Title	Joining of Advanced Materials by Combustion Synthesis
Author(s)	Liu, Weiping; Naka, Masaaki
Citation	Transactions of JWRI. 2000, 29(2), p. 1-10
Version Type	VoR
URL	https://doi.org/10.18910/10842
rights	
Note	

Joining of Advanced Materials by Combustion Synthesis[†]

Weiping LIU* and Masaaki NAKA**

Abstract

Novel joining processes are needed for the joining of similar and dissimilar combinations of advanced materials such as intermetallic alloys, structural ceramics, metal-matrix composites (MMC) and ceramic-matrix composites (CMC). Joining by combustion synthesis or self-propagating high-temperature synthesis (SHS), as a combination of SHS processing and joining technology, provides innovative joining capabilities for advanced materials. During the past decade, considerable research efforts have been directed to understanding the fundamentals of the novel joining process, joining technique developments for various materials, microstructural and mechanical characterization of the joints obtained, and process optimization and control. It is shown that SHS joining has been successfully utilized to join a variety of advanced materials. In this review article, the principle and characteristics of the joining process and the current development of the fabrication, microstructure and mechanical properties of joints and coatings made by this method were addressed. Future research directions are also indicated in this review.

KEY WORDS: (Combustion synthesis) (SHS) (Joining) (Intermetallics) (Refractory materials) (Ceramics) (Composites)

1. Introduction

With the development and practical applications of advanced materials such as intermetallic alloys, structural ceramics, metal-matrix composites (MMC), intermetallic-matrix composites (IMMC) and ceramic-matrix composites (CMC), joining of these new or advanced materials in similar and dissimilar combinations is becoming increasingly important as an indispensable means of fabricating engineering materials into structural components[1)]. Intermetallics like aluminides Ni_3Al, NiAl, Ti_3Al, TiAl, Fe_3Al and FeAl are considered to be attractive materials for high temperature structural applications because of their high strength retention at elevated temperatures, combined with relatively low density, good oxidation and corrosion resistance. For example, the elevated temperature strength and creep resistance of recently developed Ni_3Al alloys are shown to be superior to most of the commercial superalloys[2)]. Unfortunately, the weldability of these intermetallic alloys by conventional fusion welding processes is generally very limited because of their high susceptibility to hot cracking both in the heat-affected zone and in the fusion zone[3-6)]. Welding of intermetallic-matrix composite materials presents still greater challenges. The problems include the disruption of desirable arrangements of the reinforcements and their decomposition during welding followed by reprecipitation of embrittling interfacial films[7)]. In addition, there is a serious need to develop suitable methods to join structural ceramics to ceramics, to intermetallics or to conventional heat-resistant alloys for the fabrication of hybrid structures. Although a variety of brazing techniques are widely used for joining ceramics to ceramics or to metals, the high temperature performance of these brazed joints with metallic or glassy fillers has been found to be unsatisfactory[8)]. Brazing filler alloys for elevated temperature use are still to be developed. Despite its ability to produce refractory ceramic-metal joints for high temperature applications, solid state diffusion bonding requires complicated equipment and an extremely good surface finish for the parts to be joined, which is difficult and costly to obtain for ceramics. Therefore, developing new joining processes would be necessary.

Combustion synthesis or self-propagating high-temperature synthesis (SHS) has been a method for the preparation of materials by use of highly exothermic

---

[†] Received on Dec. 18, 2000

* Visiting Professor from Dalian Railway Institute, Dalian, China
** Professor

Transactions of JWRI is published by Joining and Welding Research Institute of Osaka University, Ibaraki, Osaka 567-0047, Japan.

reactions between powdered constituents, which was developed and first studied extensively by Merzhanov and co-workers[9]. It provides an attractive alternative to conventional methods of producing advanced materials. A large number of materials[10-13] have been synthesized by this method, including ceramics, ceramic composites, intermetallic compounds and their composites. These materials are used in applications that involve high temperatures and/or require high wear resistance. Besides synthesizing raw materials, this process has also been used for fabrication of near-net-shape or net-shape parts through simultaneous consolidation with the material synthesis[14-15]. More recently, this novel process has been increasingly utilized in combination with other traditional fabrication technologies such as sintering, casting, thermal extrusion forming, rolling, welding and surfacing. Combined with joining technology, the combustion synthesis process can be used for joining materials[16-35], leading to a novel joining process termed as SHS joining or pressurized combustion synthesis (PCS) joining when pressure is applied during the joining process[19,27].

The PCS joining process is particularly attractive for joining the difficult-to-weld intermetallics, ceramics and their composites[22]. This novel process has a number of potential advantages over the conventional processes. It is an energy-saving process with limited thermal effects in heat-sensitive substrates due to rapid and highly localized heating. It can be used for near-net-shape joint production with simultaneous bulk materials synthesis and in-situ joining. The joining process offers the capability of forming a composite joint interlayer for composite joining by incorporating various reinforcing particles, fibers and whiskers into the reactants or through in-situ synthesis of reinforcing phases. Using this process, it is also possible to fabricate functionally gradient material (FGM) joints for overcoming mismatches in chemical composition, physical and mechanical properties between dissimilar materials[19].

In this paper, the principle of the PCS joining and the latest developments in joining advanced materials by combustion synthesis are reviewed. Our most-recent investigations in this specific area are summarized.

2. Principle and Characteristics of the PCS Joining Process

There are two basic modes of combustion synthesis, i.e. the propagating mode and the thermal explosion (or combustion) mode[9]. In the first mode, the reaction is initiated at some point in the powder compact by a localized heat pulse, and the combustion wave propagates with a definite velocity through the whole compact. In the thermal explosion mode, however, the compact as a whole is heated uniformly by some means to above the ignition temperature (T_i), and the reaction occurs simultaneously throughout the entire compact. In both cases, the product of combustion reactions is the material to be obtained. The products obtained in this way are normally very porous. However, dense products of combustion synthesis can be obtained with the simultaneous application of external pressure by various densification techniques such as hot pressing, HIPing and use of shock waves during combustion reaction.

The combustion synthesis joining or SHS joining is a combination of SHS processing with joining technology. It utilizes exothermic combustion synthesis reactions to produce the joining or filler material and achieve bonding in-situ to the parts to be joined. In this novel joining process, the pre-compacted powder mixture of reactants, in appropriate atomic proportions, is placed between the two workpieces to be joined. The heat generated by the exothermic reactions between the powdered reactants together with externally applied heat is used, both for the synthesis in the joint filler, and for the bonding at the joint interface. The synthesized product of combustion reaction acts as the joint interlayer or filler material, and is in-situ bonded to the parent material. In an alternative method, joining of materials can be achieved simultaneously with their synthesis and consolidation in a process called "primary joining", as opposed to "secondary joining" termed for joining of two preexisting materials[22]. Since pressure is usually applied during the SHS joining process in order to produce dense joints, the process is also referred to as pressurized combustion synthesis (PCS) joining.

