of compact heat exchanger technology.

II.A. High Effectiveness

Benefits vary, dependent upon the reactor type; however, there is always a need for high effectiveness and close temperature approach: features normally associated with compact and in particular plate type heat exchangers.

II.B. High Mechanical Integrity

There is a need for high mechanical integrity. Supercritical carbon dioxide cycles are associated with pressures in the order of 20 MPa, the fast reactors tend to use liquid metal or molten salt at relatively low pressure (0.1MPa) but high pressure gas (supercritical carbon dioxide) is often considered for the secondary side. For the VHTR pressures appear to be more modest, typically (7 MPa). However, consideration needs to be given to allowable design stresses at elevated temperature. A leading material contender for VHTR applications is Alloy 617.

Assuming ASME criteria, at 950°C alloy 617 has design stress of 7.9 MPa. Clearly designing a heat exchanger or (any pressure vessel) to operate at a design pressure 7MPa that is approaching 90% of the design stress (7.9MPa) is not possible, using conventional design codes or criteria. Conventional design rules would suggest the design pressure should be less than 50% of the design stress.

Even with high integrity compact technology design pressure to design stress ratios significantly greater than 50% are not possible. It will therefore be necessary to give consideration to differential pressure design.

For exchangers where high design pressure to design stress ratio is required, such as VHTR, it is essential that joints within the exchanger core have parent metal properties. There is little value in considering high temperature alloys and then restricting their strength with joining techniques resulting in less than parent metal properties. Heatric's PCHE and FPHE with their unique diffusion bond are both exchanger types where parent metal properties can be guaranteed for the joints within the core.

II.C. Exchanger Configuration.

In addition to high thermal effectiveness and mechanical integrity, candidate compact heat exchangers will need to exhibit design flexibility.

There will be a need for counter-current configuration. Low pressure drops will require a minimum area associated with cross flow entry regions or distributors, if flow mal-distribution mechanisms are to be avoided. Heatric's "platelet" type configurations results in some of the best active area to distributor area ratios available for plate type heat exchangers (Fig. 1).

It may also be necessary to consider an exchanger with very different passage sizes on the primary and secondary side. The optimum passage geometries and sizes for a liquid metal, or molten salt are very different from those for a gas such as helium, nitrogen or carbon dioxide.

Heatric's PCHE (Fig. 2) and FPHE (Formed Plate Heat Exchanger, Fig. 3) products offer such design flexibility, between them offering passage sizes from 0.1mm semicircles to 5mm square section.

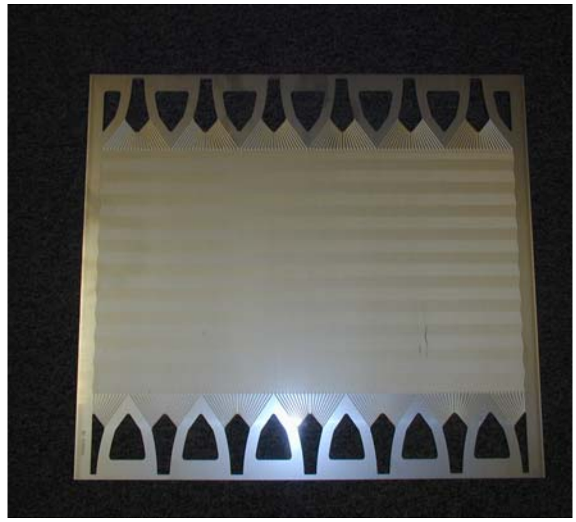

Fig. 1. PCHE platelet configuration.

