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Research on Reactive Brazed Connections of Steel/Ceramics

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Abstract: Ceramics are widely used in aerospace and industrial production due to their excellent mechanical properties. However, due to the shortcomings of ceramics, such as high hardness and brittleness, it is difficult to be processed into the shapes required for engineering applications, and it needs to be connected with metals to form ceramic/metal composites with complementary properties, and the selection of suitable connection technology becomes the key to determine the strength of ceramic/metal properties. And reactive brazing in the connection of ceramics and metal showed very superior mechanical properties. The active element can effectively wet the ceramic and form a reaction layer through diffusion, which enhances the stability and connection strength of ceramic/metal joints. At the same time, the active brazing material can efficiently relieve the residual thermal stress, which plays an important role in maintaining the integrity of the joint. This paper summarizes the current research status of ceramic/metal brazed joints at home and abroad in recent years, analyzes the microscopic morphology, interfacial structure and mechanical properties of the joints, and concludes that the joints are generally composed of the structure of metal base material + reaction layer + reactive brazing material + reaction layer + ceramic base material, of which the reaction layer depends on brazing material to a large extent. Finally, the problems of metal/ceramic in the joining process are summarized.

Keywords: Ceramics/Metal, Brazing, Complementary Properties

1. Introduction

With the continuous development of modern material science and technology, some common metallic materials have become difficult to meet the needs of people for mechanical production and manufacturing [1]. Some new requirements have been put forward for some materials, such as in special environmental conditions can still work properly. The metal materials commonly used today, steel, have excellent properties such as ductility, toughness, and ease of processing [2, 3], but their poor performance in terms of wear and corrosion resistance makes them subject to large losses in the actual use process. Ceramic materials have excellent properties such as corrosion resistance, wear resistance, oxidation resistance and high strength at high temperatures, but their poor ductility and more dispersive internal strength make it difficult to prepare structurally complex components [4, 5].

The composite components formed by joining metal steel and ceramic materials together can achieve a complementary state in terms of performance, which can integrate the respective advantages of the two materials, make up for the shortcomings of the two materials and broaden the scope of application of the two materials. Then, the technology of joining metallic and ceramic materials will become particularly important, and the selection of a suitable joining technology will result in reliable steel/ceramic structural members.

Engineering ceramics have many excellent physical and chemical properties, such as high hardness, high strength, high temperature resistance, corrosion resistance and abrasion resistance. Ceramics have important applications in aviation, energy, electrical, mechanical and optical fields [1-3]. However, ceramics have the characteristics of small ductility, large brittleness, and difficult processing, which cannot eliminate stress through plastic deformation in the using process. Therefore, it is easy to fracture and difficult to prepare large-scale complex ceramic components, which limits the engineering application of ceramics to a certain extent. Metal materials have good room temperature strength, electrical conductivity, thermal conductivity, excellent plasticity and toughness, and good machining performance, but poor mechanical performance at high temperature [4, 5]. For example, stainless steel (SS) is the most commonly used structural engineering material, which has excellent corrosion resistance, heat resistance, welding performance and polishing performance. It is easy to process and cheap, which is widely used in construction, food, chemical industry and medical fields.

Ceramics/steel composites have the potential for high fracture toughness, resistance to catastrophic failure, high strength, light weight and low thermal expansion, and they withstand high temperature with high oxidation resistance. These types of materials exhibit much greater resistance to high temperatures and aggressive environments than single steel or other conventional engineering materials.

