Materials for Nuclear Diffusion-Bonded Compact Heat Exchangers

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Abstract – *This paper discusses the characteristics of materials used in the manufacture of diffusion bonded compact heat exchangers. Heatric have successfully developed a wide range of alloys tailored to meet process and customer requirements. This paper will focus on two materials of interest to the nuclear industry: dual certified SS316/316L stainless steel and nickel-based alloy Inconel 617.*

Dual certified SS316/316L is the alloy used most widely in the manufacture of Heatric's compact heat exchangers. Its excellent mechanical and corrosion resistance properties make it a good choice for use with many heat transfer media, including water, carbon dioxide, liquid sodium, and helium. As part of Heatric's continuing product development programme, work has been done to investigate strengthening mechanisms of the alloy; this paper will focus in particular on the effects of nitrogen addition.

Another area of Heatric's programme is Alloy 617. This alloy has recently been developed for diffusion bonded compact heat exchanger for high temperature nuclear applications, such as the intermediate heat exchanger (IHX) for the very high temperature nuclear reactors for production of electricity, hydrogen and process heat. This paper will focus on the effects of diffusion bonding process and cooling rate on the properties of alloy 617

This paper also compares the properties and discusses the applications of these two alloys to compact heat exchangers for various nuclear processes.

I. INTRODUCTION

Nuclear energy is becoming more important due to increasing energy demand, concerns over climate change and reducing dependence on fossil fuels. There are now approximately 440 commercial nuclear power reactors operating worldwide, which contribute 16% of the world’s electricity. In addition more power reactors are planned or under construction and their efficiency is increasing. There are also a number of national and international programmes to develop advanced nuclear reactors such as the “Generation IV” reactors. This results in many different types of nuclear reactors. A common practice is to classify power reactors according to the nature of the coolant, which passes through the nuclear reactors and transfers the nuclear heat to the designed applications. These include gases, water, supercritical fluids, liquid metals and molten salts.

Depending on the type of nuclear power plant, its specific design features, operation conditions and particular application, the requirements for heat exchangers, one of the key components to transfer the nuclear heat, will vary. For example, supercritical carbon dioxide (SCO_2) heat exchangers typically have a relatively high design pressure, at temperatures compatible with SS316/316L. On the other hand, intermediate heat exchangers generally have a very high design temperature, for which only a few materials such as Inconel alloy 617, are suitable. However, the general requirements are as follows:

- High Reliability
  - Assured leak tightness
  - Compatible with environment
  - Long life

- Good Performance
  - High effectiveness
  - Low pressure loss
- Cost Reduction
  - Reducing quantity of materials used
  - Reducing installation and operating costs

Of the currently available technologies, compact heat exchangers such as Heatric′s Printed Circuit Heat Exchanger (PCHE), Formed Plate Heat Exchanger (FPHE) and Hybrid Heat Exchanger (H^2X) are leading candidates for meeting these requirements.[1,2,3] Core samples from these heat exchangers are shown in Fig. 1. The benefits of these heat exchangers include space and weight saving, high thermal effectiveness, low pressure drop, close temperature approach, higher design pressures, and greater flexibility, permitting unique, bespoke designs for demanding duties.

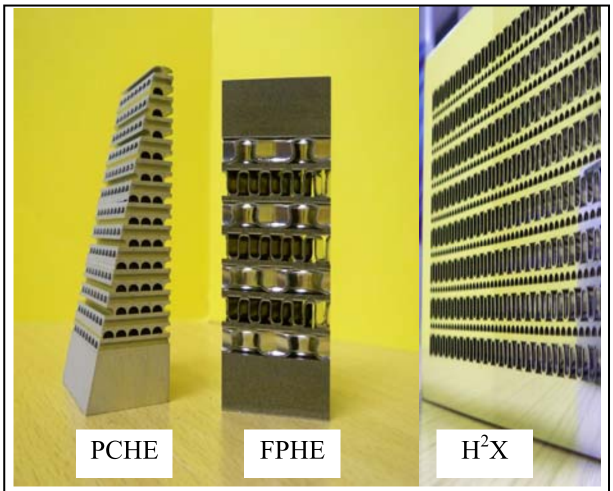

Fig. 1. Core samples from PCHE, FPHE and H^2X

A wide range of alloys have been successfully developed for the manufacture of the above diffusion-bonded compact heat exchangers. These include but are not limited to 300 series austenitic stainless steels, 6Mo super austenitic stainless steels, duplex stainless steels, titanium alloys, nickel and nickel-based alloys.