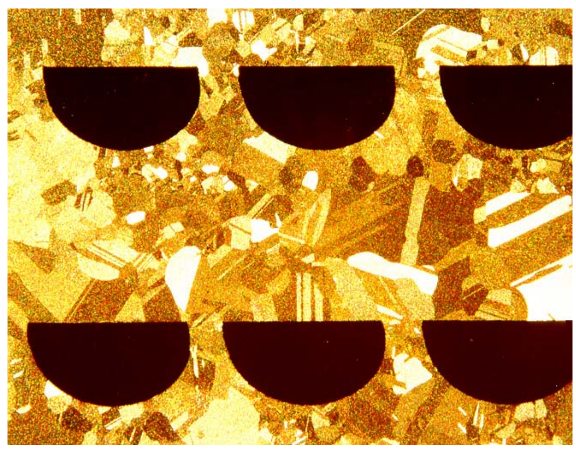

Fig. 2. Micrograph of section through diffusion diffusion bonded PCHE core

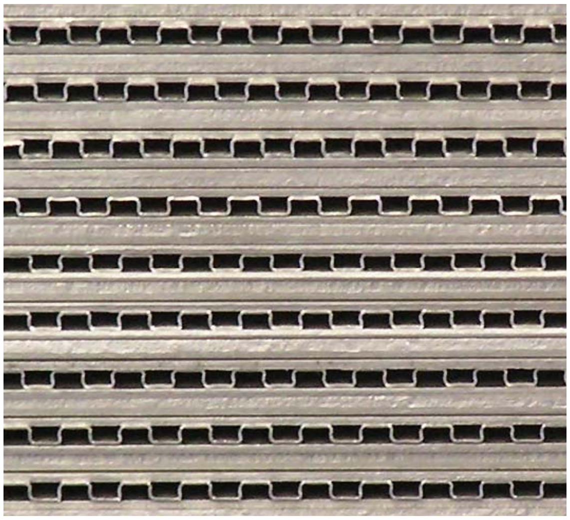

Fig. 3. Section of diffusion bonded FPHE core

II.D. Transient Characteristics

Although heat exchangers for nuclear duties will predominantly operate in steady state, there is a need to give consideration to thermal and pressure transients associated with start-ups, shut downs and unscheduled events. Some of these events will give rise to severe temperature gradients and high resultant thermal stresses. It will be important to demonstrate that the considered heat exchanger has a suitable fatigue life and failure mode, when subjected to repeated transients. Modelling and testing performed by Heatric, has already demonstrated that the PCHE is more or less immune to pressure cycles and has a thermal fatigue life in excess of that required by currently available nuclear heat exchanger specifications.

II.E. Codification Considerations

Irrespective of heat exchanger type, design codes must be extended and developed to encompass the requirements of the next generation of nuclear reactors.

Will the heat exchanger represent a nuclear boundary, requiring design and manufacture to ASME III? If the heat exchanger is placed in a containment vessel, which becomes the nuclear boundary, design to ASME VIII may suffice.

If design and manufacture to ASME III is a requirement, it will be necessary to select and qualify candidate materials to a minimum design temperature of 900°C.

Given the required temperatures, for most conventional alloys design in the creep region seems inevitable. Both creep and creep fatigue must be considered. However, it may be necessary to extend the code to allow design and manufacture in ODS or non-metallic (ceramic) materials.

It will be necessary to develop and qualify inspection procedures and techniques that are applicable to the materials of construction and compact heat exchanger technology.

III. MATERIAL ISSUES

For next generation reactors the heat exchangers materials challenges are:

- Materials will be exposed to high temperatures, up to 1000°C. This could result in metallurgical structural changes, causing strength loss.
- Materials will be exposed to very corrosive environments. Even for the helium cooled exchangers, the coolant will be contaminated and may contain N_2, CO, CO_2, H_2, CH_4, and H_2O impurities.
- Up to a 60-year operation life will be required. This necessitates very long-term materials stability.
- Process-heat for large-scale hydrogen production, will also need materials compatibility with heat transfer media and reactants

Initial material selection will focus on those materials approved by ASME VIII for the manufacture of conventional pressure vessels. We will then discuss the more advanced alloys, suitable for the aggressive environments associated with nuclear hydrogen production via thermo-chemical process, such as the SI process.

