

Optimization on the diode laser joining process of Al₂O₃ liners

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Abstract. The effects of fillers width and energy density of laser on weld morphology were studied. The mullite filler with the quality ratio of 70wt.% Al₂O₃-30wt.% SiO₂ was determined as the initial fillers for the joining of Al₂O₃ liners according to the crystalline phase of Al₂O₃ liners. The transient multiple physical model of diode laser joining Al₂O₃ ceramic liners was built by Multiphysics COMSOL, and the coupling of temperature and stress field was carried out. The thermodynamic properties of the Al₂O₃ liners and fillers were analyzed and tested. Numerical results showed that the increase of fillers width would effectively reduce the thermal stress of the joint. With the increase of the energy density, the temperature and thermal stress of the seam zone increased gradually. With the increase of laser joining heat input, weld penetration increased slowly and then sharply. The experimental results indicated that the weld zone free from pores and macro cracks can be formed with the fillers width of 3mm, laser power of 700W and welding speed of 1mm/s.

Keywords: diode laser, ceramic liners, weld penetration, thermodynamic properties, morphology of the weld, thermal stress.

1. Introduction

On account of the reliable and excellent physical and chemical properties in high temperature environment, such as high hardness, oxidation resistance, wear resistance, etc., alumina ceramic is of incomparable application value in aerospace, oil and gas transportation pipe, high temperature reactor and other fields [1-2]. Fluid media usually contains H₂S, CO₂, Cl⁻ and other corrosive media components in the oil and gas industry, forming different acidic environments in the transportation process [3-4]. Owing to the excellent wear resistance and corrosion resistance, the alumina-lined composite pipes are widely used in petrochemical industry as Konegger et al. (2005) described [5]. And it is an attractive candidate to replace traditional steel pipes used for oil and gas transportation, in order to increase their service lifespan^[6]. Because of the high melting point and relatively large size of ceramic liners, the traditional joining methods restricted by temperature and size are not suitable for ceramic joining. In addition, it is difficult to process ceramics due to its poor plasticity and great brittleness. The traditional metallurgical joining method is not competent for the acidic application environment of ceramic liners. For example, diffusion bonding requires either high temperature or high pressure [7]. 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 application [8]. Therefore, it is essential to obtain a reliable joint between the Al₂O₃ liners before electric arc welding.

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 [9-13]. To achieve this, a time-consuming conventional joining process is conducted in a furnace. On the contrary, laser radiation can heat the joining zone locally without heating the whole to the melting temperature[14-15], resulting in shorter processing time and no damage to the bilateral Al₂O₃ liners during ceramic joining. Furthermore, there is no component size restriction imposed by the internal dimensions of a sintering furnace. Previous works have shown that the diode laser in wavelength range of 808-1010nm is very well-suited for joining oxide ceramics [16-18]. Oxide ceramics 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 s [19]. Hence, the diode laser

joining was adopted in our experimental works. During laser joining process, excessive temperature gradient may cause cracking of the weld. Therefore, it is particularly important to optimize the processing parameters of laser joining process. For example, fillers of thin disk or pastes of slurry were used to join alumina ceramics in previous studies [20-21]. The capillary force of the molten pool can be fully utilized by using the thin disk to form a compact connection between the joining parts [22]. However, with rapid heating and cooling process under laser radiation, fragile ceramics would crack easily due to the thermal shock. With the increase of fillers width, the laser radiation could be totally irradiated on fillers instead of Al_2O_3 liners, which could help to avoid the crack of liners [23]. Besides, the temperature field and stress field during the joining process can be simulated numerically to analyze the mechanism of thermal cracking. In previous studies, a series of numerical calculations based on COMSOL have been carried out to predict the temperature and stress field during the interaction between the laser and joining parts [24-26]. In this paper, bar-shaped fillers were used for the joining of Al_2O_3 liners. Experimental and theoretical analysis based on COMSOL were carried out to optimize the process parameters during the joining of ceramic liners.