This paper will focus on dual certified SS316/316L and alloy 617. The former is suitable for many applications and the latter is the leading candidate material for the high temperature exchangers.[4,5]

II. MATERIAL REQUIREMENTS

The main requirements for heat exchanger materials are their properties. This section will discuss these requirements with emphasis on the physical properties, mechanical properties and corrosion resistance of dual certified SS316/316L and alloy 617.

II.A. Physical Properties

A suitable alloy for the heat exchangers should have favourable physical properties, especially thermal conductivity and coefficient of thermal expansion.

The material thermal conductivity is directly associated with the overall heat transfer coefficient of heat exchangers and a higher value is beneficial. For SS316/316L and alloy 617, the thermal conductivity at room temperature is about 13-14 W/(m K) and it increases as temperature rises.

Temperature changes within heat exchangers under transient conditions such as start-up and shut-down can generate thermal stresses when thermal expansion is restrained. These thermal stresses are generally proportional to the alloy′s thermal expansion coefficients. High local thermal stresses could eventually initiate and propagate fatigue cracks, therefore materials with lower thermal expansion coefficients may be preferable for heat exchangers with high thermal gradients. Generally speaking, materials with higher nickel content will have lower coefficients of thermal expansion. This is true for alloy 617, which has a lower thermal expansion coefficient than dual certified SS316/316L.

II.B. Mechanical Properties

Heat exchangers are pressure vessels, therefore mechanical properties of materials are very important. The selected alloys must meet code (such as ASME) required mechanical properties. For dual certified SS316/316L stainless steel, the required ASME minimum values at room temperature are 205 MPa for 0.2% proof strength (0.2%PS), 515MPa for ultimate tensile strength (UTS) and 40% for elongation (%El). For alloy 617, these values are 240MPa, 655MPa and 30% respectively.

The mechanical design of heat exchangers is directly related to the maximum allowable stresses of the material. These values are established based on both instantaneous mechanical properties and creep properties of the material.[6] Table I lists the ASME maximum allowable stresses for dual certified SS316/316L austenitic stainless steel (up to 550°C) and alloy 617 (up to 982°C). Since the onset creep temperature for Type 316 is 595°C, the maximum allowable stresses for SS316/316L stainless steel are based on its tensile strengths. For alloy 617, the ASME onset creep temperature is 825°C. Below this temperature, the stress levels decrease gradually with increasing temperature. Above 825°C, the stresses, which are based on the creep-rupture life at 100000 hours, decrease rapidly with increasing temperature.

TABLE I

ASME Maximum Allowable Stresses of Dual Certified SS316/316L Austenitic Stainless Steel and Alloy 617

Temperature (°C)	Max Allowable Stresses (MPa)	
	SS316/316L (up to 550°C)	Alloy 617 (up to 982°C)
RT	138	161
200	134	161
400	111	150
550	105	144
800	~	31.30
900	~	12.30
950	~	7.91
982	~	5.00

In order to improve cycle efficiency and reduce replacement cost, a higher operating temperature and longer lifetime are desirable for nuclear heat exchangers. To provide a reliable service under such conditions, creep, fatigue and creep-fatigue properties of materials need to be considered. Although many research works have been reported in this area,[7] the requirements of these properties are not straightforward. This is because a number of variables, such as test temperature, environment, strain range, frequency, time and type of hold, waveform, material property and damage characteristics, affect the fatigue life. The conclusions drawn in any investigation may therefore apply only to the materials and test conditions used in that study. In addition, the models for fatigue and creep damage prediction and creep-fatigue interaction have to be verified for a specific application. Correctly assessing the lifetime of a heat exchanger involves a significant amount of work. For example, in order to predict the thermal fatigue life of a heat exchanger component, the temperature distributions within the heat exchanger under many types of transient conditions must be predicted. This first requires the variations in mass flow, temperature and pressure to be correctly predicted - a task that is challenging in itself, as the behaviour of many new related components in the main power system such as the gas turbine needs to be simulated correctly. Nevertheless for the diffusion-bonded compact heat exchangers, Heatric has developed a finite difference programme to calculate fluid and metal temperature, fluid flow, and pressure change distributions within the heat exchanger under steady state and transient conditions. This tool is used to predict heat exchanger thermal-hydraulic response, and is also used to facilitate mechanical analyses and materials selection.

