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Diffusion Welding of Compact Heat Exchangers for Nuclear Applications

Trends in Welding Research

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Diffusion Welding of Compact Heat Exchangers for Nuclear Applications

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Abstract

Diffusion welding is a useful technique for assembling plate-type heat exchangers. The present work examines the diffusion welding characteristics of alloys of interest in advanced energy systems, including Alloy 800H (~45Fe-35Ni-20Cr), Alloy 617 (~55Ni-22Cr-12Co-9Mo-2Fe), Alloy N (~71Ni-17Mo-7Cr-5Fe), and Alloy 242 (~67Ni-25Mo-8Cr). A diffusion welding procedure was developed for the Gleeble thermomechanical testing machine, which allowed the critical welding parameters for diffusion welding to be explored efficiently. Stacks composed of many sheets were also welded in a vacuum hot press. Composition profiles across the finished joints were measured with SEM X-ray spectroscopy, and compared with models developed with Thermocalc/DICTRA to evaluate such modeling as a tool for rapid parameter development, and agreement was generally good. Appropriate surface treatments were found to be critical to achieving good mechanical properties, which were typically >90% of base metal properties. Diffusion welds in these alloys led to fully integrated grain structures across the previous joint. In addition, slight differences in diffusion with crystallographic direction were noted.

Keywords

Diffusion welding, compact heat exchanger, printed circuit heat exchanger, Gleeble

Introduction

The Next Generation Nuclear Plant (NGNP) program is tasked with investigating the application of a new generation of nuclear power plants to a variety of energy needs.[1] One baseline reactor design for this program is a high temperature, gas-cooled reactor (HTGR), which provides many options for energy use. These might include the conventional Rankine cycle (steam turbine) generation of electricity, but also other methods: for example, Brayton cycle (gas turbine) electrical generation, and the direct use of the high temperatures characteristic of HTGR output for process heat in the chemical industry (Fig. A-1). Another possible high-temperature source is a reactor with a molten salt working fluid, similar to the Molten Salt Reactor Experiment (MSRE).[2] Such process heat is currently generated by burning fossil fuels, and is a major contributor to the carbon footprint of the chemical and petrochemical industries.

The HTGR, based on graphite fuel elements, can produce very high output temperatures; ideally, temperatures of 900 °C or even greater, which has significant energy advantages. Such temperatures are, of course, at the frontiers of materials limitations, at the upper end of the performance envelope of the metallic materials for which robust construction codes exist, and within the realm of ceramic materials, the fabrication and joining of which, on the scale of large energy systems, are at an earlier stage of development.

Heat exchangers are required for transferring heat from the primary reactor coolant loop to process loops; the two loops generally need to be separated for radiological reasons. Heat exchangers come in many configurations, but the one examined here is the "compact heat exchanger," a plate-type heat exchanger in which many, relatively thin layers of material containing channels for fluid flow are sandwiched together in an arrangement that provides for efficient countercurrent flow, as shown in Fig. 1.[3] The particular configuration shown here is also called a "printed circuit heat exchanger" (PCHE) because the channels are formed by a photolithography and etching process similar to those used for electronic printed circuit boards.

An actual example of such a fabricated PCHE is shown in Fig. 2. The dimensions are approximately 10 x 10 x 30 cm; it was made by diffusion welding a stack of 0.062 in. Alloy 617 sheets between thicker top and bottom plates of the same material, machining to the desired final size, and welding on headers that allowed the device to be placed in a test loop to test its performance.

The present work examines the diffusion welding of materials of interest for process heat applications. Some problems (incomplete bonding and delamination) were encountered in the construction of the heat exchanger shown in Fig. 2,[3] and it was partly to address these and define their solution that the present work was undertaken. Because of material and construction code limitations, a target reactor outlet temperature of 750 °C was set, and Alloy 800H was chosen for the initial compact heat exchanger material. Most of the work described here will be in that material, although others were examined as well, including Alloy 617 (potentially useful up to 925 °C) and alloys developed for use with molten salts at about 700 °C, Haynes Alloys N and 242. In addition to the experimental work described here, modeling of the diffusion welding process with Thermocalc/DICTRA[4,5] was carried out to evaluate the usefulness of these relatively recently developed techniques for planning and evaluating the diffusion welding process.

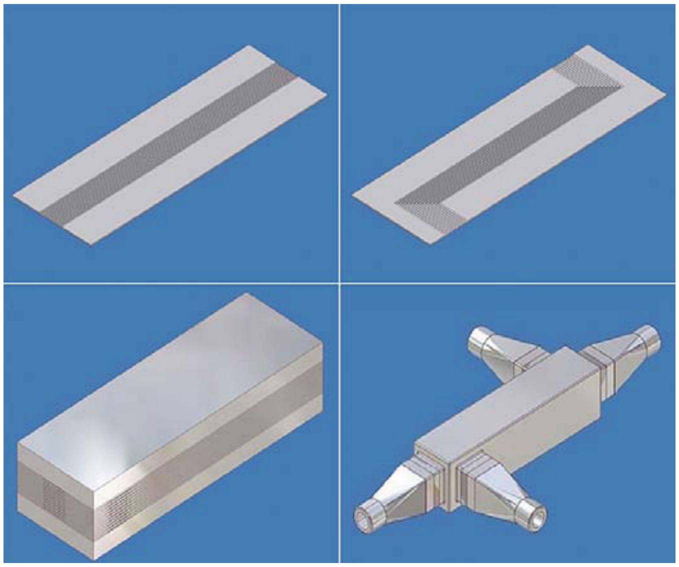

Fig. 1. The "printed circuit heat exchanger" concept for compact heat exchanger construction.