3. Methods and Apparatuses Used for PCS Joining

Several methods have been used so far for investigations of combustion synthesis joining. Messler and coworkers[19-21] uniquely employed a Gleeble thermal-mechanical simulation apparatus for SHS joining. They used a high-strength graphite cylindrical tube to contain the joint elements and the powdered reactant mixture disc which was sandwiched between the joint elements. Liu and coworkers[27-32] also utilized this type of apparatus in their studies, but without the use of any containment tubes. The Gleeble tester proved to be an excellent apparatus for studying combustion synthesis joining. This tester is able to apply controlled and pre-programmed temperature-time as well as pressure-time cycles. Heating is accomplished by using the electrical resistance of a current through the joint sample. Pressure is applied on the sample through a servo-hydraulic loading system. The arrangement of a joint sample on the Gleeble tester is

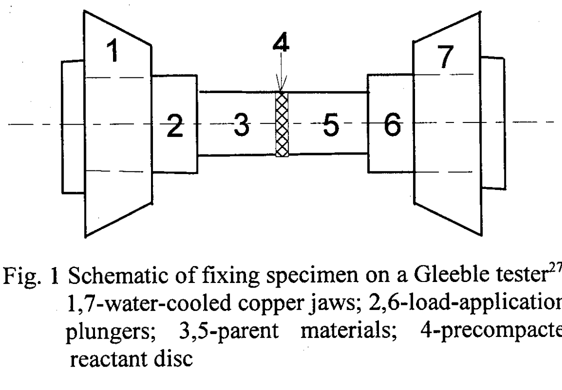

Fig. 1 Schematic of fixing specimen on a Gleeble tester[27]. 1,7-water-cooled copper jaws; 2,6-load-application plungers; 3,5-parent materials; 4-precompacted reactant disc

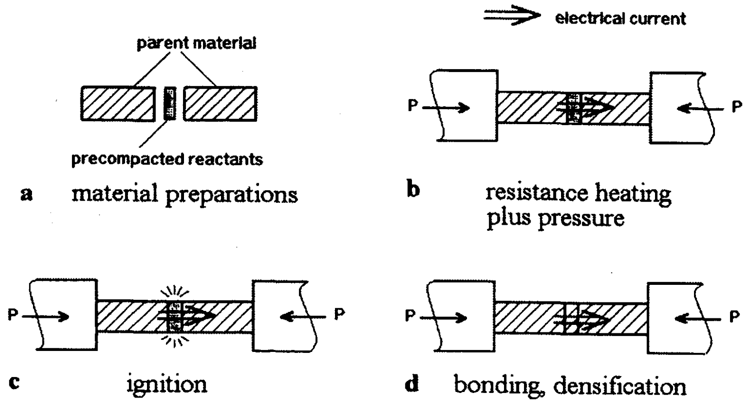

Fig. 2 Schematic of the pressurized combustion synthesis joining process accomplished in the thermal explosion mode by means of internal electrical resistance heating

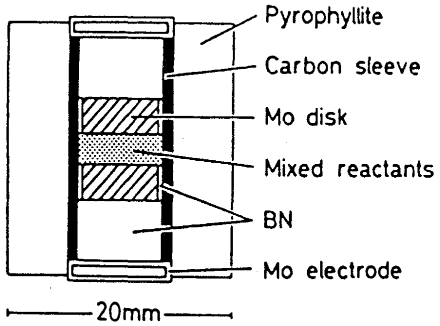

Fig. 3 High-pressure cell assemblage used for pressurized combustion welding[16].

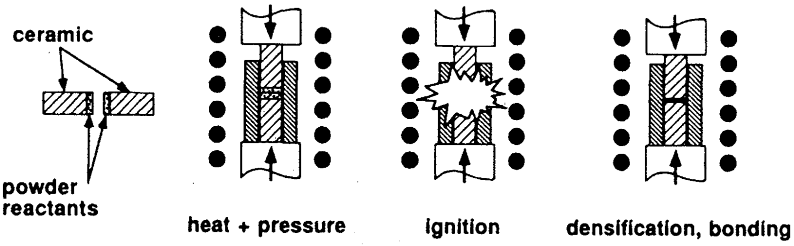

Fig. 4 Schematic of the reaction joining technique in which a hot press is used and the joining reactants made on the surface of parent material by tape casting[18].

schematically shown in **Fig. 1**. The hot junction of a thermocouple recording the sample temperature was embedded in the pre-compacted reactant disc[20] or was located at the sample surface immediately adjacent to the compacted reactant disc[27]. **Fig. 2** schematically shows this type of PCS joining process accomplished in the thermal explosion mode by means of internal electrical resistance heating. Shcherbakov and Shteinberg[25] have used a similar experimental setup for SHS welding of refractory materials in the electrothermal explosion (ETE) mode. The ETE mode can also be accomplished through induction heating by exposing the joint sample with the reactant mixture to a high-frequency electromagnetic field.

Miyamoto and coworkers[16] developed the high-pressure self-combustion sintering (HPCS) technique using a similar SHS-HIP (hot isostatic press) approach to fabricate dense TiB_2 and TiC ceramics, and utilized this method to join the synthesized ceramics to refractory metal Mo. The processing involved inserting the reactant compact sandwiched between the materials to be joined into a pyrophyllite cube cell and subjecting this cell to a pressure of 3GPa by means of a cubic anvil device. **Fig. 3** shows the high-pressure reaction cell used for joining in their research. An electrical insulating BN sleeve was placed between the Mo disks and a cylindrical carbon heater for ignition so that only the reactant compact was made contact with the heater. The reactant was ignited at the periphery through the carbon heater. In this way, the combustion synthesis joining was conducted in a self-propagating mode. In a study of joining the SiC ceramic and SiC/SiC composites using combustion reactions, Rabin[18] used a hot press for carrying out the reaction joining experiments. The method he used is shown schematically in **Fig. 4**. In a different way, the powder reactants were fabricated into thin layers directly on the surface of the materials to be joined using a modified tape-casting procedure. In this process, the powders were mixed with a solvent, deflocculent, organic binder and plasticizer to form slurry. After the slurry was milled, an adjustable doctor blade was used to cast a thin layer of the slurry on the sample surface, which dried to form the green tape. The tapes contained about 30-40 vol.% organics that would be removed by thermal decomposition prior to joining[18].

Instead of using powdered reactant mixtures, Hawk and Alman et al.[44-45] utilized the combustion reactions at the interface between dissimilar elemental metal foils to synthesize an intermetallic layer and to join these foils simultaneously as a technique for the fabrication of in-situ layered metal-intermetallic composites. **Fig. 5** is a schematic representation of this method.