II.C. Corrosion Resistance

The material selection for nuclear heat exchangers has to consider the issues of material-coolant interaction under specific service conditions.

For an intermediate heat exchanger (IHX) in a very high temperature reactor, one particular challenge will be the control of impurities such as H_2, H_2O, CO, CO_2 and CH_4 in the coolant. Otherwise, all candidate materials including alloy 617 could face a risk of oxidation, carburization and decarburization.[8,9,10,11] If the secondary coolant contains significant nitrogen, the nitriding resistance of materials should be considered.[12] Nickel based alloys generally have good nitridation resistance due to the low solubility of nitrogen in these metals. Iron tends to be detrimental, as does low concentration aluminium or titanium. Silicon forms a brittle intermetallic compound with nitrogen and can contribute to scale spallation, especially in applications with low oxygen concentrations and during thermal cycling.[4]

In the supercritical water reactor concept, which is designed for high thermal efficiency and plant simplification at pressure in excess of 22MPa, the main material challenges are related to the supercritical fluid coolant and high operating temperature. At the critical point there is a dramatic change in most of the physical properties of water including density, specific heat and viscosity. The core outlet temperature is between 500 and 620°C. One of the material requirements is low susceptibility to stress corrosion cracking under supercritical water condition. In austenitic stainless steels, this type of corrosion results from sensitisation of certain grain boundaries caused by a complex interaction of stress, material, temperature, pressure and environment.

In summary, there are many requirements for nuclear heat exchanger materials, irrespective of the exact design or purpose. The material must have adequate availability, fabrication and joining properties. In addition, the material must have good physical and mechanical properties, and compatibility with reactor coolants. The optimal heat exchanger type and configuration are in turn strongly influenced by alloy selection.

III. DUAL CERTIFIED SS316/316L AUSTENITIC STAINLESS STEEL

For Type SS316L to be dual certified as Type SS316 and Type SS316L, defined as SS316/316L in this paper, the material must meet both the lower carbon limit of Type SS316L and the higher strengths of Type SS316.

Work has been carried on two groups of SS316L austenitic stainless steel. Group one is standard SS316L and group two has an enhanced nitrogen content, referred to as SS316L(N). The material properties in as-received and diffusion-bonded conditions have been investigated and the main results are presented in this section.

III.A. As-Received SS316L and SS316L(N) Alloys

The as-received SS316L and SS316L(N) sheets were cold-rolled and then solution annealed with a nominal thickness of 1.65mm. The chemical compositions of these two alloys are given in Table II. The ASME material specifications for Type SS316L and Type SS316 are also listed in this table. The only difference between the two types is the maximum carbon content. Type SS316L, with a maximum 0.03 wt% carbon, is the low carbon version of Type SS316 and is immune from sensitisation (grain boundary carbide precipitation).

TABLE II

Material Specifications and Chemical Compositions of As-Received SS316L and SS316L(N) Alloys (wt%)

Elements	SS316L	SS316L(N)	ASME 316L	ASME 316
C	0.018	0.014	0.03	0.08
Si	0.43	0.45	0.75	0.75
Mn	0.93	0.86	2.00	2.00
P	0.024	0.030	0.045	0.045
S	0.001	0.002	0.030	0.030
Cr	17.2	17.4	16-18	16-18
Ni	11.2	10.3	10-14	10-14
Mo	2.03	2.07	2.0-3.0	2.0-3.0
N	0.042	0.080	0.100	0.100

TABLE III

ASME Requirements and Mechanical Properties of as-Received SS316L and SS316L(N) Alloys

	0.2% PS (MPa)	UTS (MPa)	El%
SS316L	309	626	63
SS316L(N)	374	680	53
ASME 316L	170	485	40
ASME 316	205	515	40

The mechanical properties of as-received SS316L and SS316L(N) sheets are shown in Table III. The ASME mechanical test requirements for Type SS316L and Type SS316 are also given in this table. Since the mechanical strengths of Type SS316 are higher than those of Type SS316L, this allows a reduction in the wall thickness of SS316/316L heat exchangers which results in both weight and cost savings.

