



Editorial

Joining Technology Innovations at the Macro, Micro, and Nano Levels

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Abstract: With the growing joining requirements of emergent engineering materials and new applications, conventional welding continues to evolve at all scales spanning from the macrodown to the micro- and nanoscale. This mini review provides a comprehensive summary of the research hot spots in this field, which includes but is not limited to selected papers from the international nanojoining and microjoining conference (NMJ) held in Nara, Japan on 1–4 December 2018. These innovations include the integration of nanotechnology, ultrafast laser, advanced manufacturing, and in situ real-time ultra-precision characterization into joining processes. This special issue may provide a relatively full picture of the state-of-the-art research progress, fundamental understanding, and promising application of modern joining technologies.

Keywords: joining; nanotechnology; nanomaterials; nanojoining; nanobrazing; nanosoldering; nanosintering; nanopastes; nanomultilayers; packaging

1. Evolution of Conventional Brazing, Tungsten Inert Gas (TIG) Welding, and Soldering

Engineering applications at high temperatures, which demand high reliability and high resistance to harsh environments, persistently drive the development of high-performance engineering materials such as high-temperature Ni- and Co-based superalloys [1], high-entropy alloys [2], and oxide-dispersion enhanced Al-based, Ti-based, and Mg-based alloys [3]. The mechanical properties of these materials are improved through either one or more of the following microstructure modifications and phase controls: grain-boundary refinement, nanoscopic precipitation, and the addition of heterogeneous nanoparticles. The mechanical strengthening, thus, arises from different mechanisms, such as grain-boundary strengthening, precipitate strengthening, and dispersion strengthening. Conventional welding is not suitable for joining these materials since these microstructures may be destroyed during conventional welding processes [4]. Additionally, the formation of brittle intermetallic compounds, solidification cracking, and liquation cracking are additional limiting factors [2,3]. The integration of nanotechnology is an effective strategy to address these issues.

Recently, metallic nanoparticles were successfully applied as brazing filler materials without the usage of conventional melting point depressants such as boron, silicon, and phosphorus. With the long-known size-induced melting point depression of nanoparticles, high shear bonding strength was achieved in joining Inconel 718 using Ni nanoparticles brazed at a lower processing temperature than a commercial brazing material (Figure 1) [5].

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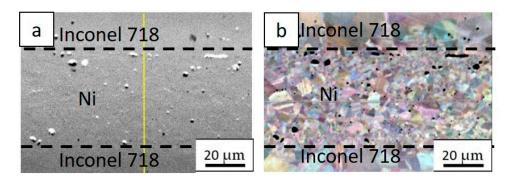


Figure 1. Inconel 718 Joint with Ni Nanomaterials as the filler metal: (a) SEM image; (b) forescatter detector image.

The usage of TiC nanoparticles to hinder the growth of grain boundaries showed that arc welding can join Al7075 without the formation of microcracks [3]. These nanoparticles can be dispersed into the metallic matrix through either mechanical alloying [6] or friction stir processing [7]. Another innovative welding method is the usage of metal-oxide powder to enhance the welding penetration through a change in the Maragoni flow in the melting pool [8]. The surface coating of SiO₂ nanoparticles displayed a significant improvement of the depth-to-width ratio with arc tungsten inert gas welding than the coating of SiO₂ microparticles.

In this special issue, cold metal transfer welding–brazing was applied for the joining of Al7075 to galvanized steel. AlSi5 filler wires were used to enhance the wettability of galvanized steel [9]. It was found that preheating the weldment at $100\,^{\circ}\text{C}$ to $200\,^{\circ}\text{C}$ could significantly reduce the loading force and suppress the creation of fractures. In another work focusing on the microstructure and fatigue damage of metal insert gas welding Al6082, incomplete penetration was identified as the dominant factor for limited welding strength [10]. For low-temperature soldering studies, Yan et al. [11] studied the aging effect of Pb-rich phase on Sn–Pb alloys, and Xue et al. [12] studied the effect of γ -irradiation on the formation of micro-voids. To enhance the bonding strength, Chen et al. induced Cu to suppress the void formation in Sn-soldering [13], and Xue et al. [14] used resin to strengthen the Pb–Bi solder paste. Watanabe et al. [15] studied the effect of Fe–C plating on the erosion resistance of lead-free solder. These studies pioneered the energetic research of low-temperature soldering and packaging.