4. Joining of Various Materials

4.1 Joining of intermetallics

Messler et al.[20-21] systematically studied the role and effects of process parameters such as processing temperature, hold time, applied pressure, and heating rate in SHS joining. They used Ni and Al powder mixtures to synthesize the Ni_3Al intermetallic compound fillers and to join the Inconel 600 nickel-base alloy end elements. They found that the processing temperature had the greatest influence on bond integrity, reaction product

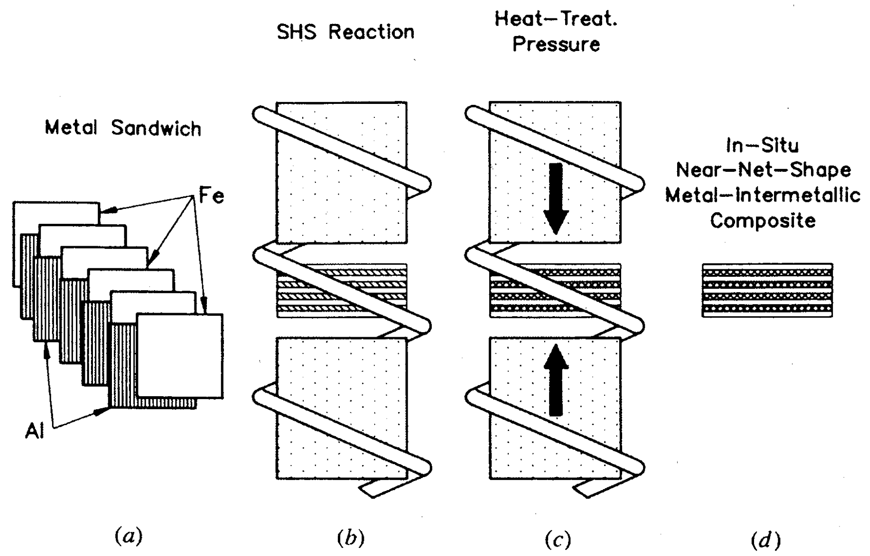

Fig. 5 Schematic representation of SHS joining used to form metal-intermetallic composites: (a) elemental foils are stacked into appropriate sequence; (b) foils are placed in vacuum hot press and heated to initiate the SHS reaction; (c) after SHS reaction is completed, pressure is applied to densify composite; and (d) resultant structure is a metal-intermetallic layered composite[45].

homogeneity and joint density. Joint densification was enhanced by increasing the pressure applied during processing. In their experiments, however, the heating rate in the range from 0.5 to 5°C/s was not found to have an effect. Wright et al.[24] have investigated joining of solid Fe_3Al hot-extruded material using combustion reactions of compacted Fe-28at.%Al and Fe-50at.%Al powder mixtures. In their experiments, the sandwiched samples were heated in the hot press of a graphite die at a rate of 0.3°C/s to 1200°C under an applied pressure of 24.5 MPa, and held at the pressure and temperature for 15 min in an argon atmosphere. The joints produced with nominal applied pressure were found to have a high level of porosity, while the hot-pressed Fe_3Al joints were near theoretical density. They also found that addition of 2% Cr in the Fe_3Al joints was effective to increase the ductility of the joints. In a study by Uenishi et al.[26], the intermetallic compound TiAl (Ti-34 mass% Al) was joined in a hot press by the SHS reaction of blended elemental Ti and Al powder mixture of the same composition used as a filler metal. The microstructure of SHS reacted filler metal was found to be inhomogeneous after joining at a temperature up to 1173 K for 1 hr and consisted of $TiAl_3$, α_2 (Ti_3Al) and Ti. After a post-joining heat treatment at 1573 K for 3 hrs, the microstructure of the filler metal was converted to a fine lamellar structure consisting of α_2 and γ(TiAl) phases, the same constituent phases as those of the base material. The SHS joined samples had a tensile strength of about 220 MPa at room temperature and at 873 K, which was about the same as that of the base material.

In the studies by Liu and coworkers[27-28], they found that both the heating rate and joining temperature had the most remarkable effects on the completeness of synthesis reaction and hence on the microstructural homogeneity in the filler material. In their experiments to join Ni_3Al intermetallic cast alloys by combustion synthesizing Ni_3Al fillers using the Gleeble tester, a joining temperature higher than 700°C appeared to be necessary for bond formation. A higher heating rate was found to be beneficial for accelerating the reactive synthesis process by decreasing the formation of diffusional pre-combustion phases and elevating the real combustion temperature in the joint filler due to reduced heat losses. Completely synthesized Ni_3Al was obtained in the joint seam at joining temperature of 1100°C for 30 min using a high heating rate. **Fig. 6** shows the microstructure of a Ni_3Al joint made at 1100°C for 60 min and an applied pressure of 35 MPa with a heating rate of 10°C/s. Dense and essentially single-phase Ni_3Al with fine equiaxed grains was formed in the joint seam. Microhardness of the fully-synthesized Ni_3Al was measured to be in the range of 350-380 HV. The Ni_3Al alloy joints with synthesized boron-doped Ni_3Al filler exhibited a tensile strength of 541 MPa and an elongation of 5.6% at room temperature[32]. Dissimilar joints were also successfully made by PCS, as demonstrated in **Fig. 7** which shows the microstructure of a joint between IC-221M (Ni_3Al alloy) and nickel-base superalloy. The researchers also

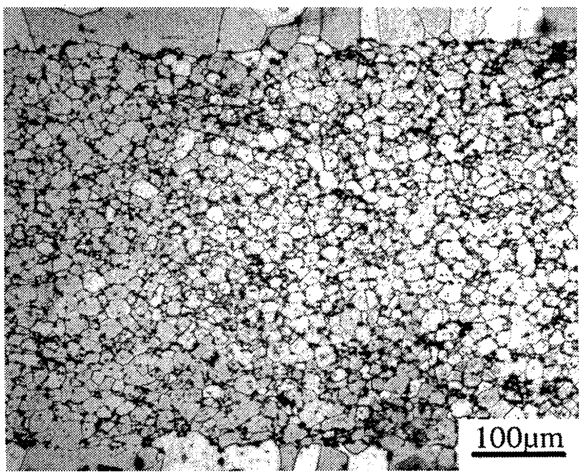

Fig. 6 Microstructure of a Ni_3Al joint made at 1100°C/60 min/35 MPa, 10°C/s heating rate.