The mechanical properties for all as-received sheets at room temperature met the ASME mechanical test requirements. The minimum values from type 316L also met the specifications for the standard grades of SS316. This means that in the as-received condition, type SS316L could be dual certified as SS316/316L.

For the two alloys discussed here, the mechanical strengths are mainly governed by the Hall-Petch relationship, i.e. the strength of the material comes from the contributions of alloying elements and grain size. Out of all the elements added, nitrogen is the most effective element in increasing material strengths followed by the carbon. The measured grain sizes for as-received SS316L and SS316L(N) sheets are 12 and 6 microns respectively. Thus the higher values of 0.2%PS (374 MPa) and UTS (680 MPa) for as-received SS316L(N) sheet are mainly due to its higher nitrogen content (0.080 wt%) and finer grain sizes than those of alloy SS316L.

Only one matrix phase exists in the as-received SS316L and SS316L(N) sheets. A typical micrograph is shown Fig. 2, which reveals a fine and eqiaxed grain structure for the as-received SS316L sheet.

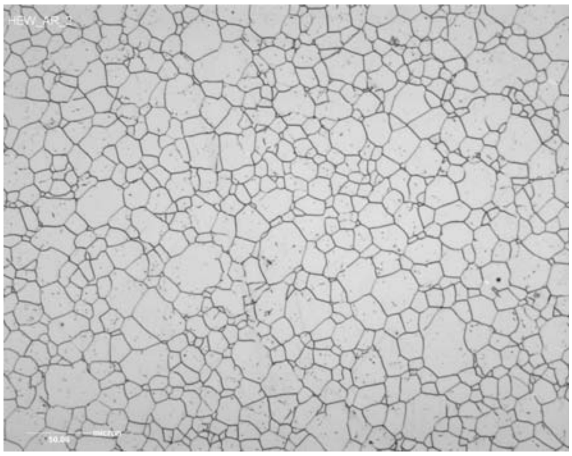

Fig. 2. Micrograph of as-received SS316L stainless steel sheet showing fine and eqiaxed grain structure

III.B. Diffusion-Bonded SS316L and SS316L(N) Alloys

To investigate the effect of alloy element and diffusion-bonding parameter on mechanical properties of diffusion-bonded SS316L and SS316L(N) alloys, many bonding trials were performed. Fig. 3 shows typical tensile test results for diffusion-bonded SS316L and SS316L(N) materials tested in the bonding direction (perpendicular to the bonded plates).

The minimum values of 0.2%PS, UTS and elongation for the diffusion-bonded alloy SS316L(N) are 208MPa, 545MPa and 81%. These values met the ASME mechanical test requirements for Type SS316 austenitic stainless steel as shown in Table III. This means that the diffusion-bonded alloy SS316L(N) can be dual certified as SS316/316L stainless steel.

Without the enhanced nitrogen (alloy SS316L), the 0.2% proof strength of diffusion-bonded material (186 MPa) may only meet the ASME requirement for Type SS316L (170MPa) but not for Type SS316 (205MPa). This means that without specified high nitrogen content, the diffusion-bonded SS316L alloy could not be dual certified as SS316/316L stainless steel.

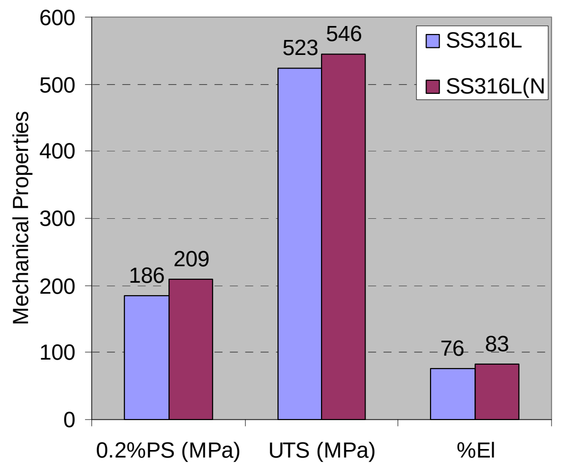

Fig. 3. Mechanical properties of diffusion-bonded SS316L and SS316L(N) alloys

The measured grain size for both alloys after diffusion bonding process is approximately 200μm. Thus, the grain size contribution to the mechanical strengths of the diffusion-bonded alloys can be treated as a constant. Apart from a very different level of nitrogen, the compositions of alloy SS316L and alloy SS316L(N) (Table II) are very similar. It is therefore reasonable to conclude that the improved mechanical properties of diffusion-bonded alloy SS316L(N) are mainly due to the higher nitrogen addition. This has been confirmed by additional tensile tests at room temperature (RT), 400°C and 530°C as shown in Fig. 4.

