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High strength alumina joints via transient liquid phase bonding

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Abstract

Low melting boron oxide, instead of metallic materials in other methods of transient liquid phase bonding, was taken as braze in joining alumina in this paper. Pure boron oxide melts at low temperature and reacts with alumina matrix to form a stable high melting compound. This transient liquid phase bonding has the advantage of producing a ceramic joint for high temperature applications at low processing temperature. In this study, alumina pieces coated with boron oxide layers in various thicknesses were bonded at 800 °C for various times in air under minor loading. The average flexural strength of joints were measured by means of four point bending, while the microstructure of the cross-section and fractured surface was observed by means of scanning electron microscopy. Phases at joints were identified by low angle X-ray diffraction. The maximum flexural strength reaches a value of 155 MPa after joining at 800 °C for 15 h with a 21 μm interlayer. Three compounds, $3Al_2O_3$–B_3O_3, $2Al_2O_3$–B_3O_3 and $9Al_2O_3$–$2B_3O_3$ have been found at the joint. It is also found that $2Al_2O_3$–B_3O_3 whiskers dominate at the joint with the maximum strength.

Keywords: A. Joining; B. Interfaces; C. Strength; D. Al_2O_3; B_2O_3

1. Introduction

Many industrial processes need parts lasting under severe conditions including high temperature, high stress, corrosive solutions, biochemical reactions, etc. Undoubtedly, fine ceramics meet the need due to their special properties, including high melting/decomposition temperature, excellent wear/corrosion resistance, biocompatibility, high strength and hardness, etc. However, these properties make ceramic objects hard to form with complicated shape or large size. To join several ceramic parts into a large, complicated assemble is a promising way. It inspires the improvement of ceramic joining techniques aiming that mechanical properties of joined objects are influenced by the joint as little as possible.

The methods for joining ceramics include solid-state diffusion bonding [1], friction joining [2], laser welding [3], ultrasonic joining [4] and brazing [5]. Except brazing, ceramic joining is usually carried out at elevated temperature which results in the degradation of joint and hinders its application. Transient liquid phase bonding (TLPB), also known as ‘‘isothermal solidification’’ academically or as ‘‘diffusion brazing’’ technically, is a joining technique in which a low melting interlayer is introduced, melts at joining temperature and either reacts to form solid compounds or diffuse into matrix isothermally. Due to the formation of high melting compound(s) the operational temperature of the joined object can be higher than the joining temperature. This method has been successfully applied for joining some metallic and ceramic systems [6–10]. For joining alumina, different metallic materials as interlayer have been experimented and fine results have already been obtained. Using the Cu/Pt, Cu/Ni/Cu, Cu/Nb/Cu multilayer combinations, the joining temperature must be above 1000 °C and a flexural strength above 200 MPa has been reached. However, metallic brazes restrict the joining and usage environments of a joined component and reduces its practicability. Using metals with high temperature and oxidation resistance, e.g. Pt, Nb [11] or Au [12], is a way to improve the joining property in expense of high material costs.

Recently, a new attempt was proposed, instead of metallic interlayer, boron oxide interlayer was used for joining alumina pieces [13]. Glassy boron oxide has a very low melting point of 540 °C. The eutectic point in the B_2O_3/Al_2O_3 system is 470 °C. Two equilibrium incongruent compounds $2Al_2O_3$–B_2O_3 and $9Al_2O_3$–$2B_2O_3$ with melting point of 1035 and 1950 °C are

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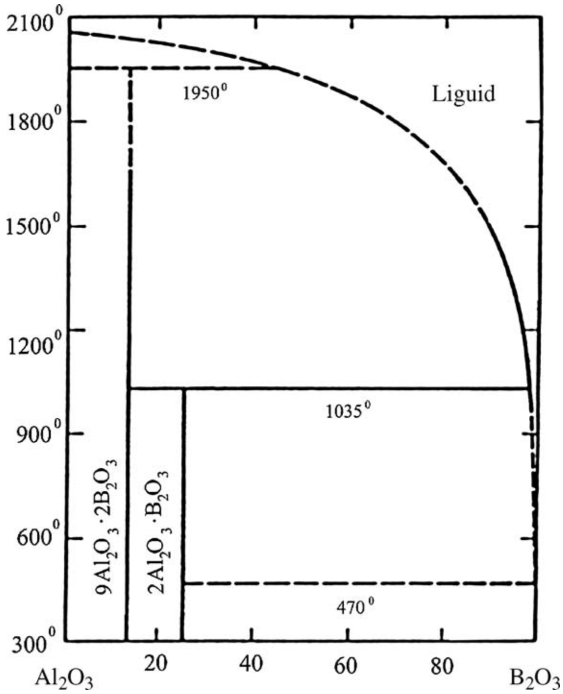

Fig. 1. Al_2O_3–B_2O_3 phase diagram [14].