Diffusion Welding

Diffusion welding is an old welding process with wide application in many industries.[6] Relatively closely mating surfaces, held together under moderate pressure and at high temperatures (typically >0.6 Tm) will eventually, through diffusion processes, eliminate surface contaminants and oxides, and reduce their surface asperities to isolated pores along the bond line which are gradually filled. Grain boundary migration and grain growth dynamics can eventually produce a joint that is microstructurally indistinguishable from the base materials.

Diffusion welding is usually carried out in vacuum or protective atmosphere, to avoid excessive oxidation. The needed pressure can be applied mechanically or by dead weight loading, or the assembly to be welded can be canned and the pressure applied via hot isostatic pressing (HIP). Typically, the pressures needed are modest; particularly for intricate structures such as heat exchangers, creep must be avoided on a macro scale. On the other hand, the pressure must be evenly applied, sometimes over large areas and through many layers of material.

Metals that do not form highly stable oxides under ambient conditions, or that have a high solubility for their own oxides, are especially good candidates for diffusion welding. The alloys under study here were designed for high temperature service, so they have high strengths at high temperatures, and, with approximately 20% Cr, develop protective oxides. They are thus somewhat more challenging to diffusion weld than other alloys. Many austenitic stainless steels have similar Cr levels, however, and are commonly diffusion welded, so the basic technology is promising, even if some adaptation may be needed.

Surface preparation is important in diffusion welding. Plated surfaces are sometimes used for oxidation protection during heating, and "filler metal," typically made from a metal foil, is sometimes used to improve contact by local deformation, and to enhance diffusion. Surface treatment with other easily diffusible materials, for example, boron (short of the amount required for the process variant of "transient liquid phase bonding"), can also enhance the kinetics of the process.

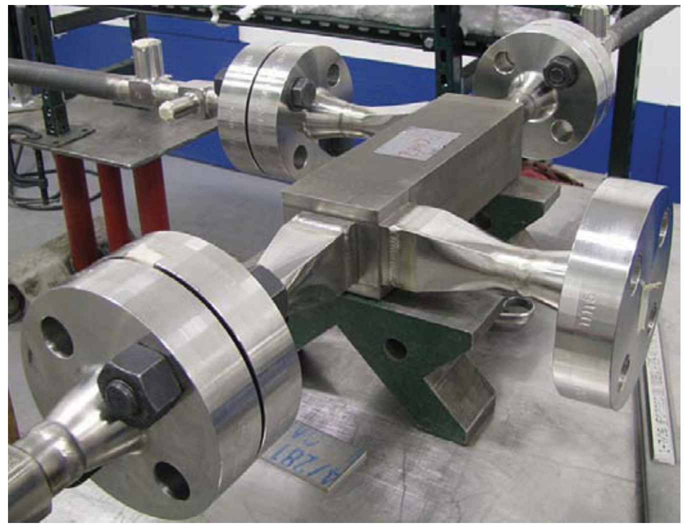

Fig. 2. A compact heat exchanger fabricated by diffusion welding from Alloy 617, in a test loop designed to measure performance with helium gas up to 900 °C.

Experimental

Gleeble

Most of the diffusion welding reported here was done using the INL Gleeble. The Gleeble is well known for its versatility in many welding metallurgy studies, particularly the rapid thermal cycling and loading typical of fusion welding processes. It also turns out to be well equipped for diffusion welding, operating at the lower end of its force capabilities and the upper end of its sustained high temperature capabilities.[7] It contains an integrated vacuum system; its vacuum level of mid-10^{-5} torr is not as high as some of the furnaces used for diffusion welding because of the compromises (sliding seals, etc.) needed for most Gleeble work, but sufficient for the present work.

Vacuum hot press diffusion welding was also performed as part of the present work; stacks of Alloy 800H material meant to approximate the geometry of compact heat exchangers are shown in Fig. 3. The size of these stacks is 2.1 inches on a side, a limit imposed by the size of the hot press used. They are an intermediate size between the large press or HIP that would be used to fabricate actual heat exchanger modules and the Gleeble, which has a practical upper limit of about 1 inch in diameter.

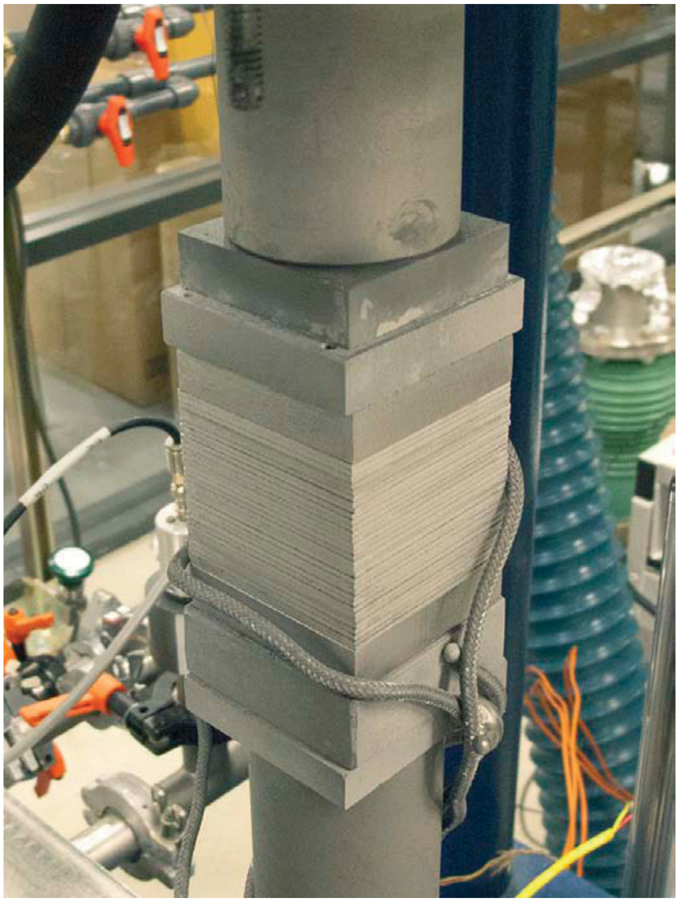

Fig. 3. Mockup compact heat exchanger stacks welded in a vacuum hot press, containing NN (left) and MM (right) sheets of 0.062 in. thick 800H sheets between 800H plates.