2. Innovative Processing on Micro/Nanojoining

Weight saving of products with keeping strength is a recent world mission in industrial fields. Dissimilar joining is one of the solutions. Joining of steel with aluminum is the most important and challenging technical issue in the automotive industry, because brittle Fe–Al intermetallic compounds (IMCs) formed between steel and aluminum decrease the mechanical properties of the joint. Controlling the IMCs is the key factor to achieve a strong steel–aluminum joint [16–19]. Yang et al. investigated the microstructure and mechanical properties of dissimilar Al/steel joints with and without nickel coating [20]. They found that the Ni coating acted as a physical barrier to avoid the direct mixing of Fe and Al elements, which inhibited the formation of brittle Fe_2Al_5 . Hence, it led to the improvement of joint strength.

Direct joining of metal with polymeric material is also an important technical issue in many industrial fields, in particular, the automotive and aeronautical industries [21]. Wang et al. produced hybrid joints of Ti6Al4V and glass fiber-reinforced polyamide using a direct laser joining method [22]. Surface texturing treatment was performed on Ti6Al4V before the laser joining process, producing a strong mechanical anchor effect between Ti6Al4V and PA66-GF30 after the solidification of PA66-GF30 melts. A modified direct laser joining process was proposed to effectively eliminate the generated micropores in the joints. The fracture strength of Ti6Al4V–PA66-GF30 joints was improved to 41.5 MPa, and fractures of the joints occurred at the bonding interface between Ti6Al4V and PA66-GF30.

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Focused ultrashort laser pulse induces a variety of nonlinear phenomena inside transparent materials, producing many attractive novel applications [23–25]. Sugimoto et al. applied the light-emitting diode (LED) array microscope to visualize femtosecond laser-induced structural modifications [26]. Structural changes induced inside BK7 glass using a femtosecond laser were observed using the LED array microscope. They demonstrated bright-field, dark-field, and differential phase contrast (DPC) images of the structural changes using an LED array as a light source and by temporally changing the illumination pattern. They also demonstrated simultaneous acquisition of bright-field and dark-field images of structural modifications in glass by color-coded Rheinberg illumination and subsequent computational image processing.

Seeing is believing. Ultrasonic bonding is widely used in industrial fields; however, there are still many unresolved phenomena. Takahashi et al. directly observed the interfacial adhesion behavior during ultrasonic bonding between an Al ribbon and transparent silica substrate using a high-speed video camera with differing frame rates [27]. Initially, friction slip occurred, producing multiple island streaks in the direction parallel to the ultrasonic vibration. The island streaks were formed as a scratch, due to surface waviness of the Al ribbon. Momentarily, a belt-shaped bond zone was formed at the center, normally due to the ultrasonic vibration. The positional relationship between the island streaks and the central belt zone was confirmed from in situ observation results of a twist and peel test of Al ribbon bonded to silica substrate. The central belt zone was between the island streaks and the silica substrate.

3. Micro/Nanoscale Joined Interfaces

The design and control of interfacial structures at the micro- and/or nanoscale is of great importance with regard to materials science and engineering in the implementation of joining processes. Realized or not, it is also one of the key elements in traditional macro-welding technologies. Recently, this is being widely used in micro- and nanoscales to develop new joining processes, ranging from nanodevices [28] to electronic packaging [29,30], soldering [31] to light alloy (Ti, Al) [32,33], and high-strength steel joining [34]. The goal of design strategy is to meet the performance requirements of as-fabricated joints through the proper determination of the necessary number of IMCs, layered structures, phases, intrinsic properties of used materials (e.g., hardness, grain size, etc.), and so on. Control strategy, on the other hand, is implemented by altering the heat source, processing time, shape and size of grains, diffusion rate, phase transformation, formation and structure of IMCs, crystallographic orientation, etc. With the proper design and deep understanding of the relation between interfacial structures and joint properties, engineers should be able to target the scientific paths of controlling methods and the optimal processing combination of joining parameters. The right combinations of joining parameters should give the as-fabricated joints predictable engineering properties, such as high strength, long fatigue time, good conductivity, etc. Typical examples were reported recently by characterizing and analyzing the interface of joints.

Fushimi et al. [29] reported that hard copper materials and fine grains could suppress the crack propagation from the thermal fatigue of ultrasonically bonded copper joints for electronic devices by providing more grain boundaries at the interface, which can be controlled by longer friction time and grain refinement. When Hang et al. [31] studied the interfacial intermetallic growth of Sn–37Pb solder joints, under –196 °C to 150 °C thermal shocking, they found that plane-type IMCs and layered Cu₃Sn could grow at the interface because of the bulk diffusion and grain-boundary diffusion, resulting in the ductile fracture to mixed ductile–brittle fracture transition and the reduction of joint strength. It is speculated that brittle fracture could also be further improved by designing the shape of formed interfacial IMCs and controlling the diffusion. Another example from Ma et al. [33] showed the importance of designing the interfacial structure for the joining of two very different materials, carbon fiber-reinforced plastic (CFRP) epoxy composite to Al alloy, using Ni/Al reactive multilayer films (RMFs). The RMFs were custom-designed to meet the requirements of heat generation to melt polycarbonate filler material and facilitate the formation of CFRP–Al alloy bonded interfaces. When dealing with such

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heat-sensitive materials, heat or temperature management during welding or joining needs innovation and creativity in both science and engineering aspects. High-temperature welding might change the phase and composition of materials (for example, low-temperature partitioned quenching and partitioning (Q&P) steel), and cause property degradation. Hausner et al. [34] used Sn or SnAg (Ag nanoparticles with an Sn sandwich filler) to lower the joining temperature to 237 °C with an unchanged microstructure of the Q&P steel.