2. Experiment procedure

2.1. Preparation and characterization of the liner and filler

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 5 mm×10 mm×20 mm by a CNC scribing cutting machine. The fillers consisted of α - Al_2O_3 , SiO_2 (99.99 % purity, from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China). The powders were pressed into pellets with a dimension of 5 mm×10 mm×x mm (the thickness of the fillers was set as 5mm). Then, the samples were pre-sintered at 1400°C for 3 h in muffle furnace. The microstructure and elements distribution of the liners and were performed by scan electron microscopy (SEM) and energy dispersive spectroscopy (EDS).

As shown in Fig. 1(a), the crystalline phases of the liner are Al_2O_3 , SiO_2 , $\text{Al}_{82}\text{Fe}_{18}$ and a few $\text{FeO}\cdot\text{Al}_2\text{O}_3$. Fig. 1(b) shows the microstructures of the liner, which are mainly composed of dark gray strip-like section (marked as A) and light gray strip-like section (marked as B). From the EDS patterns, it can be concluded that the zone A is Al_2O_3 and the zone B contains Al_2O_3 , FeAl_2O_4 and SiO_2 [29]. The researchers show that mullite phase possesses high melting point, high corrosion resistance, creep resistance, relatively low expansion coefficient and other characteristics [27]. Therefore, the glass-ceramic fillers with the mass ratio of 70wt.% Al_2O_3 -30wt.% SiO_2 were selected as fillers for the laser joining experiment.

2.2. Laser joining procedure

Fig. 1 shows the diode laser experimental joining system, which includes a 3KW DILAS didod laser generator, a beam shaper, bump power control system, a six-axis ABB welding robot and a water cooler of laser generator. The laser with wavelength 980 ± 10 nm was used to join the Al_2O_3 liners, its output is controlled by the CNC panel of ABB robot.

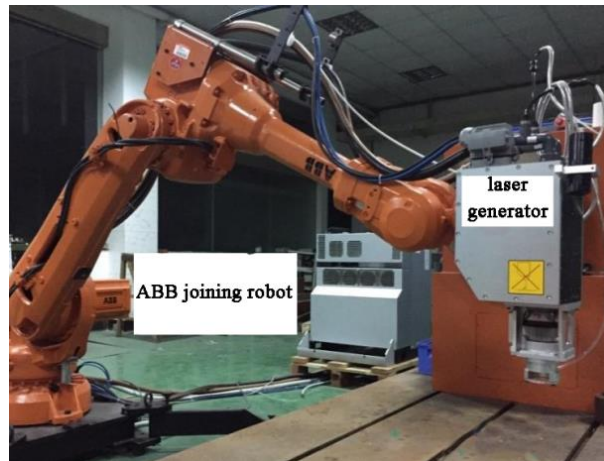


Figure 1. Experimental joining system of diode laser

Fig 2. shows the shape of the laser beam. When there was no defocusing of the laser spot, the output beam of the diode laser is approximately a Gaussian spot.

