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Laser joining of Al_2O_3 liners with Al_2O_3–MgO–SiO_2 glass-ceramic fillers

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ABSTRACT

The influence of fillers composition on microstructure, the physical and chemical properties of joints is studied. Thermal mismatch between fillers and Al_2O_3 liners during laser joining process could be effectively reduced with the increase of MgO content. The mutual diffusion and the substitution reaction lead to the formation of the eutectic spinel phase (Mg, Fe)Al_2O_4. The interfacial atomic diffusion is enhanced by the addition of MgO. The joints corresponding to fillers composed of 70 wt.% Al_2O_3-15 wt.% MgO-15 wt.% SiO_2 have the optimal mechanical performance. The average flexural strength of joints reaches 168 Mpa which is 80% of the Al_2O_3 liner (200Mpa). The weight loss of the joint interlayer in 10 wt.% HCl (pH = 0.45) for 24 h is 0.62 mg/cm^2 which is 0.16 mg/cm^2 lower than that of the liner. The softening temperature of joint interlayer exceeds 1500 °C. The reliability of Al_2O_3-lined joints could be maintained during the sequential process of arc welding for outer steel pipe layers. The corrosion of pipe layers could be effectively avoided.

1. Introduction

The alumina-lined composite pipes produced through self-propagating high-temperature synthesis consist of α-Al_2O_3, transition layer and steel from inside to outside respectively. Owing to the excellent wear resistance and corrosion resistance, the alumina-lined composite pipes are widely used in many fields such as electricity, petrochemical industry, metallurgy and chemical industries as Konegger et al. (2014) described. For example, Le et al. (2008) prepared ceramic-lined steel pipe by the thermit reaction of aluminium-ferric oxide under the centrifugal force. And it is an attractive candidate to replace traditional steel pipes used for oil and gas transportation, which might increase their service lifetime. However, it is difficult to obtain reliable bonding of the alumina-lined composite pipes. King et al. (2014) have realized metallurgical bonding between the steel pipe layers by traditional electric arc welding, but not for the alumina liners. In this case, corrosive media would corrode the steel pipe through the gap of Al_2O_3 liners. Therefore, it is essential to obtain a strong and refractory bonding between the Al_2O_3-lined layers before electric arc welding.

Several methods have been developed to join ceramics, such as diffusing bonding, active brazing and glass-ceramic joining. Ti_3AlC_2 ceramics were joined through diffusion bonding using Ti/Ni interlayer by Cao et al. (2014). Esposito et al. (1998) successfully applied diffusing bonding to join alumina with thin metal sheets of Ni, Cu and Fe. Moreover, Fernandez et al. (2016) achieved the active metal brazing of silicon nitride ceramics using a Cu-based alloy and refractory metal interlayers. Asthana and Singh (2008) realized the joining of polycrystalline alumina to itself and to Ti, hastealloy and a CVI C–SiC composite with two Ag–Cu active metal brazes containing Ti. However, each joining technology has its own limitations. For example, diffusion bonding requires either high temperature or high pressure, which is not conducive for the bonding of fragile ceramics. Active brazing needs to be conducted in vacuum or inert atmosphere environment and the corrosion resistance of the joint is not sufficient for acid environment applications as reported by Gozzelino et al. (2016). However, the silicate glass-ceramics fillers have been successfully utilized to join ceramics for corrosion resistance and high temperature applications, due to their excellent chemical durability and wettability. Zhou (2002) proposed a method for the joining of silicon nitride composites with Y_2O_3–Al_2O_3–SiO_2 mixture. Herrmann et al. (2014) succeed in the creation of homogeneously SiC ceramic joint with Y_2O_3–Al_2O_3–SiO_2 filler. Ahmada et al. (2016) realized the joining of SiC by the utilization of Nd_2O_3–Al_2O_3–SiO_2 glass solder. Several aluminosilicate glass-ceramic systems have been employed as fillers to join Al_2O_3 ceramics. Börner et al. (2010) developed the SiO_2–Al_2O_3–B_2O_3-MeO system, and the glass-ceramic joint were free from defects with the average strength value of 158 Mpa. Zhu et al. (2013) developed the CaO-Al_2O_3–SiO_2 glass for the joining of porous alumina and the joint possessed excellent thermal shock resistance. Subsequently, Zhu et al. (2014) used the ZnO–Al_2O_3–B_2O_3–SiO_2 glass ceramic to join alumina ceramics, and the optimum joint strength reached 285 Mpa which was the same as the base material strength. However, the melting temperature of the steel

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pipe layer (~1500 °C) is much higher than that of these fillers (< 1250 °C), which may cause the resoftening of the interlayer during the process of the electric arc welding. Therefore, it is strongly necessary to develop a new glass ceramic filler with high thermal resistance.

Lee et al. (1998) realized the joining of silicon carbide with Al_2O_3–MgO–SiO_2 filler above 1500 °C in Ar atmosphere. Zhang et al. (2011) analyzed the bonding layer microstructure and their effects on fracture behaviors of sapphire joint with Al_2O_3–MgO–SiO_2 filler. It is noteworthy that the Al_2O_3–MgO–SiO_2 (AMS) glass ceramics is highly refractoriness. In addition, a small amount of FeO exists in the Al_2O_3-lined layers due to incomplete thermit reaction. According to the isomorphism theory, the Mg^{2+} ions and the Fe^{2+} ions can replace mutually and form eutectic system as Mao et al. (2005) mentioned. Therefore, the MgO in fillers could contribute to the mutual diffusion and substitution between the Mg^{2+} ions from filler and the Fe^{2+} ions from Al_2O_3-lined layers. Thus, the AMS glass-ceramic filler is promising for high temperature joining application.