The parameter development, metallurgical, and modeling work reported here was done on the Gleeble. In general, compared with furnace methods, the Gleeble provides a faster turnaround, with smaller specimens, and a degree of observability during welding, as can be seen in Fig. 4(a).

The Gleeble is a servohydraulic machine capable of applying heavy loads at fast stroke (20-ton load cell, ~1 m/s stroke rate). Diffusion welding requires nothing like these maximal conditions, and it was considered desirable not to run the hydraulic system, for the many hours diffusion welding sometimes requires. The auxiliary air ram system, normally used in positioning the system and applying an initial holding force to specimens, was used for the diffusion weld loading. Although the air ram is not under automatic feedback control, it was found that loading is stable, and small manual adjustments over the several hours of the diffusion weld cycle can keep specimen loading reasonably constant. In addition, it was found that a short, high load applied hydraulically at the beginning of the cycle (up to 40 MPa, ~5800 psi, or about 1100 lbf for the typical 0.5 in. diameter specimens used) tended to seat the specimens and take care of slight misalignments. An example of a typical loading cycle is shown in Fig. 5.

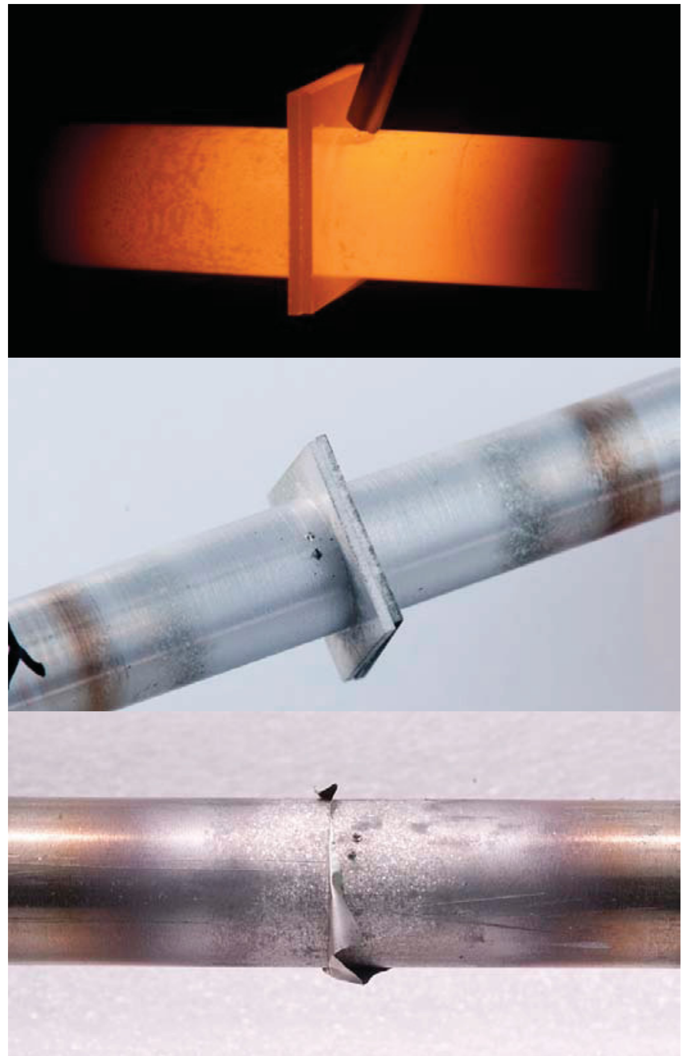

Fig. 4. (a) Alloy N base metal with Alloy N inserted sheets, diffusion weld in progress in the Gleeble; (b) same weld completed; (c) Alloy 800H weld with Ni foil interlayer.

The Gleeble grips specimens in water-cooled jaws and applies thermal cycles by power Joule heating using feedback from a thermocouple welded to the specimen, close to the diffusion joint (Fig. 4). There is thus a temperature gradient away from the joint that does not exist, for example, in furnace or HIP diffusion welding, and it might be expected to be steeper than that observed in vacuum hot press welding. Measurements with a second TC located 7 mm from the joint shows a temperature drop of about 20 °C. In most cases conditions directly at the bond line are of interest, and the TC is a good record of this temperature history.

A typical diffusion weld in progress is shown in Fig. 4(a), and the temperature gradient can be seen qualitatively by the thermal emission as well. In this particular weld, intermediate sheet material similar to a compact heat exchanger design was inserted. When more sheets are used (up to 9 have been successfully tried), the control TC is welded to the center sheet.

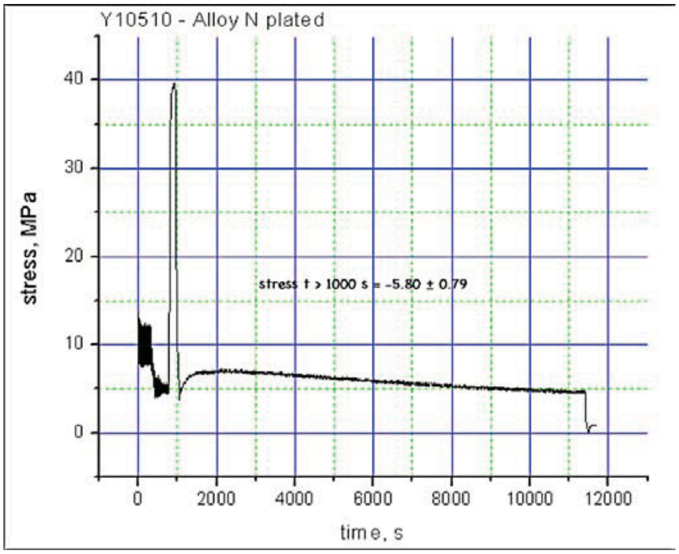

Fig. 5. Typical loading cycle for a diffusion weld under air ram control.