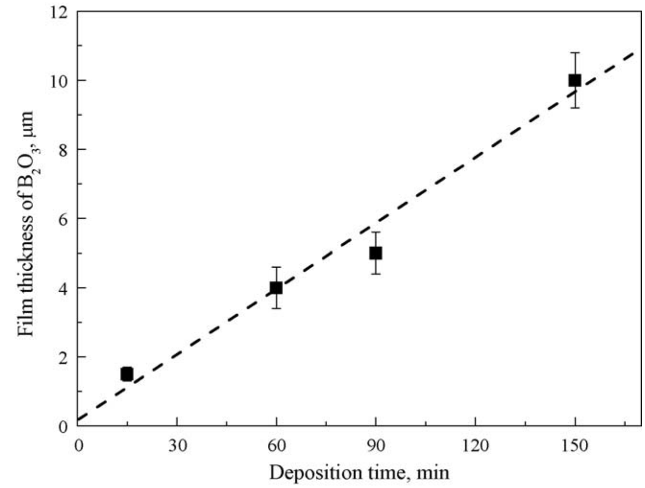

Fig. 2. Relation between the film thickness of boron oxide and deposition time.

seen in the phase diagram (Fig. 1) [14]. These high melting compounds formed at the joint of the $Al_2O_3/B_2O_3/Al_2O_3$ assembly after joining, but the maximum strength reported was merely 71 MPa with an interlayer thickness of 3 μm at a joining temperature of 800 °C. However, some advantages of this technique like moderate joining temperature, no precious metal and no protective atmosphere were needed to reduce the overall cost of joining. Besides, the ceramic interlayer may cause a galvanic effect much lower than that expected in a ceramics/metal heterogeneous system. The purpose of this work is to improve the flexural strength of alumina joint by optimizing the joining time and interlayer thickness of transient liquid phase bonding and to explain it with the formation and morphology of compounds at the interface.

2. Experimental

In this study, boron oxide was sandwiched in two alumina pieces. The raw materials are 99.6% α-alumina powder (ALCOA A16SG). Alumina slurry with 50% solid content and 0.05 wt.% dispersant (Darvan 7) was ball mixed for 24 h. Green cakes of alumina were formed by pressure slip casting with an air pressure of 7 atm, dried at 40 °C for 24 h, 60 °C for 24 h and at 200 °C for 6 h to completely remove water and dispersant. Finally, dried cakes were sintered at 1550 °C for 6 h to get alumina bulks with a density of 3.91 g/cm^2 and four-point flexural strength of 332 MPa.

Alumina bulks were slowly cut into pieces with the size of 15 mm × 20 mm × 10 mm by diamond saw. The joined surface was polished to 1 μm diamond paste and ultrasonically de-contaminated in ethanol. 99.6% boron oxide powder (STREM CHEMICALS) was heated to 300 °C on a hot plate (HP-46825) and boron oxide films in various thicknesses were deposited on water-cooled alumina surfaces in air for deposition times of 15, 60, 90 and 150 min. The relation between the film thickness and deposition time is plotted in Fig. 2. Two alumina pieces were vertically joined at the coated sides, put into an electric furnace (THERMOLYNE FB-1300) and dwelt at 800 °C for different times. A small load of 15 kPa was applied for keeping the interface in good contact.

Joined specimens were slowly cut-off in the direction perpendicular to the joint plane. The cross-section of the alumina joints was observed by SEM (JEOL JSM-5400). Six pieces of 4 mm × 4 mm × 25 mm specimens for each joining condition were four-point bended with MTS (HT-8116). In order to see the morphology and phases formed, the fractured surface of specimens was analyzed by using SEM and low-angle XRD (MAC SCIENCE MXP3), respectively.