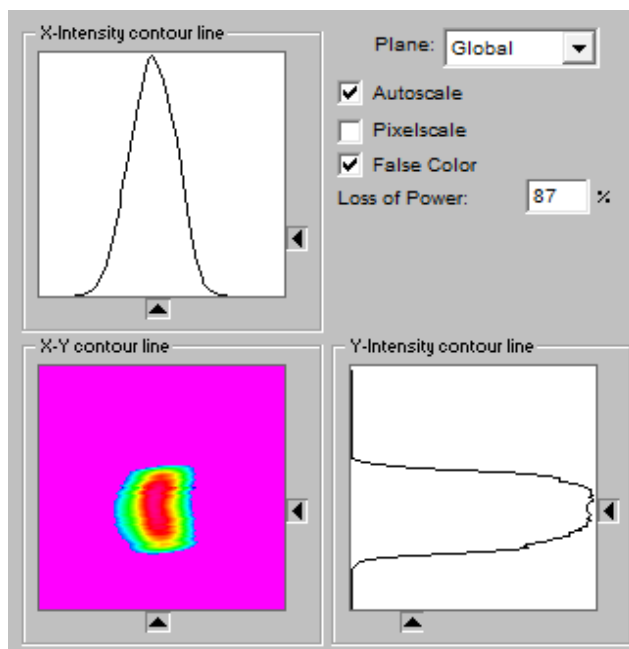


Figure 2. Diode laser generator and Gaussian beam

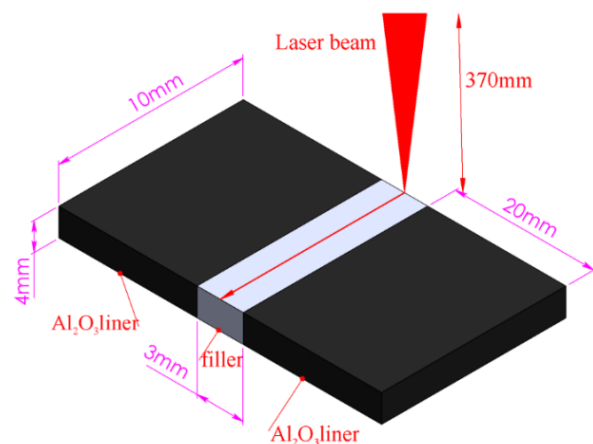


Figure 3. Laser joining of the Al_2O_3 liners with mullite ceramic fillers

Fig.3 is the schematic diagram of the laser joining of the Al_2O_3 liners with ceramic fillers. Compressed air was used in the experiment to prevent the mirror of beam shaper from any damage caused by the splash and vaporization of melted pool of sample due to the instantaneous shock of laser. The actual measured gap of ceramic liners was about 1mm. Considering the integrity of the gap filling, fillers width should be no less than 1 mm. The laser spot diameter was 3 mm at zero defocusing position, the fillers width was divided into 6 groups from 1mm to 3.5mm with the span of 0.5mm. The power of the diode laser was divided into six groups from 400W to 900W with the span of 100W. The scanning speed of was divided into six groups from 0.4mm/s-1.4mm/s with the span of 0.2mm/s. The joining process was carried out according to the process parameters in this range.

2.3. Joining quality assessment

The laser joining quality evaluation was revolved around penetration depth, macro crack and porosity etc. During the laser joining process, the weld zone absorbs laser energy and gets molten. The difference of laser power density would lead to different penetration depth. If the joint depth

reached to 5 mm, it was considered that the weld zone achieved full penetration under the laser radiation.

3. Numerical simulation

The simulation of semiconductor laser joining Al_2O_3 ceramic liners was established based on COMSOL, and the interaction mechanism of laser radiation on materials was analyzed during the process of laser joining ceramics. Considering that the depth to width ratio in the experimental joining process was close to 1:1, the laser joining ceramics model was set as thermal conduction joining. Because of the single scanning during laser joining, the temperature field stress field model of laser welding in this paper was chosen as "transient" study. The temperature field and stress field of the laser joining model based on COMSOL analysis were used to analyze the cracks and the welding penetration in the ceramic substrate during the experiment.

The laser beam was assumed to be the Gaussian beam, the heat flux of the Gaussian beam in solid heat transfer module was set as follows:

$$Q_1 = \frac{2P_0}{\pi r_0^2} \cdot e^{-\frac{2r_{focus}^2}{r_0^2}} \quad (1)$$

Where P_0 is the average illumination of the incident Gaussian beam, r_0 is the radius of the laser spot, and r_{focus} is the distance from any point to the center of the laser spot

$$r_{focus} = \sqrt{(x - x_{focus})^2 + (y - y_{focus})^2} \quad (2)$$

Where $x_{focus}=0$ was the x coordinate of the laser spot center, and $y_{focus}=u \cdot t - 0.005$ was the y coordinate of the spot center.