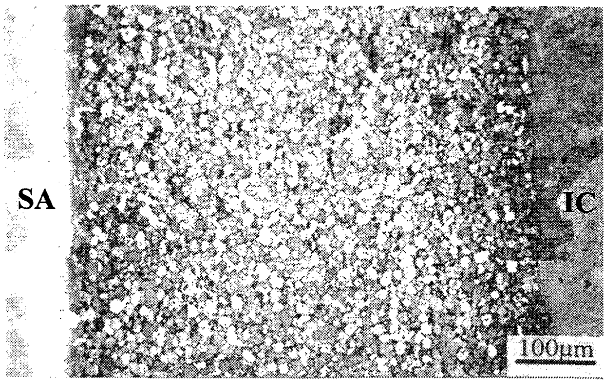

Fig. 7 Microstructure of a dissimilar joint between IC-221M (IC) and nickel-based superalloy (SA) with a synthesized Ni_3Al joint interlayer made at 1100°C/60 min/35 MPa.

investigated PCS joining through synthesizing nickel monoaluminide NiAl as the filler material. However, the joints with reaction synthesized NiAl interlayer had a lower strength of 198 MPa at the room temperature due to brittleness of the intermetallic compound. The joints failed by cleavage fracture in the NiAl interlayer[30)].

4.2 Joining of refractory materials

A variety of refractory materials such as graphite, tungsten, molybdenum, superalloys have been joined in similar and dissimilar combinations by SHS. **Table 1** lists the experimental conditions and strength data for different joints of refractory materials made by SHS using the electrothermal explosion (ETE) method from an investigation by Shcherbakov and Shteinberg[25)]. They used reactant mixtures prepared from Ti, Mo, Nb, Zr, C and B powders respectively to synthesize the carbides and borides for joining the refractory materials. The strength of graphite joints exceeded that of the base material while other joints failed in the weld seam.

As an example of practical applications of SHS joining, Shcherbakov and Shteinberg utilized this technique for the production of cathodes of powerful high-frequency generating tubes, which requires joining of tungsten and molybdenum parts[25)]. It was effected by reacting a mixture of molybdenum and boron powders to form the molybdenum boride acting as a hard braze. The welding time was only 5 seconds, and therefore no recrystallization of tungsten and molybdenum occurred. The cathodes produced by this method had better strength and better emissive properties than those made by conventional platinum brazing in a hydrogen atmosphere. The SHS joining technique had also the economic advantage over the conventional one because the reactant mixture contained no rare-earth or precious metals.

4.3 Fabrication of composite joints

Rabin used the combustion reactions in the Ti-C-Ni system for joining of silicon carbide fiber-reinforced silicon carbide composites[17-18)]. For this material, exposure to processing temperatures above 1200°C during joining must be minimized since strength reduction results from prolonged exposure to such temperatures due to degradation of the SiC fibers. In his experiments, the researcher prepared the joining mixture from Ti, C, and Ni powders with Ni content in the range of 5 ~ 15 wt%. Ni was added both to lower the ignition temperature of the system and to lower the combustion temperature. Additionally, Ni also enhanced densification by the action of capillary forces through formation of a liquid phase. The resulting joint interlayer consisted of TiC-Ni cermet structure. Room-temperature four-point bending strengths of the composite joints were averaged to be about 100 MPa.

Liu and coworkers[31-32)] investigated the fabrication of Ni_3Al matrix composite joints by pressurized combustion synthesis. They incorporated a certain amount of ceramic particles (Al_2O_3, TiC) into a mixture of Ni and Al powders and utilized the combustion reactions to produce a ceramic-particle-reinforced Ni_3Al-matrix composite interlayer material and to achieve bonding to the parent material simultaneously. The experimental results showed that a higher joining temperature and/or a longer hold time were needed to ensure a completely reacted Ni_3Al-matrix composite joint seam and to achieve good bonding with the parent

Table 1. Experiment conditions of SHS welding and joint strengths[25)]

SHS-welded materials	Composition of mixture (mass%)	Specific density of initial mixture	Current of welding (A/cm^2)	Material of welding seam	Joint strength (MPa)	Place of fracture
W-Mo	Mo-80, B-20	0.77	600	Mo_2B_5	180~200	Seam
W-Mo	Mo-64, B-16, Cu-20	0.85	850	Mo_2B_5-Cu	270~320	Seam
Graphite-graphite	Ti-86, C-14	0.6	900	7TiC-3Ti	70	Graphite
W-graphite	Ti-86, C-14	0.7	1000	7TiC-3Ti	60	Graphite
Mo-graphite	Ti-86, C-14	0.75	1100	7TiC-3Ti	70	Graphite
Nb-10X18H10T	Nb-70.9, Ni-20, C-9.1	0.8	1500	NbC-Ni	130~150	Seam
Zr-10X18H10T	Zr-79.6, C-10.4, Ni-10	0.8	1200	ZrC-Ni	90~110	Seam

material as the proportion of Al_2O_3 particles in the reactant mixture increased. It was believed that the inert ceramic particles acted as heat sinks and barriers impeding the flow of the Al-rich liquid and the diffusion of Ni and Al atoms during the reactive synthesis. The non-reacting ceramic particles also adversely affected bond formation when coming into contact with the parent material during the PCS joining process. By using appropriate joining process variables, sound joints with fully reacted Ni_3Al-matrix composite interlayers were obtained in all cases with 0–20wt% Al_2O_3 reinforcements.

Tables 2 and **3** respectively show the microhardness measurement results for the composite interlayer and the tensile test results of PCS joints at room temperature, reported in the study by Liu and coworkers[32]. The hardness of the joint seam increased with increasing proportion of the ceramic particles in the reactant mixture. Tensile strength was increased by the addition of 3wt% Al_2O_3 particles in the joint seam while further increases in the amount of particles led to decreased joint strengths. SEM fractography revealed that tensile failure of the joints occurred in the joint seam as a result of matrix-particle debonding, as shown in **Fig. 8**. The authors suggested that strength reduction in case of higher particle amounts was related to agglomeration of the reinforcements in the composite seam and to the poor bonding observed between the Al_2O_3 reinforcement and the Ni_3Al matrix. Improved mechanical properties for the composite joints would be expected with improvements in particle dispersion during mixing and by optimization of processing variables.

Table 2. Microhardness measurement results of the joint seams[32]

Composition of joint seam	Microhardness range, VHN
Ni_3Al	350~387
Ni_3Al+5wt% Al_2O_3	427~476
Ni_3Al+10wt% Al_2O_3	431~492
Ni_3Al+15wt% Al_2O_3	480~751
Ni_3Al+20wt% Al_2O_3	492~811

Table 3. Average tensile properties of the composite joints[32]

Composition of joint seam	UTS, MPa	Elongation, %
Ni_3Al	541.8	5.6
Ni_3Al+3wt% Al_2O_3	586.6	4.8
Ni_3Al+5wt% Al_2O_3	521.0	4.3

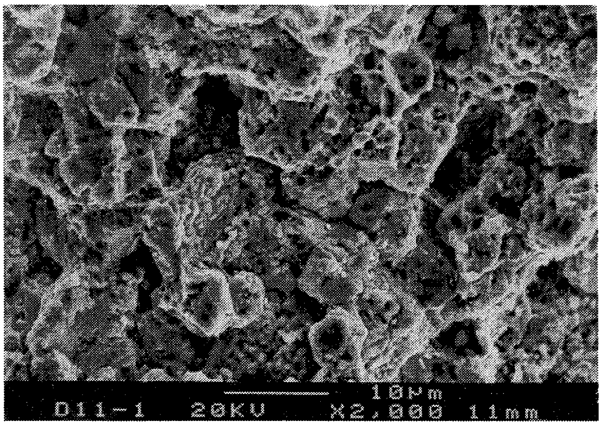

Fig. 8 SEM fractograph of the composite joint seam with 5wt% Al_2O_3 reinforcements.