3. Results and discussion

The average flexural strength of the alumina joints TLP bonded at 800 °C for different joining times and with various

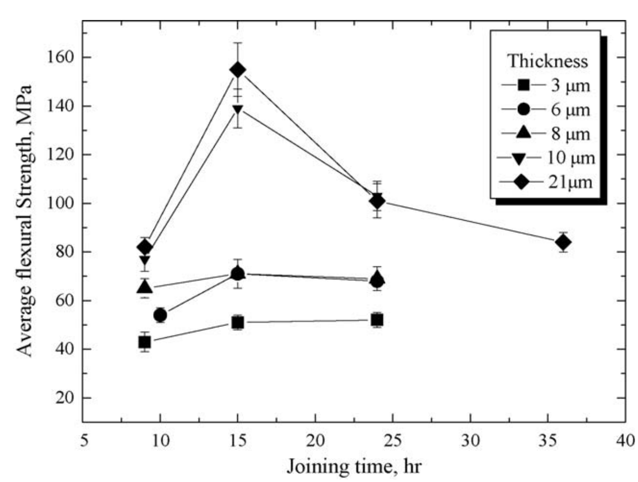

Fig. 3. Average flexural strength of the alumina joints TLP bonded for different joining times and with various interlayer thicknesses from the four-point bending test.