Results from Heatric's alloy 617 development programme, including diffusion bonding and passage formation, are also reported in this section.

III.A. ASME Candidate Materials

There are no materials approved to ASME III for temperatures approaching 900°C (1650°F). Primary materials selection therefore starts with those approved to ASME VIII.

Ten alloys are identified for use at design temperatures equal to or greater than 871°C (1600°F).[2] They are:

- Alloy 617
- Alloy 602CA
- Alloy 556
- Alloy 800H
- Alloy 800HT
- Alloy 330
- Alloy 230
- Alloy HX
- 253 MA
- Alloy 625

The compositions of these alloys with their maximum design temperatures are given in Table II. The Pitting resistance equivalent number (PREN) of these alloys was calculated according to[3]:

$$PREN = Cr + 1.5(Mo + W + Nb) + 30N \qquad (1)$$

Among these alloys, Alloy 617 and Alloy 602CA are approved to the highest temperature of 982°C (1800°F) while alloy 625 is approved to the lowest design temperature, 871°C. Other alloys are approved to 898°C (1650°F) (Fig. 4).

However, design temperature is only part of the material selection process, consideration must also be given to the mechanical requirements and high temperature corrosion resistance. Adequate material workability and product form availability must also be ensured, although this is not discussed in this paper.

For example, Alloy 602CA is approved to 982°C (code case 2359), but the allowable design stresses at high temperatures are lower than other alloys. This means this alloy could not meet some of the design stress requirements at high temperature.

Alloy 556 has very high allowable strength at high temperature, but its resistance in nitriding media is poor when compared with alloy 617, alloy 230 and alloy X.[4]

Detailed material reviews for most of the alloys listed here suggest that either alloy 617 or alloy 230 is the most suitable material for high temperature (900°) intermediate heat exchangers in a helium or nitrogen environment.[4]

For more corrosive environments consideration needs to be given to alloys, that have higher PREN values such as alloy C-276 and C-22 (Table II).

III.B. Other materials

The applications of heat exchangers in aggressive corrosive environments such as liquid metal and molten salt, may require material types that are not currently included in codes, such as ASME III or VIII. One of the technical challenges of the nuclear hydrogen initiative (NHI) is to develop high temperature, corrosion resistant heat exchanger materials for extended service under thermo-chemical conditions.

Depending on the applications, the candidate materials could be metallic, oxide dispersion strengthened (ODS) materials, composites or ceramics.

The procedures in choosing correct heat exchanger materials include:

- Identify the operating requirements (fluids, temperatures, pressures and heat duty) of the heat exchanger.
- Identify the possible materials
- Evaluate the selected materials through tests in the representative conditions

For nuclear hydrogen production using the sulphur-iodine thermo-chemical cycle, three chemical reactions are involved:[5]

$$2H_2O + SO_2 + I_2 \rightarrow H_2SO_4 + 2HI \qquad (2)$$

$$H_2SO_4 \rightarrow H_2O + SO_2 + 1/2O_2 \qquad (3)$$

$$2HI \rightarrow H_2 + I_2 \qquad (4)$$

Sulphuric acid and hydrogen iodide are generated in the central low temperature reaction (~120°C), the Bunsen reaction (Eq. (2)). Sulphuric acid is decomposed at high temperature (830-900°C) (Eq. (3)) and hydrogen iodide is decomposed at lower temperature (300-450°C) (Eq. (4)). There are significant chemical separations associated with each chemical reaction. Water is the primary solvent in the system and iodine is also an important solvent in the Bursen reaction. Thus the heat exchanger fluids may include helium, molten salt or liquid metal (primary side) and H_2SO_4, SO_2, H_2O, I_2, HI or O_2 (secondary side).