Femtosecond lasers were also used to minimize heating effects during the joining process or change the interfacial structure of contacts between nanowires [35] or nanotubes [28] to metal electrodes. Laser-induced connections can significantly reduce the contact resistance between carbon nanotubes (CNTs) and electrodes, as reported by Cui et al. [28], or possibly modify the band structure at the interface for memory device applications as demonstrated by Lin et al. [9]. By further reducing the external energy input and taking advantage of the nanosize effect, the self-powered brazing of Ti6Al4V using Ni/Al nanomultilayers was achieved by Bridges et al. [36] with a maximum reaction temperature of 683 °C to melt the BAlSi-4 brazing material. To further improve the strength of this brazed joint, ongoing research is being conducted on the usage of ductile nano multilayers and/or modification of the bonding interface configuration. The high energy of nanomaterials also enables room-temperature joining [37] for electronic packaging through self-generated heat [38] or ultra-dense atomic defect-motivated fast diffusion [39]. Aside from heat, the control of grain orientation [30], size [40], or interfacial stoichiometry [41] during the joining process could also be employed for the engineering of the high thermal conductivity, ultrahigh yield strength, or electron transportation efficiency of interfacial structures.

4. Micro/Nanojoining for Microelectronics

Recently, there was significant progress in the development of bonding materials and joining processes for power electronic devices and flexible electronics. Furthermore, new advances in the bonding of nanostructures, an important step toward the transition to nanoelectronics, were reported.

In the field of power electronics, the focus is directed at the development of novel materials for transient liquid phase bonding (TLP) and nanoparticle sintering routes. Rajendran et al. [42] proposed an Sn-coated Cu–multiwalled CNT (MWCNT) composite (Sn–Cu–MWCNT) paste for Cu interconnections, and Tatsumi et al. [43] developed a Cu–solder–resin composite paste for pressure-less die-attach technology for bonding Kovar chips onto Cu electrodes on Si₃N₄ substrates. The Sn–Cu–MWCNT composite powder [42] was fabricated by subsequent electroless plating of Cu and Sn on a multi-walled carbon nanotube. It can be applied as a paste for the fabrication of Cu interconnects with improved mechanical properties at temperatures of 260 °C and applied pressure of 10 MPa. During the process, the Sn–Cu–MWCNT composite material transformed to MWCNT–Cu₃Sn in the Cu joint zone. The Cu–solder–resin composite pastes developed by Tatsumi et al. [43] allowed a TLP sintering (TLPS) process at a temperature of 250 °C without pressure application. The as-produced TLPS joints showed excellent thermal reliability (only 14% decrease in shear strength after thermal aging at 200 °C for 1000 h). This was due to their unique microstructure consisting of a Cu particle–IMC skeleton partially filled with polyimide resin in the as-bonded state.

The growing interest in soft electronic components and devices drives the development of micro/nanojoining technology. The aim is to provide solutions for the integration of diverse passive and active components on flexible (elastic) substrates for various applications from wearables to robotics, energy storage applications, bioelectronics, and functional textiles. Wang et al. [44] reported the development of binder-free conductive composite inks with adjustable electrical and mechanical properties for flexible electronics applications. The developed binder-free composite inks were composed of silver nanoparticles, binary solvents, and conductive nanofillers of various architecture and composition. The inks were successfully used for laser-based fabrication of soft electrodes with low electrical resistance and good mechanical performance. It was shown that, using graphene filler, the electrical resistivity of electrodes could be significantly reduced, while adding silver nanowires to

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the ink improved the electrode flexibility. Thus, the properties of the joints could be tailored by the selection of the paste nanofiller according to the product requirements.

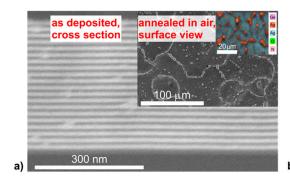
An alternative method to fabricate conductive electrodes, capacitors, and other flexible radio frequency (RF) devices is laser-direct writing (LDW), which gained a lot of interest as a simple and low-cost manufacturing method [45]. The LDW process is a noncontact, maskless, and gentle patterning method with a resolution that can go down to the submicrometer range. Li et al. [45] demonstrated that the transmission and reflection characteristics of laser-scribed microstrip transmission lines, basic components of RF circles, on flexible substrates can be simply tuned by accommodating the resistance of the conductors with a variation of the laser power.