Parameter Development

Previous preliminary work with Alloy 617 suggested diffusion welding parameters of 3 hours at 1150 °C, at an applied pressure of 3-5 MPa.[8] This was used as a starting point for the present work.

Surface Preparation

Surface preparation turned out to be a very important component of the diffusion welding of these Ni-based alloys. Ni alloys in sheet form are usually provided with a pickled finish, whereas stainless steels are generally supplied with a brushed finish, perhaps because of the different intended uses (for example, stainless steels are commonly used for architectural, appearance, or food service functions). Fig. 6 shows some representative surface profiles for as-received and further processed Alloy 800H sheet material.

Early in the experimental work, surface profiles were not closely examined, but Fig. 6(a) indicates one reason this became a matter of some concern. There is a fairly uniform distribution of pits 50-100 µm in extent and as deep as 14 µm, considerably larger and deeper than might be expected to diffuse away under reasonable diffusion welding time, temperature and pressure, or than can be overcome by the application of plating or foil interlayers. As a result, initial tensile tests showed considerable plastic deformation but ultimate strengths only about 70% of base metal, a level that roughly corresponded to the observed unbonded area on the relatively planar fracture surface.

A number of surface preparation techniques were tried. The very smooth surface seen in Fig. 6(c) resulted from grinding in a metallographic environment, with a fixture to hold the 0.5 in. diameter material vertical, and a progression from a cut surface through 600 grit. Grinding a small area like this uniformly, where length is not critical (because it is compensated for by the Gleeble fixturing) is relatively easy; the processing of the large areas of sheet stock required for plate-type heat exchangers to a similar finish, however, is another matter. Uniformity of sheet thickness and similar treatment across a wide sheet area are important for component fitup and uniformity of bonding.

Within the laboratory, a number of methods were tried. Fig. 6(b) shows the result of using a double-acting sander and grits through 240, followed by brushing with an abrasive pad ("Scotch-brite"). The occasional deeper pit is still in evidence, although the overall relief has been reduced to random scratches with a relief of perhaps 1-2 µm. This involved a great deal of manual labor, and eventually an outside contractor was found who specializes in surface preparation for oilfield corrosion applications and was able to process relatively large sheets. In this case, a similar graded grinding approach was selected; other methods such as chemical treatment and electropolishing would not be out of the question, however.

Mechanical testing

The Gleeble diffusion welds normally used here are 6 inches long. One advantage of this geometry is that the weld strength can be tested economically by machining tensile specimens from these bars, with a gauge section as per ASTM E8 and a nominal diameter of 0.375 in. and straight grip sections. Results from a variety of Alloy 800H diffusion welds are shown in Fig. 6. Early welds made with bare surfaces (ground to 600 grit but not plated) were inconsistent, and some of the lower tensile strengths were a result of partial bonding across the face of the joint, perhaps aided by oxidation and the buildup of an oxide layer during the initial heat-up—some heat tinting was observed in these cases.

A nominal joint efficiency for code qualification is 90%, and it can be seen in Fig. 7 that, as long as plating or foil interlayers were used, times ranging from 1.5 to 7 h produced full strength joints in Alloy 800H. Reductions in area for these tests were from 50-70%. Results were similar for Alloy N material, with a nominal tensile strength (for sheet material) of about 115 ksi (793 MPa) and weld strengths in a narrow range averaging 103 ksi (710 MPa), or 89.5% of full strength.

Time/temperature Cycle Selection

The production of full strength joints at a variety of diffusion welding times suggests that, under proper conditions of loading and surface preparation, complete welding may occur in 1.5 h or even less. In many applications involving high temperature performance in various environments, particularly where some kind of interlayer is used, there is also a concern about compositional gradients and whether selective attack may occur if the gradients have not sufficiently leveled out. Similarly, since diffusion welding (particularly for high temperature alloys) is carried out at rather high temperatures (for example, 150 °C higher than for stainless steels), there is some concern about grain growth and the growth of second phases in the material that might be detrimental at service temperatures. These are both instances where numerical modeling (discussed below) can provide some insight into which time/temperature cycles are required, and which, while promoting rapid bonding, may be excessive for these reasons.