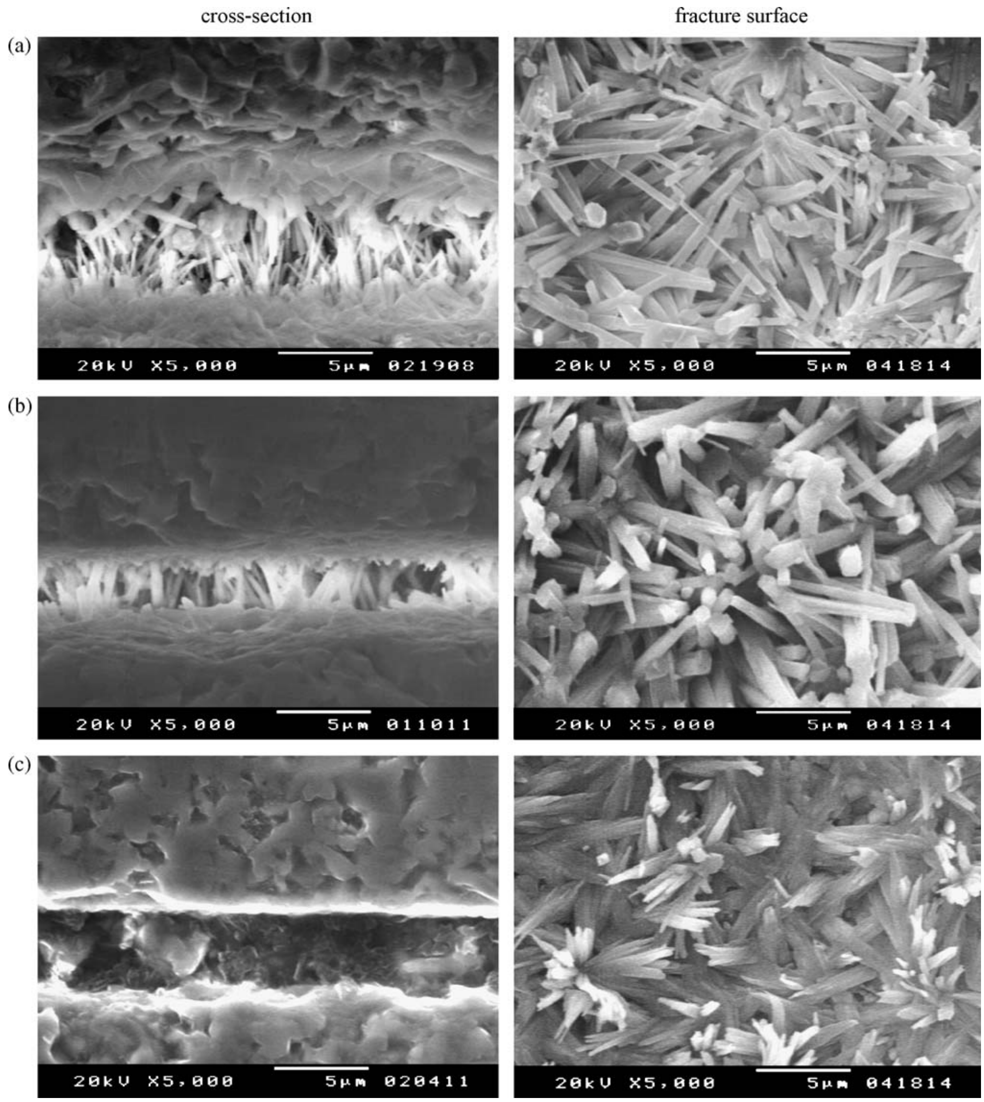

Fig. 4. SEM micrographs of the cross-section and fractured surface of the alumina joints TLP bonded at 800 °C via a 3 μm thick interlayer for (a) 9, (b) 15 and (c) 24 h.

interlayer thicknesses from the four-point bending test is drawn in Fig. 3. The average flexural strength of the alumina joints TLP bonded for 15 h via a 3 μm thick interlayer reaches 51 MPa. A longer joining does not benefit it. Fig. 4 shows the SEM micrographs of the cross-section and fractured surface of the alumina joints TLP bonded at 800 °C via a 3 μm thick interlayer for 9, 15 and 24 h. The joint thickness is about 4 μm after joining for 9 h. Whiskers initiate at the interface and grow radially across the joint (Fig. 4a). The crossing of these whiskers is observed. However, there are quite many vacant spaces between whiskers at the joint due to the deficiency of the boron oxide interlayer. The joint becomes narrower and the whiskers become thicker when the joint is heated for 15 h (Fig. 4b). Less vacant spaces are observed and this explains the increase of flexural strength. Although the joint seems to become denser from the cross-section view, the whisker structure remains unchanged as joined for 24 h (Fig. 4c). Consequently, the flexural strength remains constant.

Fig. 5 represents the XRD patterns of the fractured surface of the alumina joints TLP bonded at 800 °C via a 3 μm interlayer for 9, 15 and 24 h. The XRD pattern of alumina is also plotted as comparison. The characteristic peaks for identify the $2Al_2O_3–B_2O_3$ and $9Al_2O_3–2B_2O_3$ compounds lie at 25° and 42°, respectively. The XRD patterns indicate that the joints are basically composed of $2Al_2O_3–B_2O_3$ after a 15 h heating.

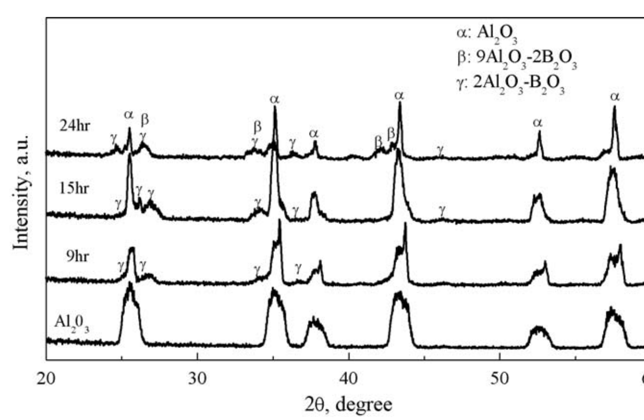

Fig. 5. XRD patterns of the fractured surface of the alumina joints TLP bonded at 800 °C via a 3 μm interlayer for 9, 15 and 24 h. The XRD pattern of alumina is also plotted as comparison.