Ultrafast laser ablation can be used for the reshaping, cutting, and joining of nanostructures, as well as for the formation of nanostructures with required optical properties and surface plasmon resonance [46]. Zhu et al. [46] demonstrated that Ag nanorods can be transformed into bone-shaped nanorods, T-shaped nanorods, and nanodots by controlling laser polarization direction and wavelength during the femtosecond pulse laser near-field ablation process. In the same way, nanojoints can be formed.

Ultrasonic welding was shown by Iwamoto et al. [47] to be an effective method for joining the stranded wires of coaxial cables with a coaxial connector in high-frequency signal devices with low transmission loss. In particular, high-quality joints of C- stranded wire consisting of seven Ag-coated Cu wires (70 microns in diameter) with Cu substrate (Ni/Au metallized) were produced by this method. As reported by various research groups, well-selected coatings play an important role in the achievement of defect-free, high-quality joints.

Material coating systems, in particular, nanostructured coatings, so-called nanomultilayers (NMLs), show unique properties and offer new opportunities for many applications. Very recently, NML coatings of brazing fillers alternated by a functional barrier (i.e., Ag/AlN, Cu/AlN, Cu/W, Al/AlN, AgCu/AlN, and AlSi/AlN) were designed to serve as low-temperature brazing nanofillers for packaging and assembling miniaturized devices and heat-sensitive components at ever-reduced processing temperature [48–54]. Fast diffusion and extensive mass transport were observed in these nanomultilayers at temperatures much below the melting temperature of the bulk brazing filler $(T > 250 \, ^{\circ}\text{C})$, and they were exploited for joining technology applications [48,50–53]. It was shown that the behavior of the NMLs, i.e., their structural evolution, strongly depends on many parameters such as geometrical constraints (layer thickness, number of repetitions), material composition, deposition parameter, interfacial structure, and internal stresses (Figure 2). The overall understanding of the effect of these parameters on the NML behavior in various service conditions is crucial for proper NML design. Using the example of Ag/Ge/AlN nanomultilayers, Cancellieri et al. [54] showed that the stress state of as-deposited NML structures can be controlled by the substrate bias during the deposition process or by the modification of the layer interfaces through the small addition of a second material. In this way, the thermal evolution and stability of the nanomulitlayers, as well as the directional mass outflow of the brazing filler material, can be tuned.

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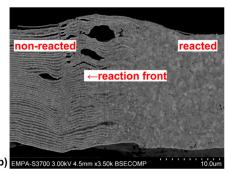


Figure 2. Nanomultilayers: A new class of joining materials: (a) non-reactive Ag/Ge/AlN nanomultilayer as a nanostructured brazing material for fast, low-temperature joining processes; (b) reactive Ni/Al nanomultilayer as a local heat source for bonding of heat-sensitive materials or for offside joining work (SEM cross-section).

Nanomultilayers can also serve as a local heating source when highly reactive materials undergoing a self-heating exothermic reaction are combined together, for example Ni/Al, Ti/Al, Zr/Al, and Pd/Al. Joining of typical packaging materials (borosilicate glass wafers, aluminum oxide, silicon, and copper) with Ni/Al reactive NMLs was presented by Rheingans et al. [55], and the effect of thermal conductivity of the base material on the joint evolution was analyzed in detail. A deep understanding of the interplay between the joint design (base components, metallization, reactive nanomultilayer, and solder), the thermal properties of the joining materials, and the resulting time–temperature profile of the joint microstructure is crucial for tailoring the joint properties.

5. Conclusions

Emergent engineering materials and nanotechnology significantly advanced conventional welding techniques, such as arc welding, soldering and brazing, activated arc welding, and nanobrazing for high temperature and aerospace applications. Nanopastes for printed electronics and low-temperature packaging are typical examples of innovations. Composite inks with adjustable properties for flexible electronics, hybrid transient liquid phase bonding materials for pressure-less die-attach technology, and nanomultilayers for low-temperature joining processes are other examples.

A wide variety of nonlinear phenomena are remarkable in the micro/nanojoining field, which has the potential to make a breakthrough for producing innovative processing. In particular, laser-based processes gained a lot of interest as an enabling technology for nanoelectronics and for the fabrication of diverse nanojoints. Interfacial structure design and control would enable engineers predict the properties of joints and develop new processes for advanced welding and joining technology from the macro- to the micro- and nanoscale.

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