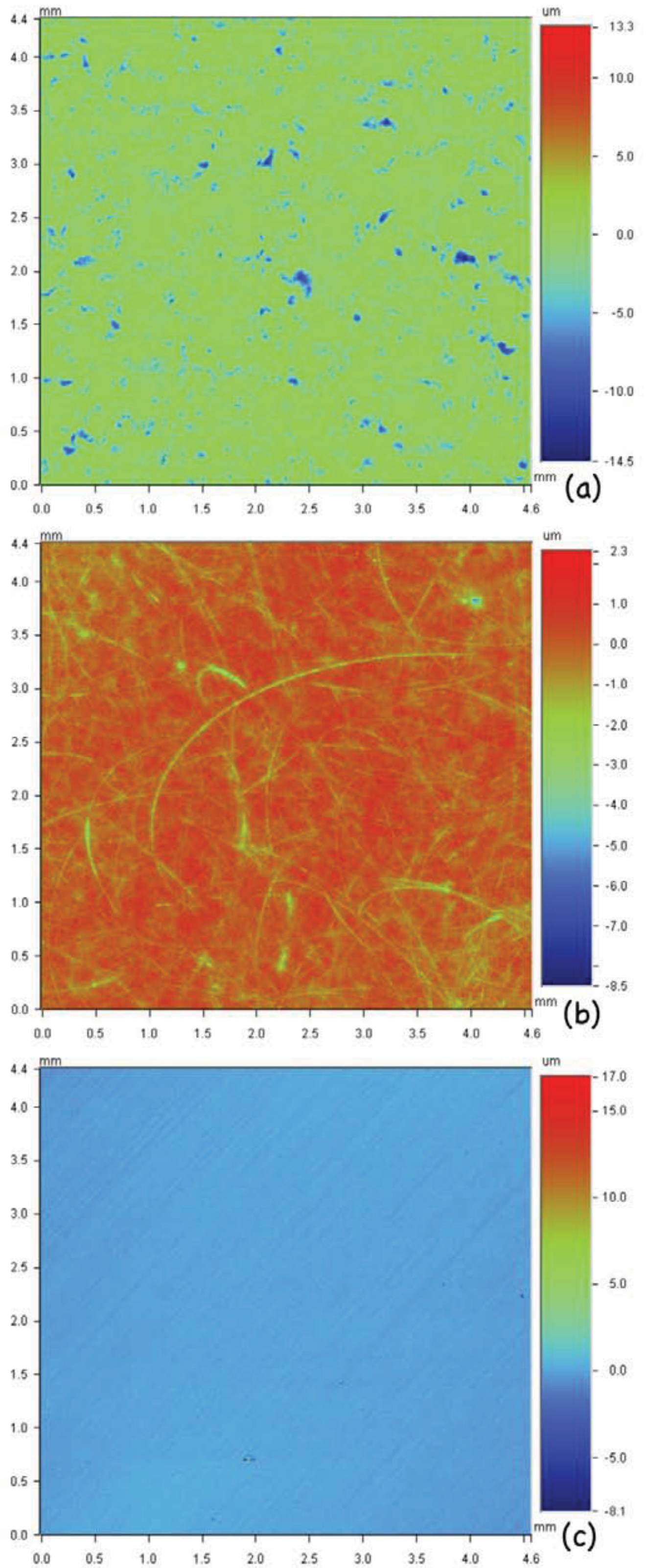

Fig. 6. Surface profilometry of: (a) as-received Alloy 800H sheet; (b) Alloy 800H after double-acting sanding through 240 grit and abrasive pad; (c) surface of base metal cylinder end after 600-grit metallographic grinding.

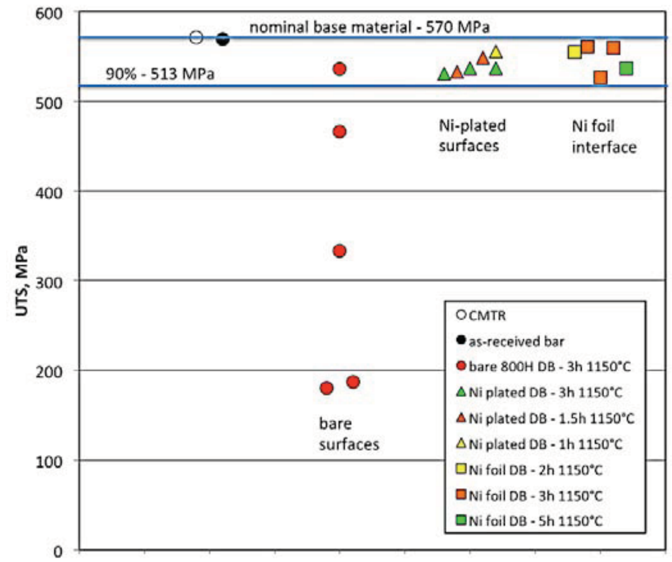

Fig. 7. Summary of ultimate tensile strengths for diffusion welds in Alloy 800H under various conditions.

Modeling

Modeling was performed with Thermocalc/DICTRA using the appropriate nickel databases.[4,5] For model verification purposes, it was convenient to use a joint design that involved a pure Ni interlayer, and to produce actual diffusion welds of the same geometry for comparison and verification. A typical arrangement was a 15 µm Ni foil at the center of a model mesh about 120 µm in length, which in actual use covered most of the diffusion effect around the joint for the standard conditions of 1150 °C and 1.5-7 hours. Because the joint design is planar, the model could be one-dimensional, and convergence times were short, typically well under an hour on PC-based computer hardware. Modeling details are covered elsewhere.[9]

No attempt was made to measure oxide layers, or to model their chemical potential and diffusion rates relative to the base metal; although this would be an interesting fundamental study to pursue, and potentially quite useful in designing diffusion welding parameters, it lay beyond the scope of the present work.

Microstructures

Fig. 8 shows a microstructure typical of diffusion welds in Alloy N. In this figure, EBSD (Electron Back Scatter Diffraction) indicates the crystallographic orientation of the grains, and is superimposed on the image of the microstructure obtained by SEM. The weld line, which originally contained a 15 µm Ni foil interlayer, can be seen as a slight rippling or wrinkling in the EBSD image, but, by and large, the microstructure of the base metal on either side is continuous across the joint. The particles visible in the image are a Mo-rich phase typical of Alloy N, and are quite large here because the base metal comes from a slab of hot-worked material about 5 x 20 inches (12 x 50 cm) in cross section. Fig. 9 shows the microstructure of the material after it has been reduced to 0.041 in. (1.04 mm) thick sheet. The particles are somewhat broken up and redistributed, but are still distinct.