The addition of nitrogen can also retard the precipitation of carbides and improve corrosion resistance of the material.[13] Additional benefit includes increasing creep and fatigue life under creep-fatigue condition.[14,15]

Metallographic examination reveals that the grains have grown across the joint interface in both diffusion-bonded alloys. A selected micrograph near plate edges in shown in Fig. 5.

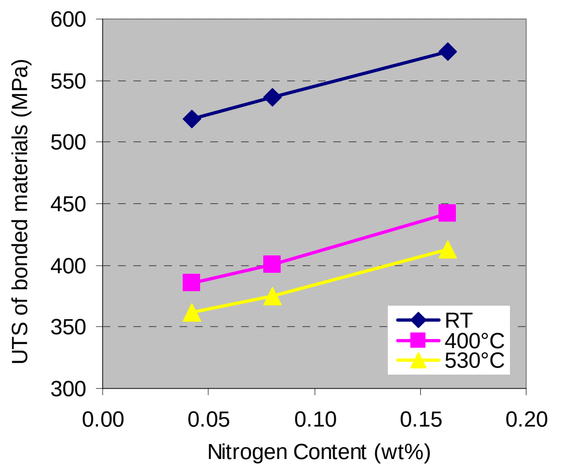

Fig. 4. Effect of nitrogen on tensile strength of diffusion bonded 316 series stainless steels

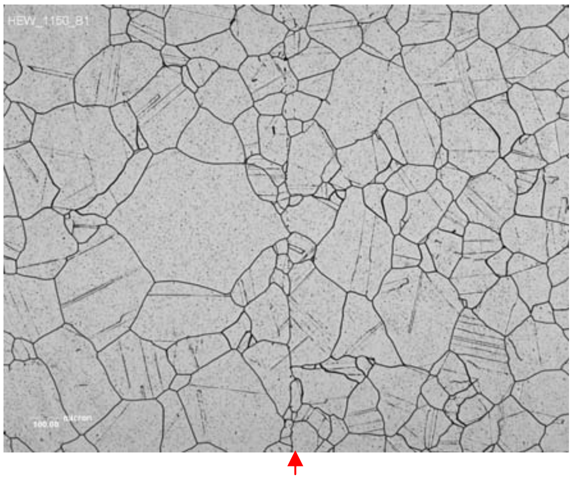
Fig. 5. Microstructure of diffusion-bonded SS316/316L sheet showing grain growth across the joint interface

Tests were also performed in the parent metal (parallel to the bonded plates) direction of diffusion-bonded SS316/316L alloys. Fig. 6 shows the comparison of 0.2% Proof Strength (0.2%PS) in both the bonding and parent metal directions. The results at various test temperatures show that the mechanical property difference between the bonding and parent metal directions is within 5% and the ASME requirements are always exceeded. Thus the mechanical properties of the diffusion-bonded materials could be treated as isotropic. This further confirms the high integrity of our diffusion bonding process which has no degradation of parent metal mechanical properties.

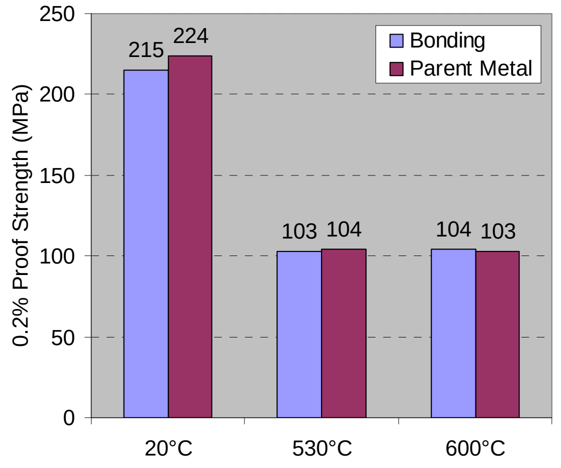

Fig. 6. Comparison of 0.2%PS for diffusion-bonded SS316/316L alloys in bond and parent directions