TABLE II

Normal Compositions of ASME Candidate Materials

Alloys	UNS No	T^{Max}°C (°F)	Ni	Fe	Cr	Mo	Nb	W	Si	N	PREN
617	N06617	982(1800)	52	2	22	9	-	-	-	-	36
602CA	N06025	982(1800)	62	10	25	-	-	-	0.3	-	25
556	R30556	898(1650)	21	30	22	3	-	3	-	-	31
800HT	N08811	898(1650)	33	42	21	-	-	-	-	-	21
800H	N08810	898(1650)	33	42	21	-	-	-	-	-	21
330	N08330	898(1650)	35	36	19	-	-	-	1.2	-	19
230	N06230	898(1650)	57	2	22	2	-	14	-	-	46
HX	N06002	898(1650)	47	20	22	9	-	0.6	0.5	-	42
253MA	S30815	898(1650)	11	-	21	-	-	-	-	0.2	21
625	N06625	871(1600)	60	3	22	9	4	-	0.3	-	41
C-276	N10276	676(1250)	58	-	16	16	-	4	3.5	-	46
C-22	N06022	676(1250)	57	-	22	14	-	3	-	-	48

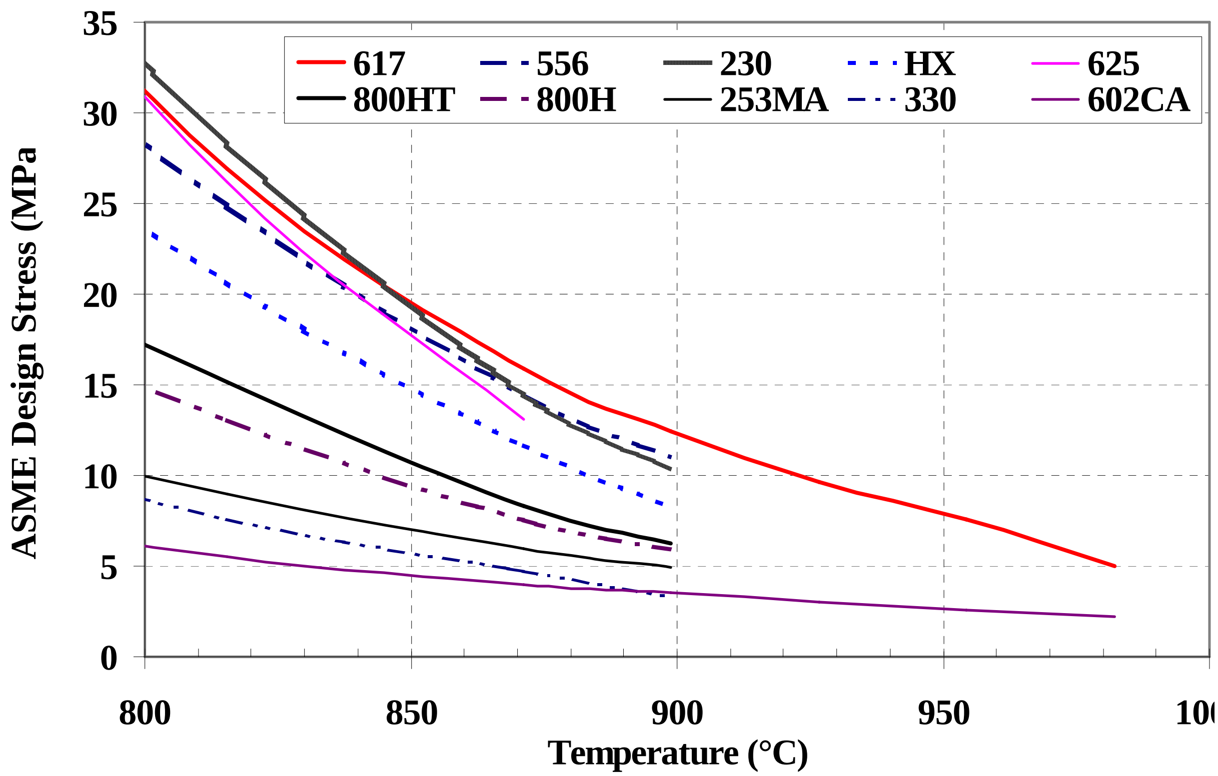

Fig. 4. Comparison of ASME allowable stress for high temperature alloys.