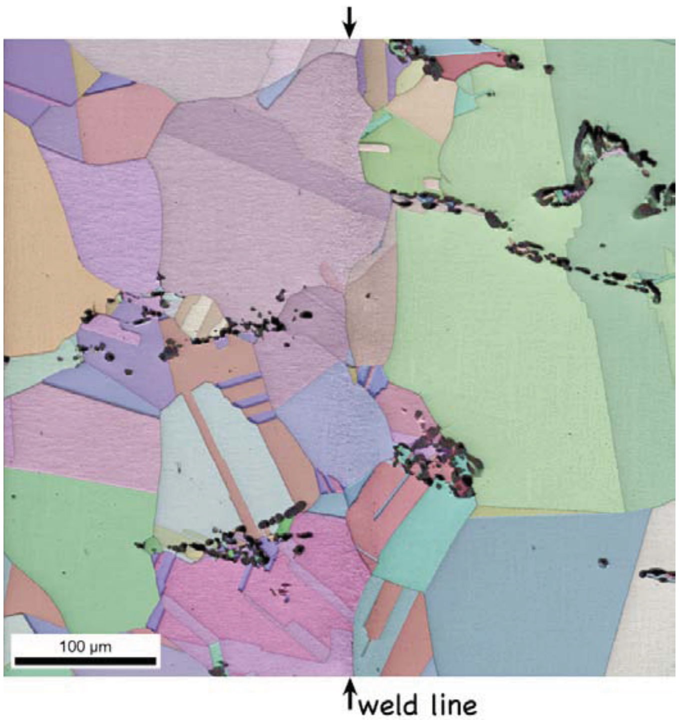

Fig. 8. Alloy N diffusion weld with superimposed EBSD data, demonstrating grain growth across weld interface. Weldline is the center of the original 15 µm Ni foil interlayer.

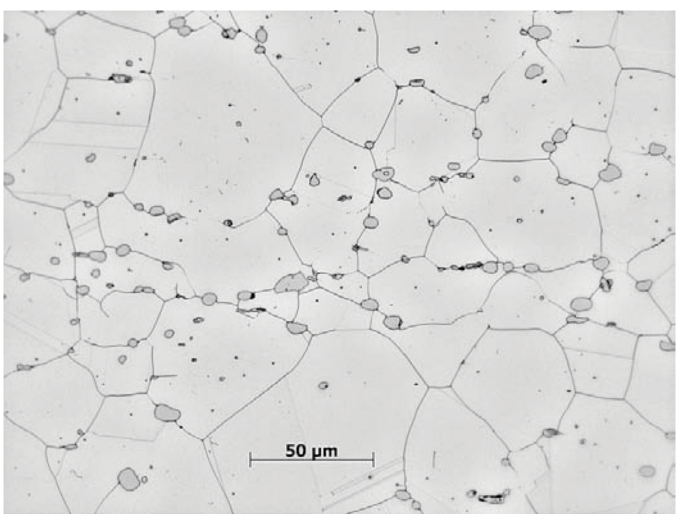

Fig. 9. Microstructure of Alloy N 0.041 in. sheet material.

Compositional Gradients

Fig. 10 shows a diffusion weld in Alloy 617, weld time 3 hours at 1150 °C with a 15 µm Ni interlayer, with X-ray analysis of the major elements superimposed, along with a stripe representing the original Ni foil. The burn marks of the analysis spots form which spectra were obtained are also visible. A feature of interest here is that both Al and Ti appear to be slightly higher in the original pure Ni foil area than in the base metal, as opposed to the other elements, which appear to indicate a simpler dilution effect. This may be due to the formation of Ni, Ti, and Al intermetallics, though the modeling did not indicate this for the thermal conditions used, and the noise of the spectral data at these low concentrations does not point unambiguously in this direction. Nonetheless, this is an interesting observation that may bear further investigation.

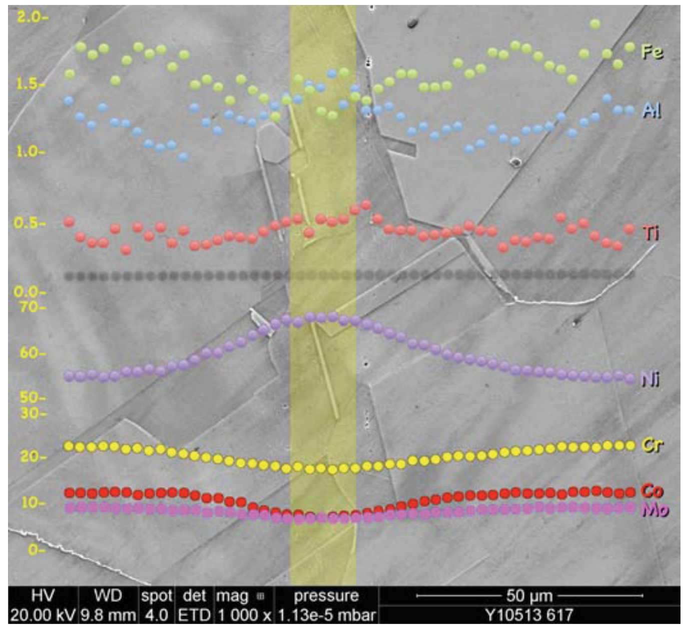

Fig. 10. Diffusion weld in Alloy 617 with 15 µm Ni foil interlayer (shaded vertical stripe shows foil's original location).

Model/Experiment Correlation

The model results were correlated with experimental observations for a number of diffusion welds; two examples are shown in Figures 11 (Alloy 617) and 12 (Alloy 800H). In most cases (Co in Alloy 617 is an exception), the experimental profiles of the major elements are somewhat flatter than the models predict, suggesting that the model tends to underestimate the diffusivity.