2. Active Brazing

Active brazing is more widely used than inactive brazing because of the active metal elements in the brazing material added. The active metal elements inside the filler braze not only react metallurgically with the steel side under high temperature, but also diffuse to the surface of the ceramic and interact with each other, so that the braze in the molten state can extend better on the ceramic surface. Among them, Cu-Ti-based series of active brazing materials are widely used in various brazing applications because they can form TiCu metal compounds with certain extensibility at high temperatures [6]. Ou Ting et al [7] applied AgCuTi brazing material for vacuum brazing to join SiC ceramics with 316L stainless steel. As shown in Figure 1a, the brazed area was mainly composed of TiC, Ti_5Si_3, Ag(s,s), Cu(s,s), Fe-Ti, and Ti-Cu intermetallic compounds. The fracture occurred at the SiC ceramic and braze bond interface and the maximum shear strength intensity reached 32 MPa. L. M. Pan et al [8] performed vacuum brazing of Ti_3AlC_2 ceramics and steel using the active braze AgCuTi, as shown in Figure 1b. The results showed the presence of Ag and Cu solid solution as well as four metallic phases, $AlCu_2Ti$ and Al_4Cu_9, in the weld region. The fracture location was the ceramic/braze bond interface with a maximum shear strength of 196.4 MPa. L. X. Zhang et al [9] used AgCuTi braze to join SiO_2 glass-ceramics and 30Cr3 high-strength steel. As shown in Figure 1c, the experimental results showed the formation of $TiSi_2$, Ti_4O_7, Ti_2Cu, Cu_3Ti_3O, Ag(s.s), Cu(s.s), and $TiFe_2$ compounds in the central region of the joint, achieving a reliable connection between ceramics and steel with a maximum shear strength of 37 MPa in the joint. K. L Lin et al [10] used AgCuTi brazing material vacuum brazing to join stainless steel and yttria-stabilized ZrO_2 ceramics (YSZ), as shown in Figure 1d. It was found that Fe_2Ti compounds were formed at the steel/braze interface, titanium atoms combined with oxygen atoms of YSZ to form Ti_2O_3, and Fe diffused to the ceramic side and combined with Ti_2O_3 to further form Fe_2Ti_4O. The maximum shear strength of the joint reached 48.4 MPa.

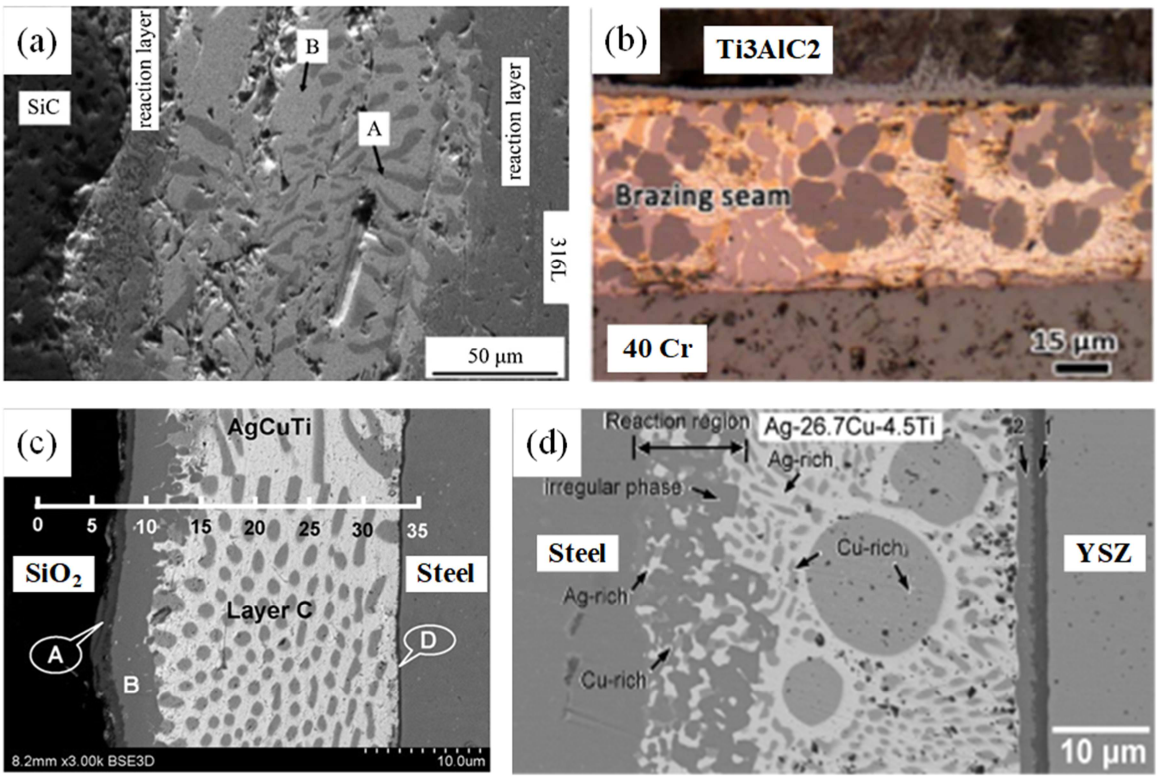

Figure 1. *Microscopic appearance of the joint: (a) SiC/AgCuTi/S316L. (b) Ti_3AlC_2/AgCuTi/40Cr. (c) SiO_2/AgCuTi/Steel. (d) Steel/AgCuTi/YSZ.*