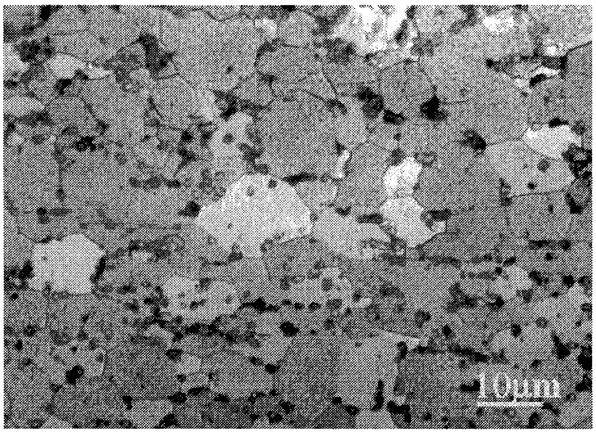

Fig. 9 Microstructure of a joint seam synthesized from a blended Ni and Al powder mixture, showing fully-synthesized Ni_3Al grains with a small amount of Al_2O_3 particles.

It was noted that, even without addition of any ceramic reinforcements, a small amount of oxide particles was formed in the combustion-synthesized joint seam[32]. **Fig. 9** shows the microstructure of a joint seam synthesized from a blended Ni and Al powder mixture. It consisted of fully-synthesized dense Ni_3Al grains with a small amount of Al_2O_3 particles existing mainly along the grain boundaries and sometimes inside grains as well. The volume fraction of Al_2O_3 particles was measured to be about 2.3% by determining the area fraction. These Al_2O_3 particles were considered to be formed in-situ as a result of reduction of surface oxides of the powders and by additional internal oxidation of Al particles during the reactive synthesis joining process. This phenomenon was utilized for fabrication of intermetallic-matrix composites through reactive hot compaction (RHC) of pre-oxidized starting elemental powders[46-47].

A composite joint seam can also be formed through in-situ synthesis of ceramic reinforcements and the matrix during combustion synthesis joining. In-situ composites are generally characterized by fine reinforcing particles, which are uniformly distributed in the matrix, and by a clean particle/matrix interface. In a study by Liu et al.[33], elemental powders of Ni, Al, Ti, C were used to prepare the reactant mixture as the filler material for fabrication of joints with NiAl-TiC (25wt% TiC) composite seams. High-energy ball milling of the elemental powder mixture was conducted before joining, which was found to greatly decrease the ignition temperature of the combustion synthesis reactions for the Ni-Al-Ti-C system during subsequent heating for joining. The composite joints were produced at a joining temperature of 1100°C for 60 min.

As mentioned before, the combustion synthesis joining is a promising method for fabrication of FGM joints between dissimilar materials. Zhu[35] conducted a feasibility study for formation of a FGM-type joint seam. In his experiment, pre-compacted thin layers of different reactant-powder compositions in the required stoichiometry were prepared from elemental powders of Ni, Al, Ti, C. These thin layers were stacked manually and pressed into a multi-layer powder compact, which was then sandwiched between nickel-based superalloy parent materials. Although joints with NiAl-TiC FGM-type interlayer of varying TiC contents from 0 to 50wt% were produced, they exhibited a considerable amount of porosity in the portion of the joint with high contents of TiC. Kudesia et al.[36] studied the fabrication of a TiC-NiAl FGM joint between a ceramic (TiC) and an intermetallic compound (NiAl). In their study, both the FGM joint and the materials to be joined were produced by combustion reactions. The researchers did not present much of their experimental results but a theoretical analysis of this approach was provided for designing the fabrication process in this reference.

In a different way, Hawk et al.[44] and Alman et al.[45] applied combustion synthesis joining to metal foils and produced different in-situ metal-metal aluminide layered composites. In their experiments, layers of thin Al foil (e.g. 0.15 mm thick) were alternately stacked between layers of thin metal (Ni, Ti, or Fe) foil, and SHS reactions were initiated at the interfaces between the dissimilar elemental metal foils under low pressure and followed by post-SHS aging at 1100 K for 1 hr under pressure (27.5 MPa). Fully dense, well-bonded metal-intermetallic layered composites were fabricated using this technique. By altering the thicknesses of the starting elemental foils, the composites could be designed to possess high-strength and high-toughness properties. This processing technique was reported as an economical method for production of light-weight and high-performance layered composites.

4.4 Joining of ceramics

Miyamoto et al. utilized a pressurized combustion reaction of the powdered mixture of Ti and B or C for ceramic-to-metal welding[16]. They obtained the Mo-TiB_2-Mo and Mo-TiC-Mo joints with tensile strengths of 20-40 MPa and 10 MPa respectively. The synthesized TiB_2 ceramic was dense with a mean grain size of 5 μm, while a porous TiC with grain size of 15 μm was obtained. Wide reaction regions (80~170 μm) were observed at the ceramic/metal interfaces, with the major reaction products being Mo_2B and Mo_2C respectively. Rabin joined dense SiC using a Ti + C + 10 wt% Ni mixture, and obtained an average bending strength of 98 MPa with a maximum value of 128 MPa for the joints. Failure of the specimens was observed to initiate at the interface between the joint interlayer and the SiC ceramic, and crack propagation occurred partly within the joint interlayer and partly along the interface.