Conventional joining process is completed in a furnace and takes hours. On the contrary, laser radiation locally heats the joining zone instead of the global. Processing time is thus considerably shorter, and the joining of ceramic can be achieved without damage to the bilateral Al_2O_3 liners. Furthermore, alumina-lined composite pipes with a diameter of meters and the length of kilometers obviously cannot be joined in a furnace. While there is no component size restriction for laser joining. Experimental selection processes have shown that a diode laser in the wavelength range of 808–1010 nm is well-suited for joining both non-oxide and oxide ceramics. Oxide ceramics such as ZrO_2 are partially transparent for the specific wavelength of diode lasers. This results in absorption of laser radiation as well as thermal energy into the volume of the ceramic body. And it is the reason why oxide ceramics, despite having low thermal conductivities, can be heated using laser radiation without excessive thermal stress as mentioned by Börner et al. (2014). Hence, diode laser welding was adopted in our experiments. In previous works, fillers of thin disk or pastes of slurry were used to join alumina ceramics. However, with rapid heating and cooling process under laser radiation, fragile lined layers crack easily due to the thermal shock. Thus, Bar shaped fillers are used to match the laser spot and prevent absorption by liners.

In this paper, Al_2O_3–MgO–SiO_2 glass-ceramics (AMS) are developed as the fillers to join the Al_2O_3 liners of ceramic-metal composite pipes by using laser welding technology. The crystalline phases, microstructures and atomic diffusion in the interfacial regions are investigated, and the mechanism of affection on the atomic diffusion process by MgO addition is analyzed. In addition, the effects of the MgO content on the quality of joints are studied in detail.

2. Experiment

2.1. Preparation and characterization of liners and fillers

The Al_2O_3 liners (provided by the Green Aus Energy Technology Co., Ltd, Beijing, China) were peeled off from the ceramic-metal composite pipes and were cut into bar-shaped samples with a dimension of 4 mm × 10 mm × 20 mm by a CNC scribing cutting machine. The fillers consisted of α-Al_2O_3, SiO_2, MgO (99.99% purity, from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China). The chemical compositions of fillers are listed in Table 1. The powders were ball-milled for 6 h at 400 rpm in a ball mill. Subsequently, the powder mixtures were dried for 4 h at 80 °C in an automatically controlled drying oven. Furthermore, the powders were pressed into pellets with a dimension of 4 mm × 10 mm × 3 mm. Then, the samples were pre-sintered at 1400 °C for 3 h in muffle furnace. The microstructure and elements distribution of the liners and fillers were determined by scan electron microscopy (SEM) and energy dispersive spectroscopy (EDS). The crystalline phases of liners and fillers were identified by an X-ray diffractometer (7000 S/L, Shimadzu Corp., Japan). The laser absorbance of the filler was measured by a spectrophotometer (Lambda 35). Furthermore, the thermal expansion behavior of AMS fillers and Al_2O_3 liners should be measured using a thermal dilatometer.

Table 1
Chemical composition and sintering temperature of the fillers.

Fillers	Chemical composition (wt.%)			Sintering temperature(°C)
	Al_2O_3	SiO_2	MgO	
1	70	30	0	1400
2	70	25	5	1400
3	70	20	10	1400
4	70	15	15	1400
5	70	10	20	1400

2.2. Wetting experiment

Five groups of 3 mm × 3 mm × 3 mm fillers as shown in Table 1 were placed on the polished surface of Al_2O_3 liners respectively. Fillers were at the focus of a semiconductor laser with the spot diameter of 3 mm. Laser radiation with the power of 500 W lasted only for 1 s. A computerized image analysis system was employed to record samples geometry and calculate the contact angle after samples were completely cooled.

2.3. Laser joining procedure

A semiconductor laser with maximum power of 3.0 kW and wavelength of 980 ± 10 nm was used to join the Al_2O_3-lined layers. The samples were placed at the focus of lens which is 370 mm under the lens. The power and facula-moving speed of the laser were 700 W and 1 mm/s, respectively. The scanning path of the laser is shown in Fig. 1.

2.4. Testing method of the joint

The microstructure, phase composition and elements distribution of joints were measured by SEM, XRD and EDS respectively. The flexural strength of joints was evaluated by the three-point-bending test with the span of 30 mm by electronic universal testing machine (CMT4104, Meitesi Industry Co. Ltd, China). The samples were cut into bars of 4 mm × 5 mm × 36 mm for bending tests and six samples were used to determine the average value of joints strength. The flexural strength was calculated according to the following Eq. (1):

$$\sigma_B = \frac{3LF}{2b^2d} \quad (1)$$

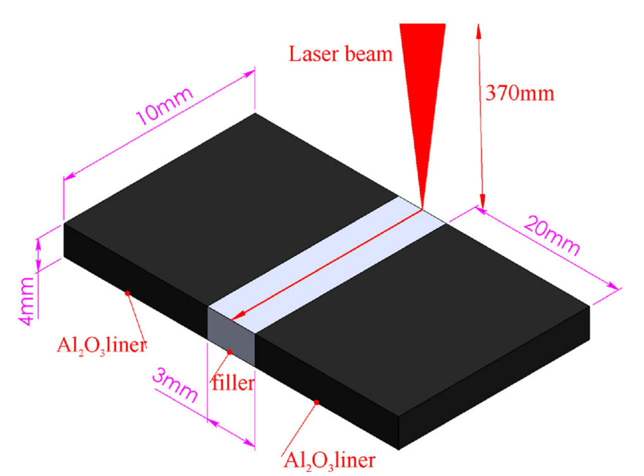

Fig. 1. Schematic diagram of the laser joining process.