The University of Nevada, Les Vegas (UNLV) and General Atomic have carried out corrosion studies of heat exchanger candidate materials[6] in the SI environment. More than 20 coupons from four classes of materials (refractory and reactive metals, superalloys and ceramics) were screened for 100 hours or more each in liquid HIx ($HI+I_2+H_2O$). Suitable candidate materials for the HI decomposition heat exchanger include Ta-40Nb, Nb-1Zr and SiC ceramic.

Based on material, mechanical and corrosion properties screening, candidate ceramic materials have been selected for the sulphuric acid decomposition heat exchanger. The preferred materials are Nb- and Ta- based alloys, and SiC based ceramic materials.[6]

In addition work is being conducted at MIT[7] to develop a material that can act as both the structural material for the heat exchanger and catalyst for the acid decomposition reaction. Alloy 800 and 617 plus up to 30 wt% platinum were studied, and two alloy chemistries were identified: alloy 800 with 1wt% Pt addition and alloy 617 with 1wt% Pt addition.

III.C. PCHE/FPHE: Alloy 617 Development work

PCHEs are constructed by chemically milling flow passages into flat metal plates, and then stacking and diffusion bonding the plates into a single block. The key fabrication technologies for PCHEs are:

- Diffusion bonding
- Flow passage formation

The process of diffusion bonding is dependent on a number of proprietary parameters, including time, pressure, and temperature[8]. Diffusion bonding can be categorised into a number of variants, dependent on the form of pressurisation, the use (or not) of interlayers and the formation (or not) of a transient liquid phase. Each finds specific application dependent on the materials and geometries that need to be joined.

Recent alloy development work at Heatric revealed that alloy 617 can be successfully diffusion bonded without using an interlayer. A typical bend test for the diffusion-bonded alloy 617 is shown in Fig. 5, which demonstrates no cracks and good ductility. Tensile tests at room temperature, 538°C (1000°F) and 982°C (1800°F) were also performed to confirm the mechanical properties through the bonds meet ASME parent metal requirements (Fig. 6 and Table III). Helium leak tests were performed on diffusion-bonded alloy 617 cores, the results confirmed that the bonds were leak tight (Fig. 7). A typical micrograph (Fig. 8) shows the grain growth across the bonded interface.

Fig. 5. Bend test for diffusion bonded alloy 617.

Fig. 6. Tensile test for diffusion bonded alloy 617.

Table III

Tensile Test Results for Diffusion Bonded Alloy 617

Temp (°C)	0.2%PS (MPa)	UTS (MPa)	%EL
RT	334	696	52
538	195	517	51
982	93	159	44

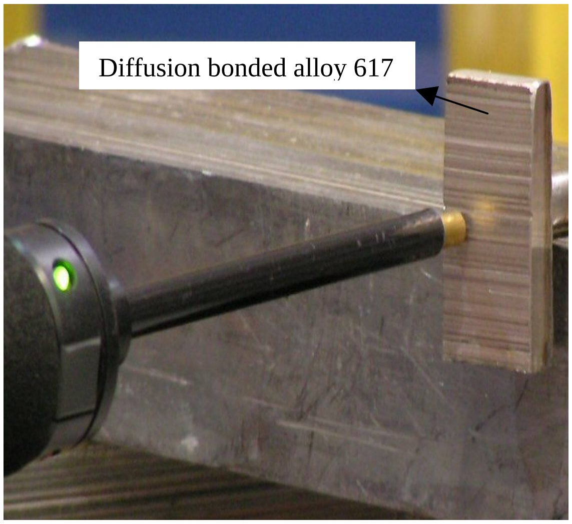

Fig. 7. Leak test on diffusion bonded alloy 617.

