



Multiple self-exothermic reactions for room-temperature aluminum bonding via instantaneous melting



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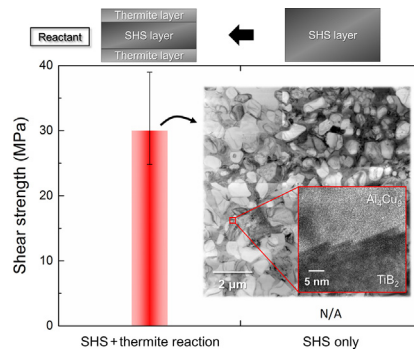
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HIGHLIGHTS

- Combination of self-propagating formation reaction and thermite reaction enable room-temperature bonding.
- The thermite reaction facilitates the inflow of molten metal, contributing to pore filling and formation of composite grains.
- TiB₂ and Al-Cu intermetallic composites are bonded in the joint owing to lattice defects relieving the interfacial misfit.
- Distribution of products is responsible for the joint strength, revealing the improvement by appropriate reactant design.

GRAPHICAL ABSTRACT



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ABSTRACT

We propose a room-temperature multiple-reaction bonding method for aluminum, by combining the thermite reaction of Al-CuO powder and self-propagating formation reaction of Ti-B powder with no external heat source. The joint microstructure consists in various composites (Al₂O₃, TiB₂ and Al-Cu intermetallic), and their distribution affects the joint bonding properties. The multiple reaction leads to a compact joint layer with a very low porosity, in contrast to joints formed only by a formation reaction. This observation is explained by the inflow of molten aluminum metal into the joint layer during the joint formation, a process which is central in the formation of a sound joint. Furthermore, bonding between different composites in the joint is achieved by lattice defects at their interface. Analysis of shear tests show that fracture occurs in regions at the metal base/joint interface where Al₂O₃ particles were segregated. It is concluded that appropriate reactant design plays a significant role in controlling the microstructure, and thus, the bonding properties of the joint.

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1. Introduction

Suppression of energy in the bonding process is necessary to prevent mechanical property degradation and to satisfy the demand for energy-saving applications. As localized heating is difficult to achieve for metals

with high thermal diffusivity, particularly for Al alloys, a large heat input is required for bonding [1,2]. Hence, there is considerable interest in developing a bonding method without external heat source since it facilitates the control of energy to form functional joint.

Self-propagating formation reaction, commonly called self-propagating high-temperature synthesis, is one type of chemical reaction using an inherent energy of materials. Exothermic heat is produced via material synthesis depending on the free-energy changes before and

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after the reaction. Whenever the amount of generated heat exceeds the activation energy for synthesis, a continuous chain of reactions occurs throughout the entire reactant [3–5]. Several studies have been conducted on the application of self-propagating formation reaction to bonding [6–13]. The use of nano-multilayered reactant is typically employed in the brazing [6–8] or bonding of various materials, such as bulk metallic glass and refractory materials [9,10], exploiting the high reactivity and large combustion speed due to the size effect that smaller particles facilitate them [14]. In addition, formation reaction using a mixture of powders has also been applied to bonding [11–13]. As the synthesis involves pore formation, it is generally used for the fabrication of biomaterials such as synthetic bone [15], as well as filtration media for molten metals [16]. On the other hand, the formation of a metal matrix composite or functionally graded materials can be achieved during the bonding process through the addition of a second phase to the pores [11,12]. However, there are several crucial problems in the bonding method using mixture of powders. One is that the bonding temperature should be raised close to the melting temperature of the reactant or bonding material in order to drive the reaction sufficiently. Another is the formation of large pores in the joint due to the local absence of heat since produced heat is consumed for heating the added powder. The presence of pores is a serious drawback as it reduces the joint strength [13].

Here, we suggest a bonding concept based on a multiple reaction combining formation and thermite reactions to replace the requirement for bonding temperature raise. The thermite reaction, an oxidation-reduction reaction, has been used to weld railroad or electronics parts [17], and produces both exothermic heat and pure metals [18], which can also be applied to formation of composite material [19]. The sole use of thermite reaction commonly results in pore formation owing to the formation of gas since the reaction temperature exceeds the boiling point of the produced materials [20]. However, a material synthesized by the formation reaction could act as a framework confining the product of the thermite reaction. Furthermore, the thermite reaction could drive the formation reaction when different reaction activation energies are appropriately chosen, so that no external energy supply would be needed. In the present study, we demonstrate the room-temperature bonding of Al alloy utilizing multiple reactions without any external heat source. Ti-B is used for the formation reaction because it has high adiabatic temperature [21], and the metal matrix compounds exhibit enhanced mechanical properties as well as enhanced thermal conductivity [22–24]. Moreover, Al-CuO is used for the thermite reaction, as it has a low activation energy (318 kJ/mol for Ti-B [25], but 128 kJ/mol for Al-CuO [26]).