This remains unresolved at present. An unaccounted experimental factor is the time it takes a well-mated interface to develop (the elimination of asperities) and the inevitable oxide layers to break down (presuming they are a substantial barrier to diffusion). This, however, might be expected to delay diffusion, and to make the experimental profiles steeper than the modeled ones, which do not account for this initial effect. The basic purpose of the modeling, however, is to predict the composition profile across the joint for evaluating the susceptibility of the joint area (or of a joint area in dissimilar metals, where an asymmetrical composition gradient will exist) to environmental degradation in aggressive high temperature environments—for example, molten salts at 700 °C, or fast moving gas at up to 900 °C. Longer welding times, or post-weld heat treatments (at welding or other temperatures), might be expected to further flatten the gradients in similar metal welding, and these effects, as well as the development of minor second phases, can be evaluated by modeling, as well.

The 15 µm Ni foil was a convenient interlayer for validating the model, given the resolution of the SEM. Thinner foils (down to 5 µm) were also tried experimentally, and electroplated coatings of a combined thickness of 1-2 µm were also used in many welds after the inconsistent effects of

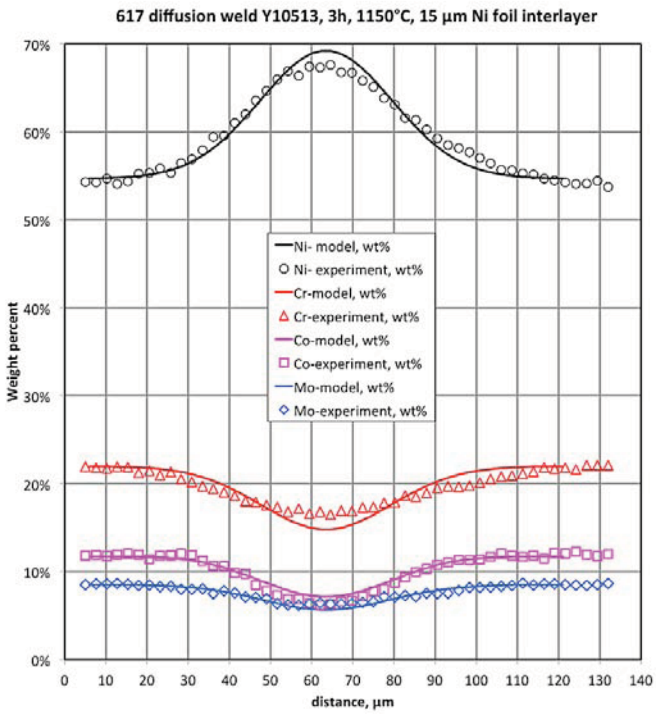

Fig. 11. Modeled and measured compositions for Alloy 617.

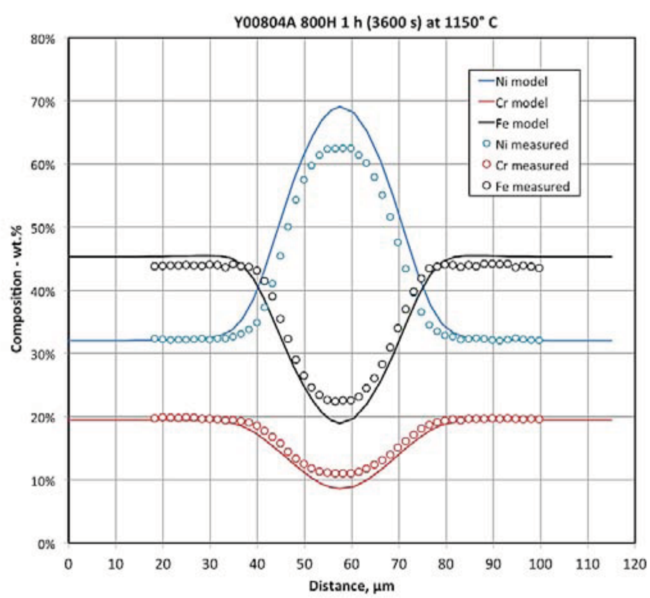

Fig. 12. Modeled and measured compositions for Alloy 800H after 1 hour.

welding bare metal (as in Fig. 7) were noted. Fig. 13 applies the model to a thickness of 1 µm of Ni. As might be expected, after three hours, such a thin Ni layer is almost entirely consumed by diffusion, and Fig. 13 is normalized to the original composition of each element, indicating that the Ni layer after welding is within 1% of the base metal composition.

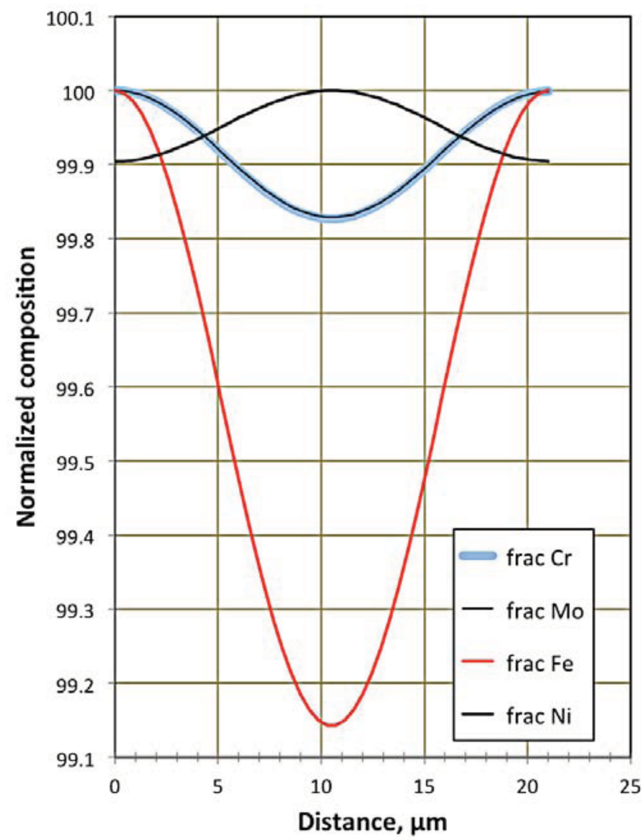

Fig. 13. Modeled compositions after 3 hours at 1150 °C, with Ni plated layers totaling 1 µm in thickness. Normalized to 100% of original concentration of each element in Alloy N.