III.C. Applications of SS316/316L heat exchangers

Depending on the technique used to form the fluid flow channels on the metal plate, the diffusion bonded compact heat exchangers can be characterized as Printed Circuit Heat Exchanger (PCHEs), Formed Plate Heat Exchangers (FPHEs) or Hybrid Heat Exchangers (H^2Xs). For PCHEs, the fluid flow channels are formed by photo chemical machining or electrochemical machining. For FPHEs, the fluid flow passages are manufactured by mechanical fin forming. The fluid flow passages in H2Xs are a combination of those from both PCHEs and FPHEs. These heat exchangers especially the PCHEs have been widely used in the oil, gas and petrochemicals industry for over 20 years. The products are receiving much interest from the nuclear industry. Examples of the nuclear applications are:

- SCO_2 recuperators and gas chillers in supercritical CO_2 Brayton cycle test loop[16]
- Recuperators for the helium-cooled Pebble Bed Modular Reactor (PBMR)[17]
- Mercury cooler for the high power proton accelerator project
- Steam generator for nuclear marine propulsion
- Gas – water cooler
- Gas-gas and sodium-gas heat exchangers for gas-cooled or sodium-cooled reactors.[18] The gas here can be nitrogen, helium, helium-nitrogen mixture or supercritical CO_2

IV. NICKEL BASED ALLOY 617

This section presents the results from Heatric's alloy 617 development programme for diffusion bonded compact heat exchangers. These results include microstructural characterization and mechanical testing in both as-received and post-bonding conditions.

IV.A. As-Received Alloy 617

Alloy 617 (UNS N06617) is a solid solution, nickel-chromium-cobalt-molybdenum alloy with exceptional high temperature metallurgical stability and mechanical properties. The alloy also has excellent resistance to a wide range of corrosive environments and is readily formed and welded by conventional techniques.

Two heats of alloy 617 sheets, which have different sheet thickness and grain size, have been investigated. The chemical compositions of the as-received Alloy 617 sheets fully comply with the ASME material specification for the alloy as shown in Table IV.

TABLE IV

ASME Material Specification and Chemical Compositions of Alloy 617 Studied (wt%)

	Heat 1	Heat 2	ASME
Ni	Bal	Bal	44.5 min
Cr	21.5	21.5	20.0-24.0
Co	12.20	12.05	10.0-15.0
Mo	9.31	9.26	8.0-10.0
Fe	2.12	1.24	3.00
Mn	0.12	0.06	1.00
Al	0.92	0.96	0.8-1.5
C	0.058	0.060	0.05-0.15
Cu	0.01	0.06	0.5
Si	0.08	0.11	1.0
S	<0.001	0.006	0.015
Ti	0.41	0.38	0.60
B	<0.001	0.002	0.006

The mechanical properties of as-received alloy 617 sheets and ASME mechanical test requirements are shown in Table V.[19] Both heats met the ASME requirements. One of the many factors which contribute the higher mechanical strengths of Heat 2 alloy 617 is its finer grain size.

TABLE V

ASME Mechanical Test Requirements and Mechanical Properties of as-Received Alloy 617

	0.2% PS (MPa)	UTS (MPa)	El%
Heat 1	345	759	62
Heat 2	352	825	55
ASME 617	240	655	30