Joining of Al_2O_3 ceramic was studied by Liu et al.[34] using the exothermic synthesis reactions in the Ni-Al-Ti system. **Fig. 10** shows a micrograph of an Al_2O_3 (purity of 99 wt%) ceramic joint bonded with reaction-synthesized Ni_2AlTi interlayer at 1350°C for 2 hrs and an applied pressure of 35 MPa. In this case, two pieces of the ceramic were joined using a pre-compacted powder mixture composed of 2Ni + Al + Ti (in atomic proportions). A 400 μm thick dense joint interlayer was obtained. Good bond was achieved in spite of the 2~3 μm surface roughness of the ceramic. No distinguishable reaction product was found at the interface between the ceramic and the synthesized interlayer. They also produced ceramic joints with a composite interlayer. **Fig. 11** shows a SEM micrograph of the Al_2O_3 ceramic

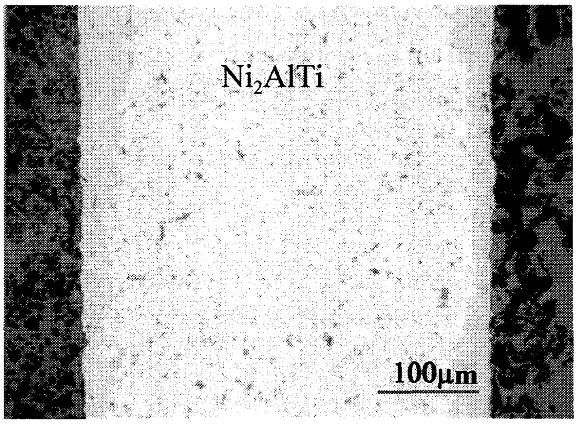

Fig. 10 Micrograph of an Al_2O_3 ceramic joint bonded with a reaction-synthesized Ni_2AlTi interlayer at 1350°C/120 min/ 35 MPa.

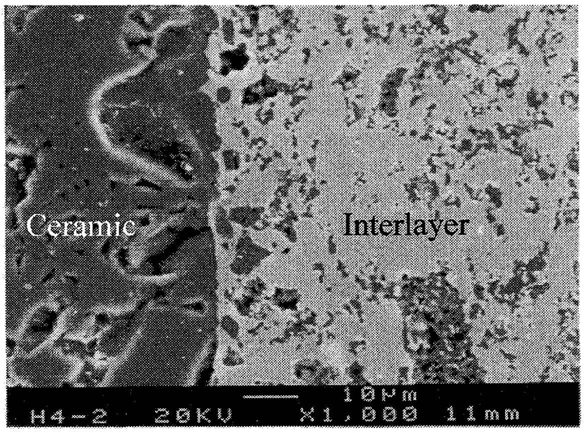

Fig. 11 SEM micrograph of Al_2O_3 ceramic bonded with a reaction-synthesized $Ni_3Al(Ti)$-Al_2O_3 composite interlayer at 1250°C/120 min/35 MPa.

bonded with a reaction-synthesized $Ni_3Al(Ti)$-Al_2O_3 composite interlayer at 1250°C for 2 hrs and an applied pressure of 35 MPa. In this case, two pieces of the ceramic were joined using a compacted reactant mixture composed of 90wt% (3Ni+Al) + 5wt% Ti + 5wt% Al_2O_3 (particles). Ti was added to improve the bonding between Ni_3Al and the alumina (the parent material and the particles as well). In addition to strengthening effects, a composite joint interlayer was also considered to be beneficial for decreasing residual thermal-stresses of the joint because of a reduced difference in coefficients of thermal expansion[48)]. No reaction layer was observed at the joint interface. However, concentration profiles of elements measured by EDX indicated somewhat higher concentrations of Ti existing at the Al_2O_3-particle/$Ni_3Al(Ti)$-matrix interface[34)].

4.5 Coating and cladding by SHS

SHS joining can also be applied in the fabrication of coatings and claddings on substrate materials. Such coatings and claddings can significantly improve chemical, physical and mechanical properties of the surfaces. There are many configurations of materials that can be used to form a coating on a substrate by SHS. All the material systems described above for joining two parent materials can also be applied for this purpose, e.g. intermetallic compounds, ceramics and composites etc.. Wright et al.[24)] studied the fabrication of a Fe_3Al coating on a carbon steel substrate using combustion synthesis reactions. A layer of blended Fe and Al powder mixture of the Fe-28at% Al composition was pressed onto a substrate with a rough surface texture in order to achieve improved bonding. The resulting compact was heated in a graphite die to 1000°C under an applied pressure of 28 MPa and held at this temperature for 1 hr. A well-bonded Fe_3Al coating was formed with a fine grain structure of about 5 μm. Interdiffusion of elements between the coating and the steel substrate occurred during the reaction processing, which was considered to beneficial for good bonding.

Uenishi et al.[37-38)] investigated the formation of an Al_3Ti surface layer on TiAl intermetallic compound substrate. A mixture of Al and Ti powders with composition Al-25at% Ti was compacted onto the TiAl cast material, and the sample was hot-pressed at 1023 K. A single-phase Al_3Ti surface layer with little porosity was formed by SHS reaction. Matsubara et al.[39)] studied the fabrication of a thick Al_3Ti surface layer on Ti substrate by reactive pulsed-electric current sintering (PECS). PECS was reported as an effective method to densify powder products at a lower temperature and for a shorter time than other conventional processes by charging a pulsed electric current directly through the powders. The researchers used mechanically milled Al and Ti powder mixtures for reaction synthesis. During heating by PECS, Al and Ti reacted to form an Al_3Ti layer, and simultaneously reacted with the Ti substrate to achieve bonding between the surface layer and the substrate. It was reported that a fully dense and homogeneous Al_3Ti surface layer with a thickness of about 1.6 mm was obtained by processing at 1100 K for 3 min under 40 MPa. However, a higher temperature (1210 K) or longer holding time (over 30 min) was required to eliminate voids at the Al_3Ti/Ti interface. The Al_3Ti layer obtained exhibited almost the same hardness, wear and oxidation properties as the cast Al_3Ti material[39)].

A combustion-synthesized thick NiAl coating was also produced on an ultralow-carbon steel in a study by Matsuura et al.[40)]. They found that the processing conditions to obtain the desired coating depended upon the thickness of the compacted powder mixture. When the thickness of the powder compact on the steel was above 5 mm, a self-propagating combustion reaction of Ni + Al → NiAl was induced by heating to approximately 900 K at a heating rate of 1 K/s under a pseudo-isostatic pressure. The maximum temperature of the compact exceeded the melting point of NiAl (1911 K), and a NiAl coating on the steel was produced in a very short time. When the compact thickness was less than 5 mm, on the other hand, the synthesis reaction was incomplete with intermediate products such as Ni_3Al, $NiAl_3$, Ni_5Al_3 and unreacted Ni remaining in the coating. In this case, a fully-reacted coating had to be obtained by additional heating to a higher temperature (1473 K) and holding at the temperature for 540 s. The resultant NiAl coatings showed a tensile bonding strength of over 150 MPa between the coating and the substrate[40)]. In another investigation, these researchers studied the fabrication of a NiAl cladding on steel substrate by reactive casting[41)]. By pouring molten Al and Ni liquids onto a steel substrate placed in a crucible, molten NiAl was exothermically synthesized. A surface layer of the steel substrate was melted due to the reaction-generated heat. A NiAl-based (Ni, Fe)Al intermetallic compound cladding was formed and well-bonded to the steel substrate after it solidified. Room-temperature four-point bending tests showed an average bending strength of 220 MPa for the NiAl/steel claddings. It was also reported that the obtained cladding exhibited excellent corrosion and oxidation resistance.