The heat exchange mode of the sample itself was set as the transient heat conduction in the solid state. The heat conduction equation is:

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p u \cdot \nabla T + \nabla q = Q \quad (3)$$

$$q = -k \nabla T \quad (4)$$

where ρ was the density of material (kg/m^3), C_p was heat capacity at ordinary pressure [$\text{J}/(\text{kg} \cdot \text{K})$], u is scanning speed of the diode laser, Q was the heat source of the diode laser (W/m^2), k was the coefficient of heat conduction of the samples.

The heat exchange course between the surface of this model and the outside was simplified into heat convection in the air.

$$Q_2 = H \cdot (T_{ext} - T) \quad (5)$$

Where T_{ext} is the external temperature, namely room temperature, which is 293.15K, H is the convective heat transfer coefficient between the surface and the environment, and the unit is $\text{W}/(\text{m}^2 \cdot \text{K})$.

Considering that the temperature field of weld zone could cause dramatic changes over time under the initial period of laser radiation, the temperature field of this time cannot reveal the actual joining process suitably. The temperature of whole samples would rise by heat conduction and the temperature field welding zone under the irradiation of laser would be gradually stabilized with the

laser scanning process. In this model, the temperature field and stress field was calculated when the laser spot center moved to the end edge of fillers.

4. Result and discussion

4.1. Numerical studies on joint quality with different joining processes parameters

The binary phase diagram of Al_2O_3 - SiO_2 is shown in Fig. 4 [28]. According to Fig. 5, the crystalline phase of 70wt.% Al_2O_3 -30wt.% SiO_2 fillers after sintering was in the mullite phase region. Therefore, in the subsequent temperature simulation, the fillers was considered to be molten when the temperature exceeded 1650°C. The laser absorbance of the fillers under solid state was measured about 0.15. Given the dramatic rise of the laser welding penetration with 700W laser power, it could be concluded that the laser absorbance of the fillers was relatively high. In this paper, the laser absorbance of the fillers was set to be 0.7.

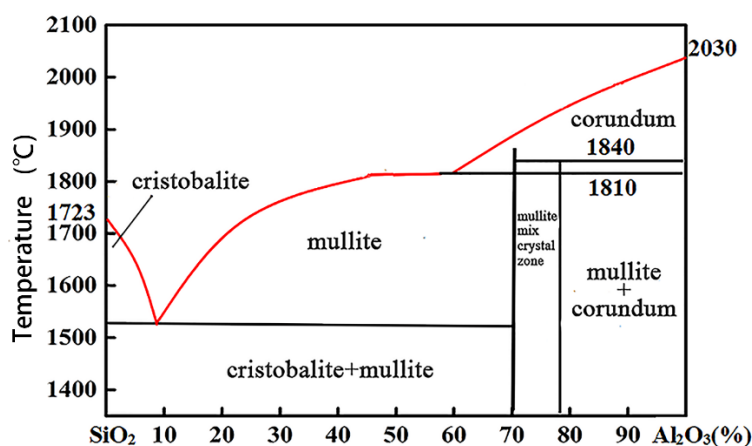


Figure 4. SiO_2 - MgO binary phase diagram

The mechanical properties of the Al_2O_3 liners were tested. Fig. 5 demonstrates the elasticity modulus of Al_2O_3 liners. Fig. 6 shows that the elasticity modulus of Al_2O_3 liners is about 80Gpa, while the defect-free Al_2O_3 liners is about 390Gpa [30]. This can be explained by the SHS process of Al_2O_3 liners, which would lead to the porous liners.