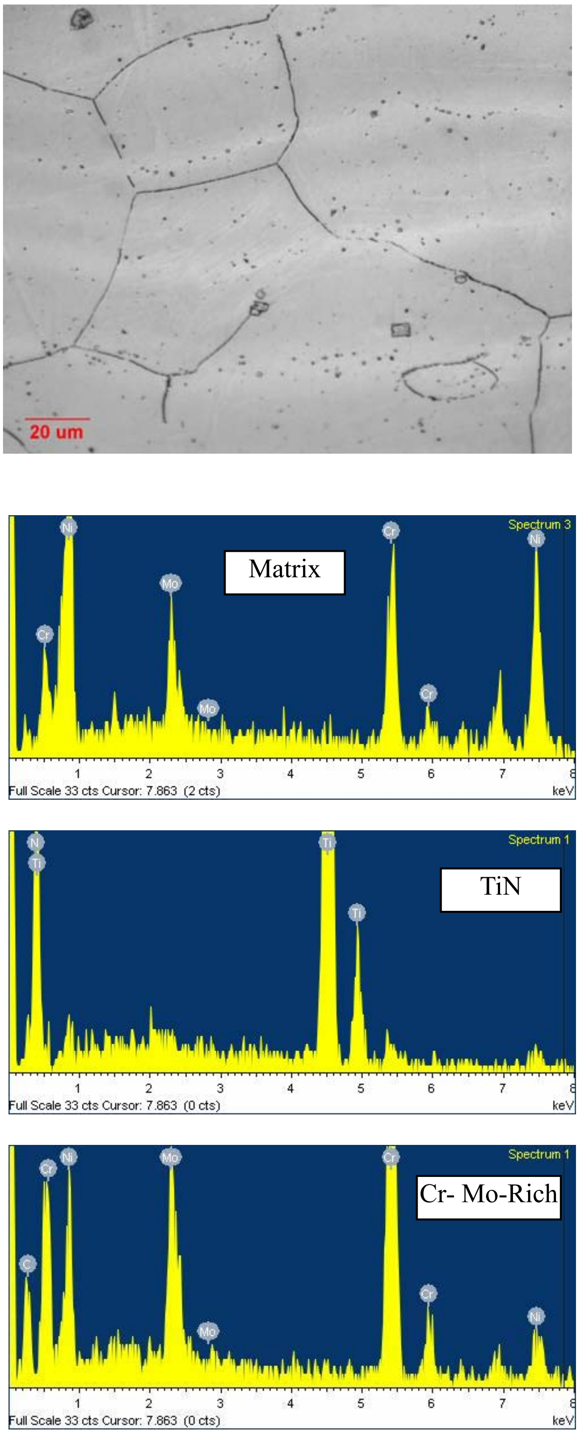

Fig. 7. Microstructure and EDX analysis of as-received alloy 617 showing the presence of FCC matrix, Titanium nitrides and Cr- and Mo-rich carbides

Compared to SS316/316L stainless steel, alloy 617 has a coarse grain structure. A typical microstructure of as-received alloy 617 is shown in Fig. 7. The major phase present is an austenitic nickel matrix in which chromium, cobalt and molybdenum are dissolved as solute elements. Small amounts of precipitates were found within the matrix as titanium nitrides (cubic shape) and Cr- and Mo-rich carbides (spherical shape within the matrix and elongated shape on grain boundaries). These phases are confirmed using the Energy Dispersive X-ray (EDX) analysis (Fig. 7).

IV.B. Diffusion-bonded alloy 617

The above alloy 617 sheets were diffusion-bonded under controlled bonding parameters. The quality of the bonds were evaluated by both mechanical testing and microstructure examination.

The tensile test results at room temperature are presented Table VI. The direction of the tests was perpendicular to the diffusion bonded plates. The values of 0.2% proof strength (0.2%PS), ultimate tensile strength (UTS) and elongation all met the ASME mechanical test requirements for alloy 617. Slightly better mechanical properties of Heat 2 diffusion-bonded alloy 617 have been noticed. Although the strengths are consistent with the as-received material properties (Table IV), further work is needed to establish the effect of chemical composition and grain size on mechanical properties of the diffusion-bonded alloy 617.

TABLE VI

Mechanical properties of diffusion-bonded alloy 617

	0.2% PS (MPa)	UTS (MPa)	El%
Heat 1	320	663	41
Heat 2	336	682	46

Microstructure examination for all diffusion-bonded materials reveals grains have grown across the entire joint metal interface and a typical micrograph is shown in Fig. 8.

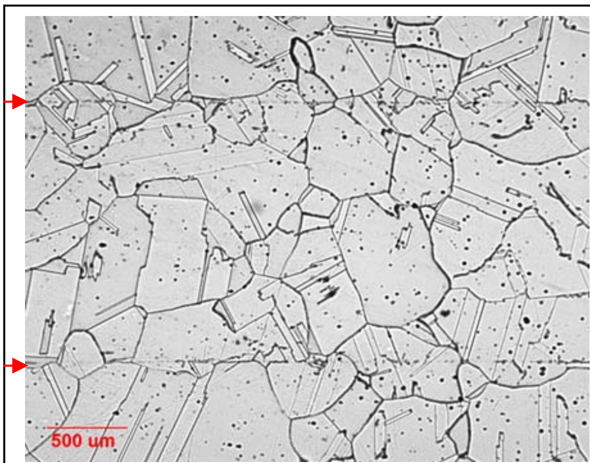

Fig. 8. Microstructure of diffusion-bonded alloy 617 showing grain growth across the joint metal surfaces