Crystallographic Effects

The effects of crystallographic orientation most often appear in welding studies in connection with solidification, where fast and slow epitaxial growth directions with respect to isotherms have an influence on grain structure in fusion welds. A less common appearance is in very large-grained (or single-grain) materials, or strongly textured materials, such as magnetic alloys.

Fig. 14 shows a somewhat fortuitous indication of the influence of crystal orientation in the area of diffusion welding, in this case in Alloy 242. Like most of the other welds discussed here, this weld originally contained a 15 µm Ni foil, and after welding showed complete interpenetration of base metal grains across the interface. However, a quirk of the etching process (Br in methanol) brings out a pale line perhaps 20 µm from the original foil centerline, on both sides. It is unclear exactly what this is, although evidently it marks a critical composition for the etchant's interaction with the surface. It is also visible in optical micrographs and with Alloy N, although it is especially clear in this specimen.

The step changes in the position of this line when it crosses grain or twin boundaries is most easily explained by enhanced or retarded diffusion rates, due to changes in crystallographic orientation in these neighboring regions. The program effort did not allow for the detailed exploration of this phenomenon, for example, obtaining EBSD data to identify the absolute direction of the crystals, or fine scale (e.g., TEM or very high resolution SEM) examination of the compositional gradient within each grain. But it is an interesting side effect of the planar nature of these diffusion welded joints.

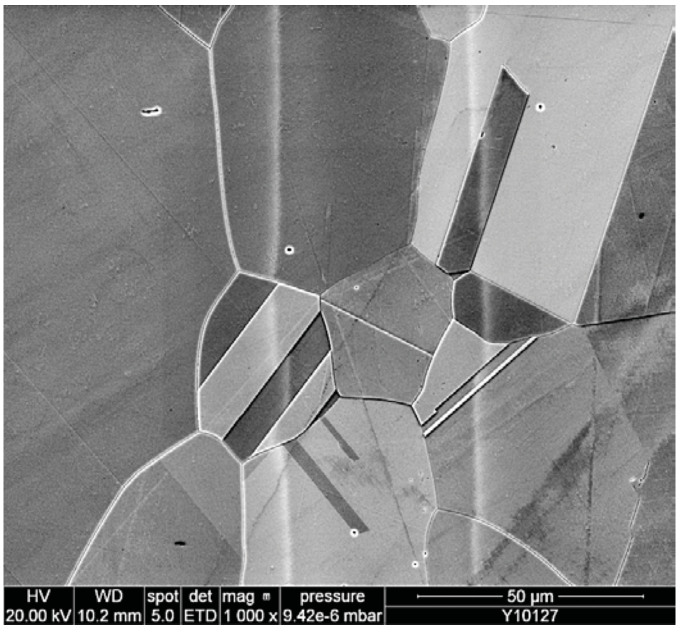

Fig. 14. Alloy 242 weld with 15 µm Ni foil, showing different diffusion rates according to crystal orientation.

Conclusions

The Gleeble is a useful tool for the development of diffusion welding parameters, including time, temperature, pressure, and the addition of filler metals or multiple layers of material, up to several layers thick, in dimensions up to about one inch in diameter and 6 in. long. Alloys 617, 800H, N, and 242 are readily welded with this technique, and exhibit full grain growth across the joint, provided the mating surfaces are carefully prepared. Ni electroplating or a Ni interlayer enhance diffusion weldability and composition gradients are substantially leveled under welding conditions of approximately 3 hours at 1150 °C and pressures of 3-7 MPa. Modeling with Thermocalc/DICTRA matched experimental composition gradients within a few percent, and, thus validated, are useful tools in designing diffusion welded assemblies and predicting their microstructural behavior during welding and in service. The influence of crystallographic orientation at the weld line was fortuitously indicated by some etching artifacts suggesting that diffusion along different crystal directions is measurably different.

Acknowledgments

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Appendix

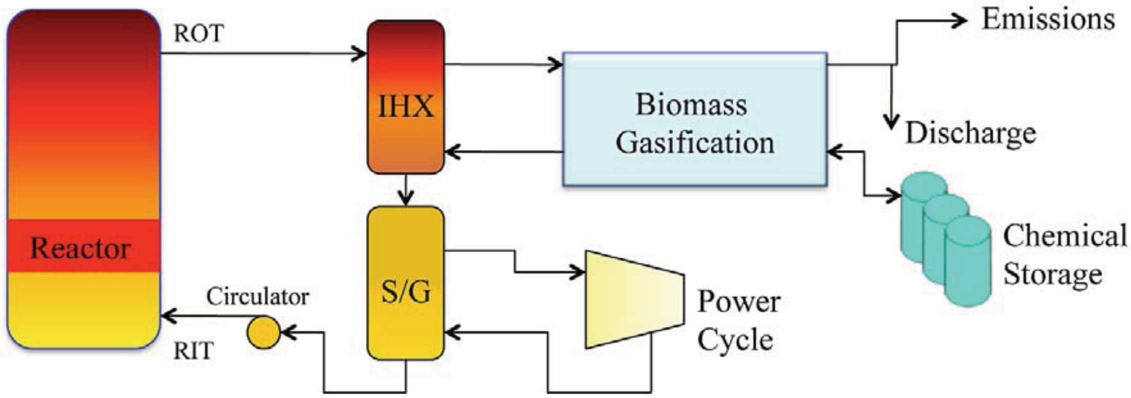

Fig. A-1. Schematic diagram of industrial process applications of nuclear heat. Compact heat exchangers are candidates for a number hf applications within such a system