Shcherbakov and Shteinberg fabricated TiC-30Ni coating layers on a steel-45 substrate by SHS joining for production of stamp products[25)]. Trofimov and Yukhvid[42)] produced a wear-resistant coating consisting of chromium carbide and an iron-based binder on a steel substrate using SHS reactions accomplished in a medium-frequency electromagnetic field. The powder reactant mixture was prepared from FeO, Cr_2O_3, Al and C. In fact, the thermite-type SHS reactions have been successfully applied in combination with centrifugal motion as a production method to deposit corrosion-resistant coatings on the inner wall of pipes[43)].

5. Summary

Joining by combustion synthesis or self-propagating high-temperature synthesis (SHS) possesses innovative joining capabilities for advanced materials. During the past decade, considerable research effort has been directed to understanding the fundamentals of the novel joining process, joining technique developments for various materials, microstructural and mechanical characterization of the joints obtained, and process optimization and control. The principle and characteristics of the joining process, the current development in fabrication, microstructure and mechanical properties of joints and coatings made by this method have been reviewed. The review of the available literature shows that SHS joining has been successfully utilized to join a variety of advanced materials including intermetallics, refractory metals, ceramics, and composites. Although SHS joining is largely still in the stage of laboratory investigation, there have been several examples of practical applications. Through further research and development, this attractive new process will fully exhibit its great technical and economical potential for advanced materials joining and related net-shape manufacturing.

Based on this literature survey, further studies are considered to be needed and expanded in the following directions. Much more research is needed into the fabrication of composite joints by incorporating various reinforcing fibers and whiskers, in addition to particles, into reactants and, more significantly, through in-situ synthesis of reinforcements. Technique developments are also necessary for production of FGM joints, e.g. ceramic-to-intermetallic, and ceramic-to-metal FGM joints. Research and development work is also badly needed in near-net-shape fabrication of joints through a combined process of parent materials preparation and joint production, also referred to as primary joining, preferentially for production of composite joints and FGM joints. Process optimization and extensive assessments and optimization of mechanical and other properties of SHS joints of various materials are to be further conducted as a prerequisite for practical applications.

References

1) R. W. Messler, Joining of Advanced Materials, Butterworth-Heinemann, Stoneham, Mass. USA, 1993.
2) V. K. Sikka, in High Temperature Aluminides and Intermetallics, ed. by S.H. Whang, C.T. Liu et al., Minerals, Metals and Materials Society, Warrendale, USA, 1990, 505-511.
3) S. A. David, W. A. Jemian, C. T. Liu, J. A. Horton, "Weldability of an Iron-Containing Ni_3Al Alloy", Welding Journal, Vol. 64 (1985), 22s-28s.
4) W. A. Baeslack III and T. Broderick, "Effect of Cooling Rate on the Structure and Hardness of a Ti-26Al-10Nb-3V-1Mo (at.%) Titanium Aluminide", Scripta Met., Vol. 24 (1990), 319-324.
5) H. Li, R. H. Jones, "Effect of Pre-welding Heat Treatments on Welding a Two-Phase Ni_3Al Alloy", Materials Science and Engineering, Vol. A192/193 (1995), 563-569.
6) G. Cam and M. Kocak, "Progress in Joining of Advanced Materials", Inter. Mater. Rev., Vol. 43 (1998), No.1, 1-44.
7) W. F. Gale, "Diffusion Brazing of Advanced Materials", in Brazing, High Temperature Brazing and Diffusion Welding, DVS Ber.166 (Düsseldorf, DVS Verlag, 1995), 231-234.
8) R. R. Kapoor and T. W. Eagar, "Oxidation Behavior of Silver- and Copper-Based Brazing Filler Metals for Silicon Nitride/Metal Joints", J. Am. Ceram. Soc., Vol.72 (1989), 448-454.
9) A. G. Merzhanov, "Self-propagating High-temperature Synthesis – Twenty Years' Search and Findings", in Combustion and Plasma Synthesis of High-Temperature Materials, ed. by Z.A. Munir and J.B. Holt (VCH Publishers, New York, 1990), 1-18.
10) J. J. Moore and H. J. Feng, "Combustion synthesis of advanced materials: Part I. reaction parameters", Prog. in Mater. Sci., Vol. 39 (1995), No.4/5, 243-273.
11) J. J. Moore and H. J. Feng, "Combustion synthesis of advanced materials: Part II. Classification, applications and modelling", Prog. in Mater. Sci., Vol. 39 (1995), No.4/5, 275-316
12) A. Bose, B. Moore, R. M. German, N. S. Stoloff, "Elemental Powder Approaches to Ni_3Al-Matrix Composites", Journal of Metals, Vol.40 (1988), No.9, 14-17.
13) J. P. Lebrat, A. Varma and A. E. Miller, "Combustion Synthesis of Ni_3Al and Ni_3Al-Matrix Composites", Metallurgical Transactions, Vol. 23A (1992), 69-76.
14) S. D. Dunmead, Z. A. Munir, J. B. Holt and D. D. Kingman, "Simultaneous Synthesis and Densification of TiC/Ni-Al Composites", J. Mater. Sci., Vol.26 (1991), 2410-2416.
15) W. C. Williams and G. C. Stangle, "Fabrication of Near-Net-Shape Al_2O_3-Fiber-Reinforced Ni_3Al Composites by Combustion Synthesis", J. Mater. Res., Vol.10 (1995), No.7, 1736-1745.
16) Y. Miyamoto, T. Nakamoto, and M. Koizumi, "Ceramic-to-Metal Welding by a Pressurized Combustion Reaction", J. Mater. Res., Vol. 1 (1986), No.1, 7-9.
17) B. H. Rabin, "Joining of Fiber-Reinforced SiC Composites by in-situ Reaction Methods", Materials Science and Engineering, Vol. A130 (1990), L1-L5.
18) B. H. Rabin: "Joining of Silicon Carbide/Silicon Carbide Composites and Dense Silicon Carbide Using Combustion Reactions in the Titanium-Carbon-Nickel System", J. Am. Ceram. Soc., Vol. 75 (1992), 131-135.
19) R. W. Messler, T. T. Orling, "Fundamentals of the SHS Joining Process", in Proc. MRS Symp. on Joining and Adhesion of Advanced Inorganic Materials, Vol. 314, ed. by A.H. Carim, D.S. Schwartz and R.S. Silberglitt (Materials Research Society, Pittsburgh, Pa., 1993), 177-182.
20) R. W. Messler, T. T. Orling, "Process Parameter

Effects in Pressurized Combustion Synthesis or Reactive Joining", Journal of Materials Synthesis and Processing, Vol. 2 (1994), No.5, 315-328.