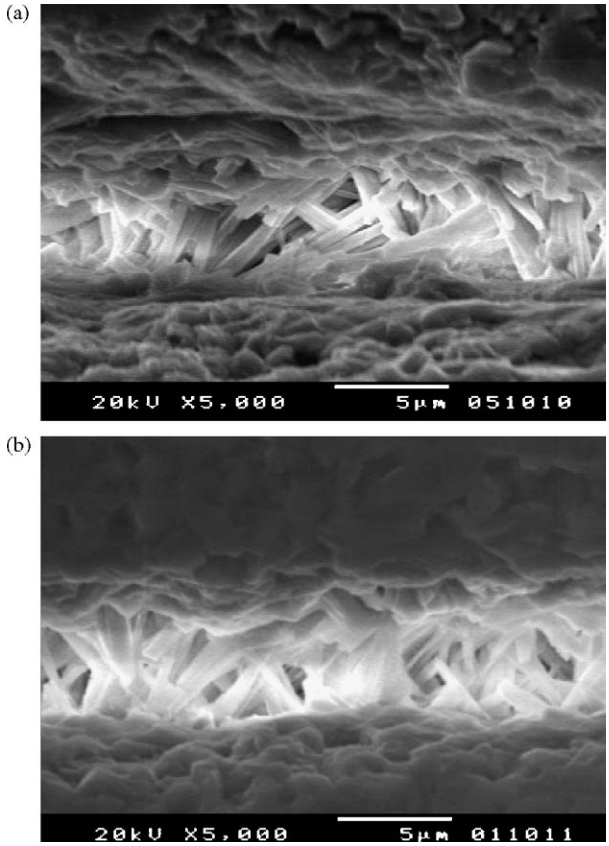

Fig. 6. SEM micrographs of the alumina joints TLP bonded at 800 °C for 15 h with interlayer thickness of (a) 10 and (b) 21 μm.

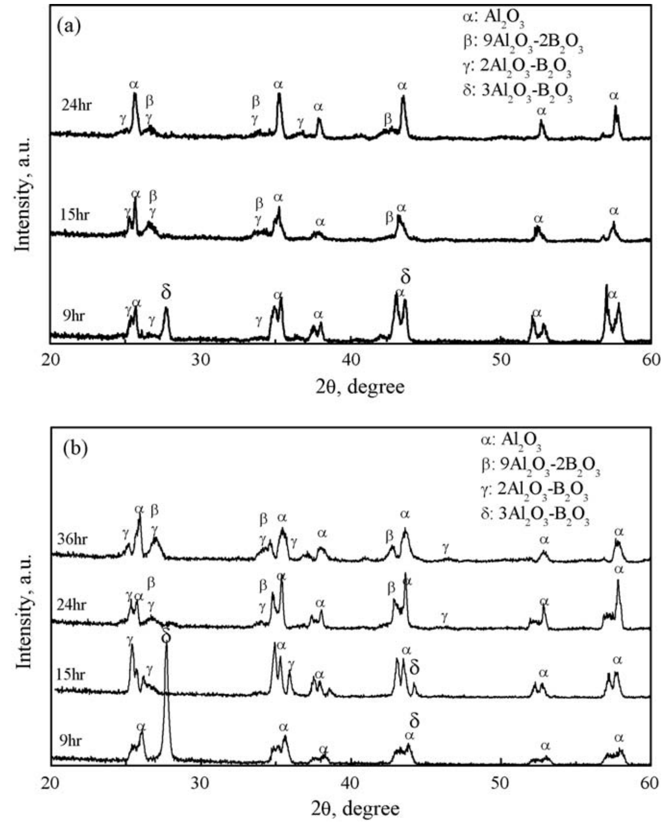

Fig. 7. XRD patterns of the fractured surface of the alumina joints TLP bonded at 800 °C via a (a) 10 and (b) 21 μm interlayer for different joining times.

As heating lasts for 24 h, $9Al_2O_3$–$2B_2O_3$ forms and replaces the $2Al_2O_3$–B_2O_3 compound.

It was proposed above that the deficiency of boron oxide interlayer may limit the strength improvement. Indeed, increasing the interlayer thickness enhances the joint. Joining with a thicker, 6 or 8 μm interlayer produces a joint with an average strength up to 71 MPa for all joining times (Fig. 3). The morphology and phase evolution at the joints are similar to that at the joints TLP bonded with a 3 μm interlayer.

A conspicuous enhancement is observed at the joint with even thicker interlayer. The data for the 10 and 21 μm samples in Fig. 3 indicate a twofold to threefold enhancement in comparison with that for the 3 μm sample. In both cases, joining respectively with 10 and 21 μm interlayer, the average flexure strength reaches the maximum at a joining time of 15 h. It is 139 MPa for 10 μm and 155 MPa for 21 μm samples. However, the average strength declines with further heating at 800 °C.