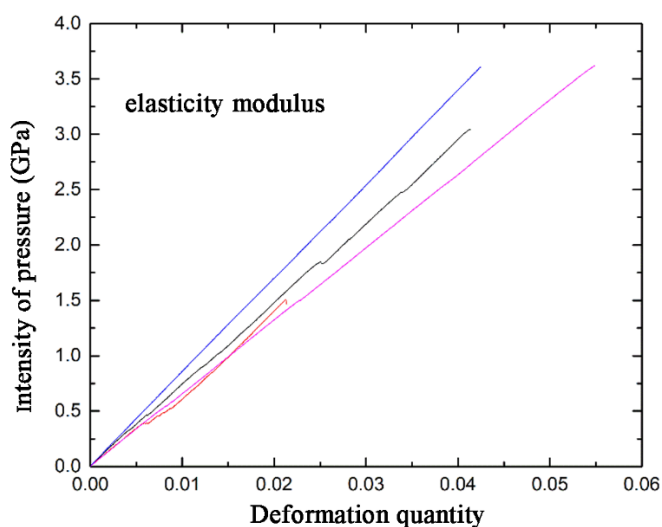


Figure 5. The elasticity modulus of Al_2O_3 liners

4.1.1. The effect of fillers width on the stress field and temperature field of the laser joining model

In this model, the power and the scanning speed of the diode laser was set as 700W and 1mm/s respectively. The temperature filed of the samples with different fillers width were demonstrated in

Fig. 6, where Figs. 6(a)-(f) represented the fillers, width ranging from 1mm-3.5mm. Fig. 7 shows the temperature distribution of surface of fillers at the laser spot center after 10s, where (a)-(f) represented the fillers width of 1mm-3.5mm, respectively. The arc length represents the fillers width in Fig. 8. As can be seen from Fig. 7, the undersurface of the fillers with width of 1mm-3.5mm can be completely melted. Moreover, with the increase of the filler's width, the temperature distribution along the upper edge tended to be symmetrical, and the temperature along the upper edge gradually decreased, which indicated that the increase of fillers width was helpful to alleviate the excessive thermal impact at the interface between the Al_2O_3 liners and the fillers. The difference of temperature profile for Fig. 7(e)-(f) from (a)-(d) could be explained by the increase of fillers width. Especially when the fillers width was 3.5mm, the edge of fillers failed to absorb the energy of the laser directly, and the temperature of the upper edge could decrease sharply. However, penetration depth was an important factor affecting the quality of joint, and it was necessary to calculate the temperature distribution of the undersurface to judge whether the penetration depth meets the requirements.