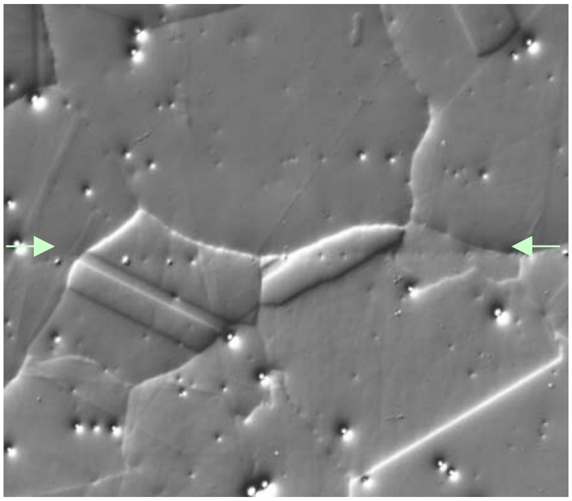

Fig. 8. Optical micrograph of diffusion bonded alloy 617 showing grain growth across the joint interface.

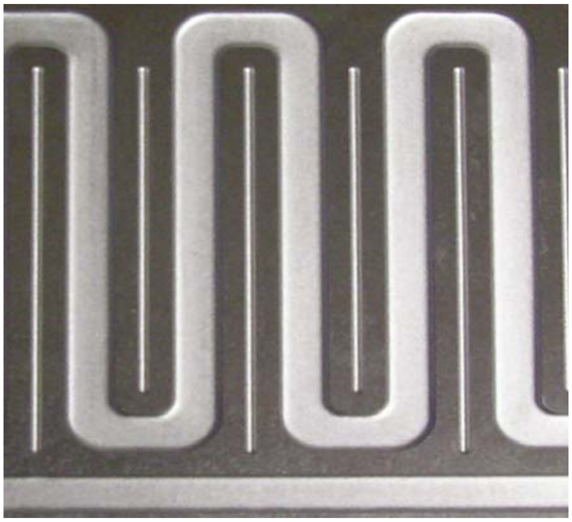

Fig. 9. Chemical machined alloy 617 plate.

Fig. 10. Different forms of formed plate.

In addition to Heatric's traditional method of passage forming, photo chemical machining (also called chemical milling), alternative processes have been developed to address the challenge associated with high temperature materials including electro-chemical machining and formed plate construction. Formed plate (FPHE) is a diffusion bonded plate fin derivative.

Selection of the appropriate passage forming technology is dependent upon alloy composition, properties and cost.

A typical alloy 617 photo-chemically machined plate is shown in Fig. 9. Fig. 10 shows typical forms of geometry that are available with formed plate.

The work on 617 laid the foundation not just for high temperature PCHEs, but also open the possibility for developing self-catalytic materials for the SI process, for example using 800H and 617 with platinum additions.[7]

IV. CONCLUSIONS

1) Heat exchangers such as Heatric's PCHE and FPHE products are fully developed, commercially available and capable of meeting the heat exchange requirements of the next generation of nuclear reactor.

2) The thermal, hydraulic, mechanical and life characteristics of the PCHE and FPHE are fully developed.

3) Pressure vessel design codes need to be established and developed to meet the requirements of the next generation of nuclear reactor. Moreover these codes need to give consideration to the appropriate compact heat exchange technologies and the required materials, which may need to include ODS and ceramic materials.

4) Of the conventional nickel based alloys, alloy 617 or alloy 230 appear to be the most suitable materials for manufacturing high temperature IHX in helium/ helium nitrogen environments. However, more research relating to high temperature time dependent properties and the effects of environment is required.

5) Heatric have successfully developed a technique to diffusion bond Alloy 617 with parent metal properties achieved, without using an interlayer.

6) Heatric have developed heat exchanger fabrication techniques, including passage forming procedures, compatible with current and emerging material requirements.

7) For more corrosive environments such as nuclear hydrogen production (SI process), ceramics (or non conventional pressure vessel materials) may be a requirement of the heat exchanger and other pressure boundary components.

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