2. Experimental procedures

Ti (99.9%, 38 μm) and B (95%, 3 μm) powders were prepared as reactants for the formation reaction. Al (99.9%, 20 μm) and CuO (99.9%, 1 μm) powders were also prepared as reactants for the oxidation-reduction reaction (thermite reaction). Ti and B were mixed at a 1:2 atomic ratio by planetary ball milling for 5 h. Al and CuO were mixed at a 2:3 atomic ratio using an agate mortar for 10 min. Fig. 1 shows a schematic illustration of the room-temperature bonding method based on a multiple reaction. A trilayer structure composed of Al-CuO (two external layers) and Ti-B (internal layer) powders was prepared, with a mass ratio of 1:3:1, and pressed at 500 MPa to form a green compact (ϕ : 10 mm) for the multiple reaction (Fig. 1(a)). A single Ti-B layer was also prepared for a single formation reaction for comparison (Fig. 1(b)). The mass and thickness of the trilayer and single layer compacts were 0.350 g, 1.53 mm and 0.352 g, 1.66 mm, respectively.

X-ray diffraction (XRD) measurements (CrK α irradiation) were performed using a RINT-2000 (Rigaku) in a Bragg-Brentano geometry in order to investigate the structure of the composites formed during the multiple reaction. The X-ray beams had a vertical and horizontal size

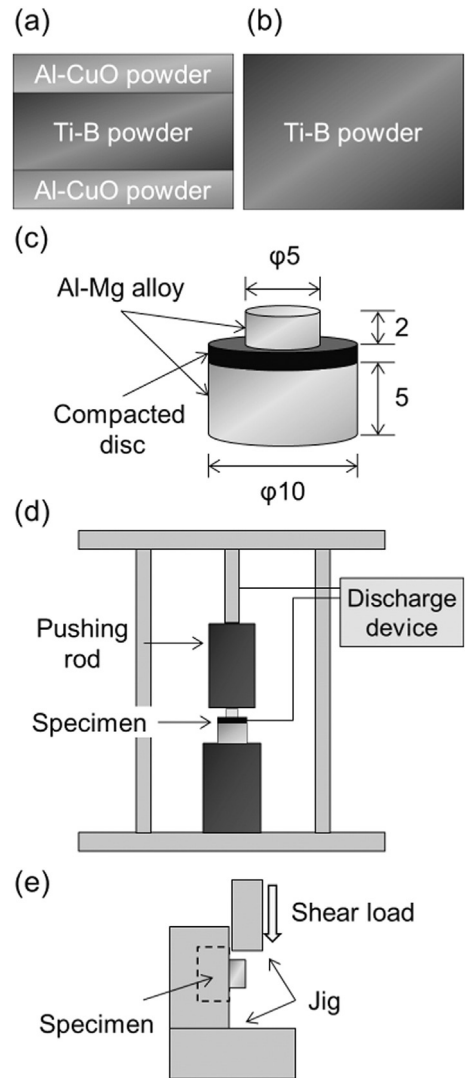


Fig. 1. Schematic illustration of the bonding method. (a,b) Structure of the trilayer and single-layer compacted discs. (c) Disposition and dimensions (in mm) of the specimen consisting of a compact disc between lower and upper Al-Mg cylindrical samples. (d) Bonding apparatus in ambient air. The ignition of the bonding reactions was carried out by capacitor discharge on the Al-CuO layer. (e) Shear test set-up.

of 1 mm. Both the reactant powders (Ti-B or Al-CuO mixed powders) and the green compact subjected to a multiple reaction (without base metal) at room temperature were investigated.

Al-Mg (5052 aluminum) alloy was used as the base metal to facilitate its identification through the presence of the Mg element. Its chemical composition is shown in Table 1. A green compact was inserted between lower and upper Al-Mg cylindrical samples, which were 5 and 10 mm in diameter and 2 and 5 mm in length, respectively (Fig. 1(c)). The bonding process, consisting in both the formation and thermite reactions, was driven at room temperature via instantaneous localized heating of the Al-CuO layer through capacitor discharge in the air at a pressure of 10 MPa (Fig. 1(d)).

The joint strength was measured by shear tests at a rate of 30 mm/min using AUTOGRAPH DCS-10t (Shimadzu) (Fig. 1(e)). The shear strength was defined as the value of maximum shear load during

Table 1
Chemical composition of 5052 aluminum (mass %).

| Si | Fe | Cu | Mn | Mg | Cr | Zn | Al |
|------|------|------|------|------|------|------|------|
| 0.09 | 0.28 | 0.01 | 0.03 | 2.65 | 0.18 | 0.02 | Bal. |

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