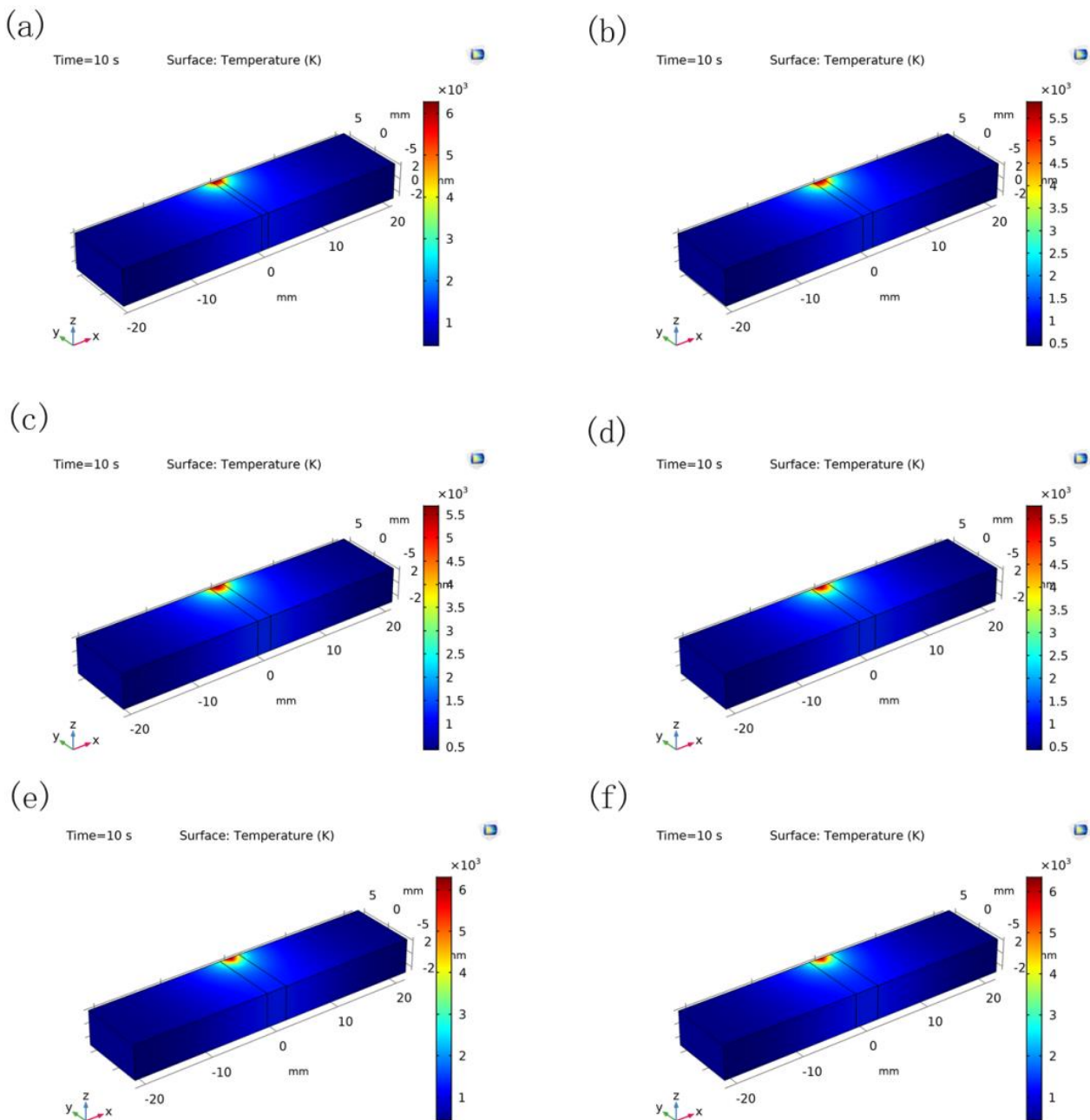


Figure 6. The temperature field of the samples with different fillers width; (a) 1mm; (b) 1.5mm; (c) 2mm; (d) 2.5mm; (e) 3mm; (f) 3.5mm.