21) R. W. Messler, T. T. Orling, "Joining by SHS", in Advanced Processing Techniques – Particulate Materials - 1994, Vol. 6, 273-285.

22) R. W. Messler, M. A. Zurbuchen, T. T. Orling, "Welding with Self-Propagating High-Tempeature Synthesis", Welding Journal, Vol.74 (1995), No.10, 37-41.

23) T. T. Orling and R. W. Messler, "Self-Propagating High-Temperature Synthesis as a Process for Joining Materials", Welding Journal, Vol.75 (1996), No.3, 93s-100s.

24) J. K. Wright, R. N. Wright and G. A. Moore, "Combustion Synthesized Iron Aluminide Coatings and Joints", Scripta Metallurgica et Materialia, Vol. 28 (1993), 501-506.

25) V. A. Shcherbakov and A. S. Shteinberg, "SHS Welding of Refractory Materials", Int. J. SHS, Vol.2 (1993), No.4, 357-369.

26) K. Uenishi, H. Sumi and K. F. Kobayashi, "Joining of the Intermetallic Compound TiAl Using Self-propagating High-tempeature Synthesis Reaction", Z. Metallk. Vol. 86 (1995), No.1, 64-68.

27) W. Liu, D. Zhou and F. Zhai, "Joining Advanced Materials by Presssurized Combustion Synthesis", in Joining Ceramics, Glass and Metal, ed. M. Turwitt (Düsseldorf: DVS Verlag, 1997), 231-234.

28) W. Liu, F. Zhai, C. Ding, "Joining of Ni_3Al by Pressurized Combustion Synthesis Using Gleeble 1500 Test Simulator", Acta Metallurgica Sinica, Vol. 13, No.1, 217-222.

29) W. Liu, S. Liu, F. Zhai and G. Elssner, "Pressurized Combustion Synthesis as a Technique to Join Advanced High Temperature Materials", in Proceedings of the 3rd Pacific Rim International Conference on Advanced Materials and Processing, ed. by M.A. Imam, R. DeNale, S. Hanada, Z. Zhong and D.N. Lee (TMS, Warrendale, USA, 1998), 2275-2282.

30) W. Liu, D. Zhu and G. Cong, "Combustion Synthesis of NiAl and In-Situ Joining to Ni-based Superalloys", J. Mater. Sci. Technol., Vol. 17, No.1, 179-180.

31) W. Liu, Y. Gong, and M. Xie, "Fabrication of Intermetallic-Matrix Composite Joints by Pressurized Combustion Synthesis", Paper presented at Materials Congress'98, Cirencester, UK, April 1998.

32) W. Liu, F. Zhai and S. Liu, "Fabrication of Ni_3Al Matrix Composite Joints by Pressurized Reaction Synthesis", in Advanced Brazing and Soldering Technologies (International Brazing & Soldering Conference Proceedings), ed. by P.T. Vianco and M. Singh (Albuquerque, NM, USA, 2000), 73-80.

33) W. Liu, D. Zhu and M. Naka, "Reaction Synthesis of NiAl-TiC Composites and In-situ Joining to Ni_3Al Alloys", Proceedings of the National Conference of Japan Welding Society, April 2001 (in press).

34) W. Liu, F. Zhai and S. Liu, "An Investigation of Joining Al_2O_3 by Combustion Synthesis", Journal of Dalian Railway Institute (in Chinese), Vol. 20 (1999), No.1, 57-62.

35) D. Zhu, "Combustion Synthesis of TiC/NiAl Composites and In-situ Joining", M. Sc. Thesis (in Chinese), Dalian Railway Institute, June 2000.

36) R. Kudesia, S. E. Niedzialek and G. C. Stangle, Ceram. Eng. Sci. Proc., Vol. 13 (1992), No.7/8, 374-379.

37) K. Uenishi and K. F. Kobayashi, "Processing of Intermetallic Compounds for Structural Applications at High Temperature", Intermetallics, Vol. 4 (1996), S95-S101.

38) K. Uenishi, T. Matsubara and K. F. Kobayashi, "Combustion Synthesis of the Intermetallic Compound Al_3Ti and its Simultaneous Joining with TiAl", Z. Metallk. Vol. 90 (1999), No.2, 163-167.

39) T. Matsubara, T. Shibutani, K. Uenishi and K. F. Kobayashi, "Fabrication of a Thick Surface Layer of Al_3Ti on Ti Substrate by Reactive Pulsed-Electric Current Sintering", Intermetallics, Vol. 8 (2000), 815-822.

40) K. Matsuura, K. Ohsasa, N. Sueoka, and M. Kudoh, "Nickel Monoaluminide Coating on Ultralow-Carbon Steel by Reactive Sintering", Metall. and Mater. Trans.Vol. 30A (1999), 1605-1612.

41) K. Matsuura, H. Jinmon and M. Kudoh, "Fabrication of NiAl/Steel Cladding by Reactive Casting", ISIJ International, Vol. 40 (2000), No.2, 167-171.

42) A. I. Trofimov and V. I. Yukhvid, "SHS Surfacing in an Electomagnetic Field", Int. J. SHS, Vol.2 (1993), No.4, 343-348.

43) O. Odawara and J. Ikeuchi, "Study on Composite Materials with a Centrifugal-Thermit Process", J. Japan Inst. Metals (in Japanese), Vol. 45 (1981), No.3, 316-321.

44) J. A. Hawk, A. V. Petty, C. P. Dogan and J. C. Rawers, "Self-Propagating High-Temperature Synthesis as a Technique to Join Metals", in Proc. MRS Symp. on Joining and Adhesion of Advanced Inorganic Materials, Vol. 314, ed. by A.H. Carim, D.S. Schwartz and R.S. Silberglitt (Materials Research Society, Pittsburgh, Pa., 1993), 183-194.

45) D. E. Alman, J. C. Rawers and J. A. Hawk, "Microstructural and Failure Characteristics of Metal-Intermetallic Layered Sheet Composites", Metall. and Mater. Trans. A, Vol. 26A (1995), 589-599.

46) H. Doty and R. Abbaschian, "Reactive Hot Compaction of NiAl with in situ Alumina Reinforcement", Materials Science and Engineering A, Vol. 195, 101-111.

47) D. Padmavardhani, A. Gomez and R. Abbaschian, "Synthesis and Microstructural Characterization of NiAl-Al_2O_3 Functionally Gradient Composites", Intermetallics, Vol. 6 (1998), No.4, 229-241.

48) W. Liu, D. Zhou and F. Zhai, "Finite Element Analysis of Residual Stresses in Ceramic/Metal Joints with Functionally Gradient Material Interlayers", in Brazing, High Temperature Brazing and Diffusion Welding, ed. by E. Lugscheider (Düsseldorf, DVS Verlag, 1998), 346-350.