In addition to conventional Ag-based brazing materials, there are some other types of active brazing materials, such as Cu-based, Ni-based, Sn-based, and Cu-Ti-based series brazing materials [11, 12], all of which have good wetting effects on ceramic surfaces. X. Y. Wang et al [13, 14] successfully realized the vacuum brazing connection of Al_2O_3 ceramics and Q235 steel by CuTi brazing materials. The experimental results showed that the solidified brazing material was closely connected to the base material on both sides, and the interface was mainly distinguished into liquid brazing material-ceramic reaction layer, Ti-Cu brazing region and steel side reaction layer. The interface mainly formed $AlCu_4$, Cu_3TiO_4, Cu_3Ti_3O, TiFe, $TiFe_2$ and Cu(s,s). The maximum shear strength of the joint reached 99.3 MPa. G. Blugan et al [15] successfully realized the vacuum brazing bonding of composite ceramics to steel by CuSnTiZr brazing material, and the bending strength at the joint was only 73 MPa. As shown in Figure 2a, the interfacial bond was densely organized and free of defects such as micropores, and mainly new phases of Fe_2Ti and Sn-Cu were formed. At the bonding interface, the reaction between titanium and iron produced a brittle Fe_2Ti phase and the generated residual thermal stresses were the factors of strength reduction. Subsequently, the addition of WC particles in the brazing region improved the weld organization and further enhanced the strength of the joint. T. H. Deng et al [16] used CnSnTiNi reactive brazing material for vacuum brazing of jointed Al_2O_3/CuSnTiNi/Cr12 steel. As shown in Figure 2b, the brazed weld was mainly divided into three regions: I, II and III, with Cu-Fe-Ti metal compounds generated in the I region near the ceramic, Cu_XTi_Y and Ti_XC_Y reactants and (Cu,Sn) solid solution generated in the intermediate II region, and $TiFe_2$ and TiC compounds formed in the III region on the steel side. The shear strength of the joint was 118 MPa by means of post-weld heat treatment.

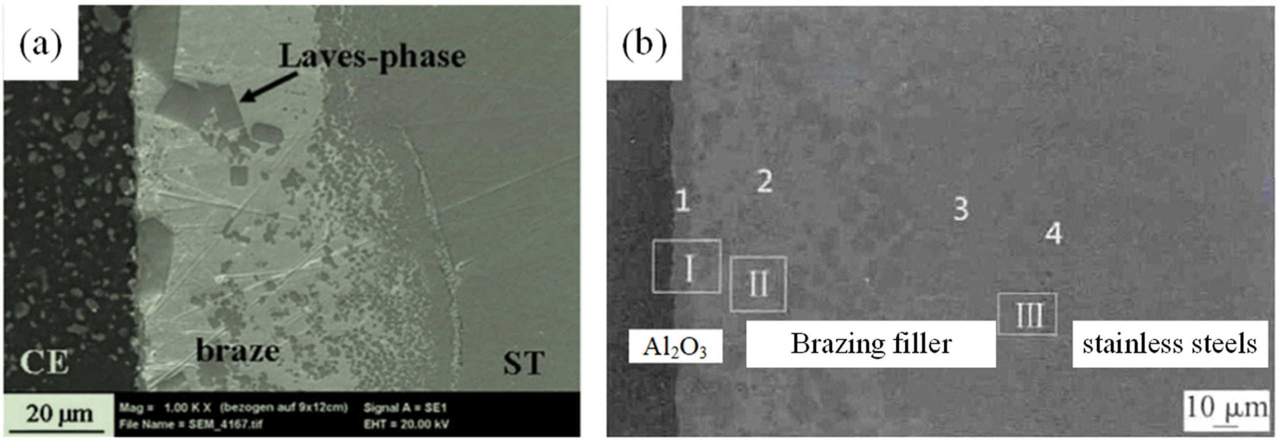

Figure 2. *Microscopic appearance of the joint: (a) Si_3N_4-TiN/CuSnTiZr/14NiCr14. (b) Al_2O_3/CuSnTiNi/Cr_{12}.*