The SEM micrographs of the alumina joints TLP bonded at 800 °C for 15 h via respectively 10 and 21 μm interlayer are shown in Fig. 6. It is clearly seen that denser structures of the interlayer and less vacant spaces are found in these samples in comparison with the samples with a thinner interlayer as shown in Fig. 4. The diameter of the whiskers slightly increases when a larger amount of boron oxide interlayer is initially introduced. The connection between the alumina matrix and the compounds in interlayer is obviously improved. It is believed that more boron oxide at the interface acts as a filler material and this provides an effect of densification. The fact that the joint thickness does not change much with interlayer thickness prevents the possible drawback of a thick joint which is weaker than the host.

The XRD patterns for the 10 and 21 μm samples TLP bonded for various times are plotted in Fig. 7. Peaks for the metastable $3Al_2O_3$–B_2O_3 compound labeled as δ were found in the patterns for samples heated at 800 °C for 9 h. This metastable phase has been observed previously [13]. The protruding characteristic

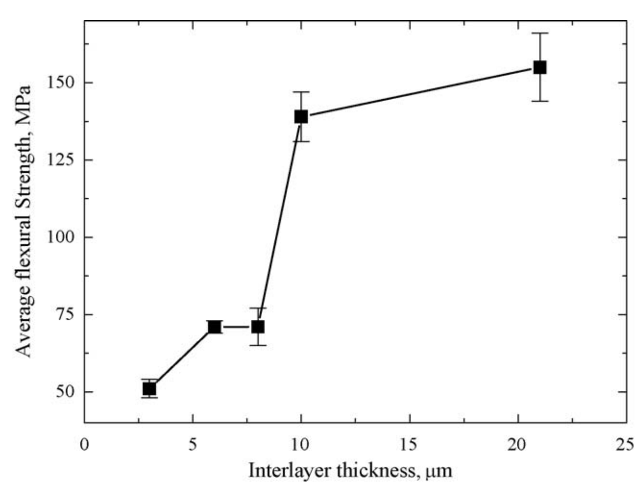

Fig. 8. Average flexural strength of the alumina joints TLP bonded at 800 °C for 15 h via an interlayer with various thicknesses.

peak for $3Al_2O_3–B_2O_3$ at 27.7° indicates that this metastable phase dominates at the alumina joint under that condition. $3Al_2O_3–B_2O_3$ disappears as the joint is heated for more than 15 h. Instead, $2Al_2O_3–B_2O_3$ and $9Al_2O_3–2B_2O_3$ appear in sequence, while the latter increases with joining time. The fact that the maximum strength occurs at the 15 h joining and that $9Al_2O_3–2B_2O_3$ appears in the joints heated for more than 24 h suggests that the formation of the $9Al_2O_3–2B_2O_3$ compound leads to the weakening of the joint.

Finally, the average flexural strength of the alumina joints TLP bonded at 800 °C for 15 h related to the interlayer thickness is drawn in Fig. 8. The strength increases with increasing thickness. The maximum strength obtained in this work is ca. 155 MPa while joining with a 21 μm boron oxide interlayer. The effect of interlayer thickness on flexural strength is consistent with the statements previously made by observing the structure of interfaces.

4. Conclusion

The transient liquid phase bonding via ceramic interlayers is proposed to join alumina in this work. Alumina pieces are joined at 800 °C in air with a boron oxide interlayer as a ceramic braze instead of metallic brazes. This technique has some advantages over the ‘‘traditional’’ processes using metallic interlayers, like moderate processing temperature, low material cost, no vacuum or reducing atmosphere needed. The interlayer thickness and joining time, which are crucial in the joining process, were varied to find the optimal joining condition. The cross-section and fractured surface of the alumina joints were observed by means of SEM, while the interfacial compounds were identified by XRD. The maximum flexural strength of the alumina joint reaches 155 MPa when joining with a 21 μm thick interlayer for 15 h. The sequence of formation of compounds at the interface proposed is $3Al_2O_3–B_2O_3$, $2Al_2O_3–B_2O_3$ and $9Al_2O_3–2B_2O_3$, where the first one is a metastable phase. The flexural strength is strongly influenced by the densification of the microstructure at joints in which compounds in the form of whiskers from. The optimal joint is mainly composed of $2Al_2O_3–B_2O_3$. When the $9Al_2O_3–2B_2O_3$ compound appears, after 15 h heat treatment, the flexural strength declines.

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