The mechanical properties of diffusion-bonded alloy 617 are directly related to the cooling rate of the post-bond heat treatment, which is normally performed between 1150°C and 1200°C. As shown in Fig. 9, the post-bond heat treatment followed by water quench (WQ) is essential to achieve the ASME required mechanical properties. The worst mechanical properties result from the slowest cooling of furnace cooling (FC) and their values are much lower than those in the as-bonded condition (AB). The mechanical properties of as-bonded alloy 617 can be improved by the post-bond heat treatment followed by the air cooling (AC). But the UTS value (649MPa) is still below the ASME required 655MPa. These results confirmed the sensitivity of microstructure involving grain size, amount and type of precipitates and their distribution on mechanical properties of diffusion-bonded alloy 617.

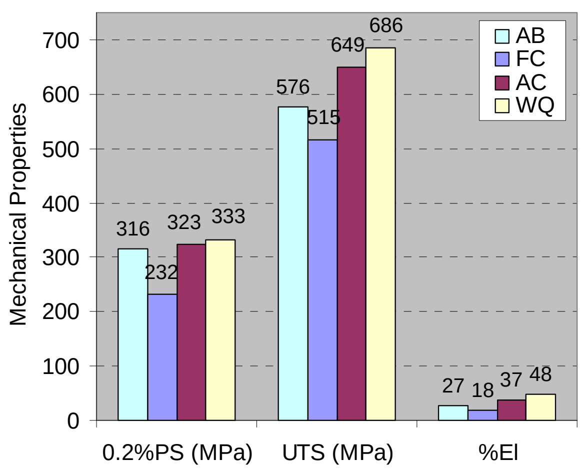

Fig. 9. Effect of cooling rate on mechanical properties of diffusion-bonded alloy 617

IV.C. Applications of alloy 617 heat exchangers

As reported previously, Heatric has successfully developed the fabrication technique of alloy 617 diffusion-bonded heat exchangers.[20] This includes the formation of fluid flow passages on alloy 617 plates and joining these plates together through diffusion bonding. The high integrity of the diffusion bonds was confirmed through mechanical testing and microstructure examination. The diffusion-bonded alloy 617 heat exchanger core achieved a leak-tightness of 8.6E-9 mbar.l/s, which exceeded our in-house standard of 2.0E-7 mbar.l/s and the standard for brazed Al plate fin heat exchangers of 1.0E-3 mbar.l/s.[21]

The diffusion-bonded alloy 617 compact heat exchanger is mainly for high temperature applications. Based on the ASME Section VIII Div.1, alloy 617 has the highest maximum temperature limit of 982°C. Therefore Heatric's high-integrity heat exchangers, designed and constructed in alloy 617, would be an excellent candidate for high-temperature heat exchangers for the Next Generation Nuclear Plant (NGNP), and for other high temperature applications.[22]

V. CONCLUSIONS

Heatric can manufacture diffusion bonded heat exchangers (PCHE, FPHE and H^2X) in a range of alloys to meet the demands of the heat exchanger markets.

This paper discusses dual certified SS316/SS316L and alloy 617 which market research has identified as leading candidate materials for the nuclear renaissance and NGNP.

From the information presented in the paper the following conclusions can be drawn:

- Heatric's bonding process results in material properties that meet or exceed the requirements of ASME for both SS316 and alloy 617
- Techniques to form fluid flow passages are developed and characterised
- Welding and other manufacturing techniques have been developed to ASME standards.

The selection of one of these two alloys should meet many of the requirements of the duties, fluids and operating parameters associated with next generation nuclear reactors.

SS316 is compatible with helium, nitrogen, carbon dioxide and liquid sodium and dependent upon creep life requirements and environments will provide an economic solution for nuclear heat exchangers with design temperatures up to 550°C and in some circumstances beyond that temperature.

Beyond those temperatures the metallurgical and mechanical properties of alloy 617 is suitable for most applications with design temperatures up to and in excess of 900°C.

NOMENCLATURE

PCHE	Printed Circuit Heat Exchanger
FPHE	Formed Plate Heat Exchanger
H^2X	Hybrid Heat Exchanger
IHX	Intermediate Heat Exchanger
SCO_2	Supercritical Carbon Dioxide

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