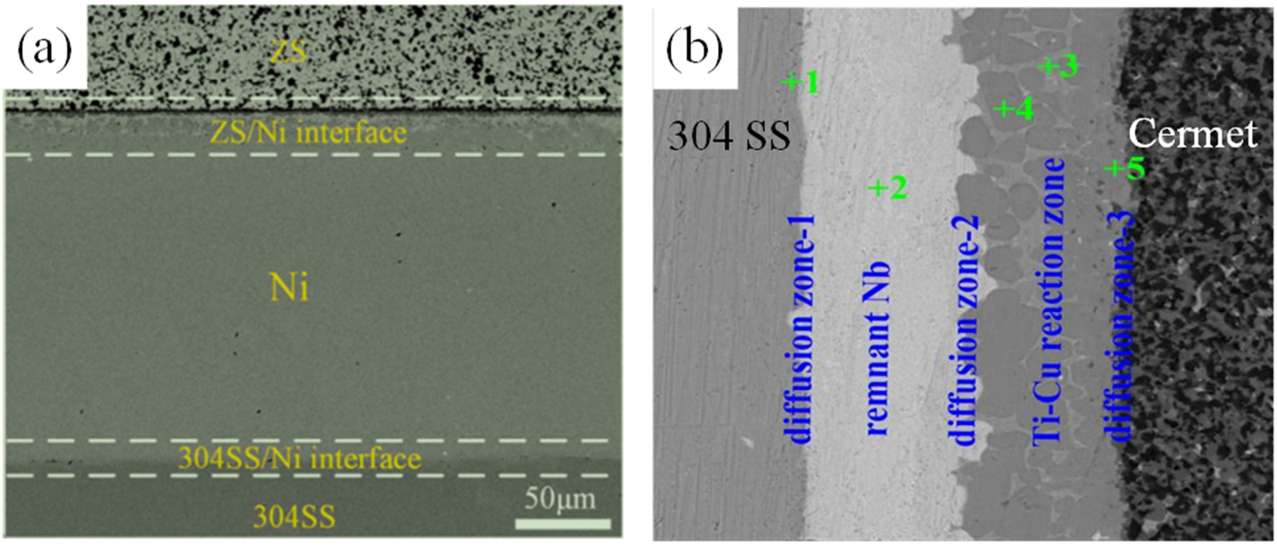

Figure 3. *Microstructure of joints: (a) ZrC-SiC/Ti/Ni/304SS. (b) TiC/Ti/Cu/Nb/304SS.*

W Tillmann et al [17] used Ni and CuNi brazing to join yttria-stabilized zirconia ceramics to Crofer22APU stainless steel. It was found that small amounts of Zr and Ti reactive elements deposited on the surface of the stainless steel promoted the brazing to wet the ceramic surface and form a good bond, as well as limiting the formation of some brittle

TiXOY and ZrXOY phases. J. C. Lin et al [18] used Ti/Ni composite metal foil for diffusion joining of composite ceramics and stainless steel, as shown in Figure 3a. It was shown that Ti_2Ni, Ti_3Ni_2Si, $TiNi_3$ and Ni_5Zr phases were formed in the weld region. The joints fractured along the $TiNi_3$/Ni boundary with a maximum shear strength of 107 MPa. Li Jia et al [19, 20] connected TiC cermets and 304 stainless steel by partial transient liquid-phase diffusion of Ti/Cu/Nb multilayer interlayer. As shown in Figure 3b, there was a clear reaction layer formation at the interface and the brazing material was tightly connected to the base material on both sides. In the process of joint forming CuTi, $CuTi_2$ intermetallic compound, the maximum shear strength of the joint was 106.7 MPa.

3. Conclusion

As can be seen above, the inclusion of reactive elements in the brazing material effectively solves the problem of ceramic wettability, and the joining of ceramics to metals can be achieved by reactive brazing. However, the reaction at the brazing interface is complex and brittle intermetallic compounds are formed in the joints along with the formation of the reaction layer required for joining, further in-depth study and improvement of the joints' properties are still needed.

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