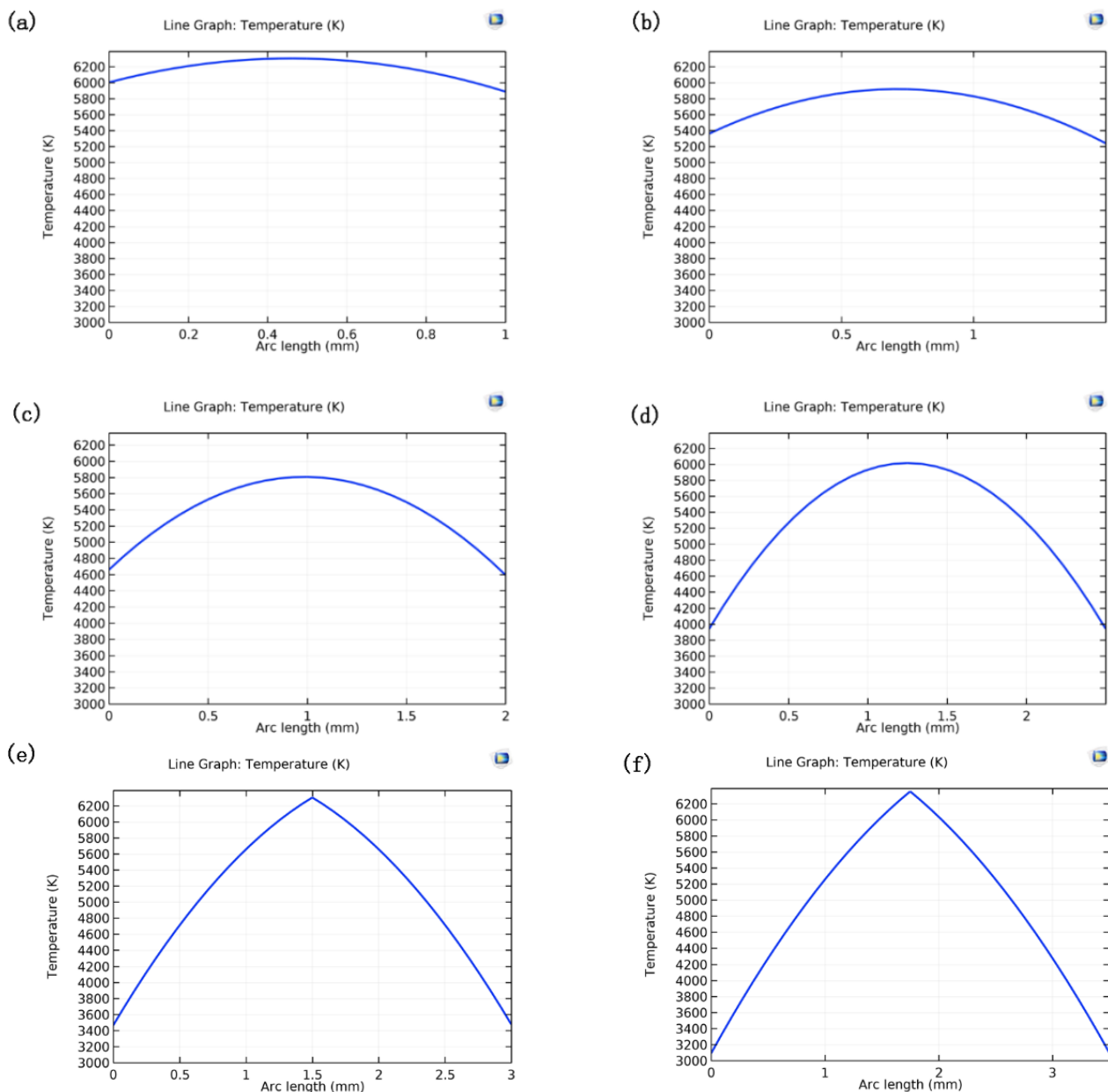


Figure 7. The temperature distribution of surface of fillers at the laser spot center with different fillers width; (a) 1mm; (b) 1.5mm; (c) 2mm; (d) 2.5mm; (e) 3mm; (f) 3.5mm.

Figs. 8(a)-(f) represent the temperature distribution of fillers undersurface at the laser spot center after 10s, where (a)-(f) represents the fillers width of 1mm-3.5mm. The arc length represents the fillers width in Fig. 8. As can be seen from Figs. 8(a)-(b), under the laser joining process parameters of 700W, 1mm/s, the highest temperature of the undersurface cannot reach the melting point of the fillers when the fillers width was 1mm and 1.5mm, and the penetration depth of the joint was relatively shallow. As can be seen from Fig. 8, with the increase of fillers width, the maximum temperature of the undersurface firstly increased and then decreased. When the fillers width was 2mm-3.5mm, the maximum temperature of the undersurface exceeded the melting point of the fillers. When the fillers width was 2.5mm and 3mm, the maximum temperature of the undersurface exceeded 1925K, meanwhile the average temperature of the undersurface was relatively high. On condition that the laser absorbance of the fillers remained unchanged, the increase of the filler's width would be beneficial for the laser absorption of the fillers. When the fillers width was 2-3.5mm, considering that the stirring movement of molten pool during the actual process of laser joining would promote the heat conduction, it can be assumed that the actual temperature of the undersurface should be higher than that of the simulation calculation results. On the condition that the upper surface was

complete melted, the wider molten pool due to the wider fillers was conducive to heat conduction. As a result, the penetration depth for those groups of samples could be considered as 5mm in the actual laser joining process.

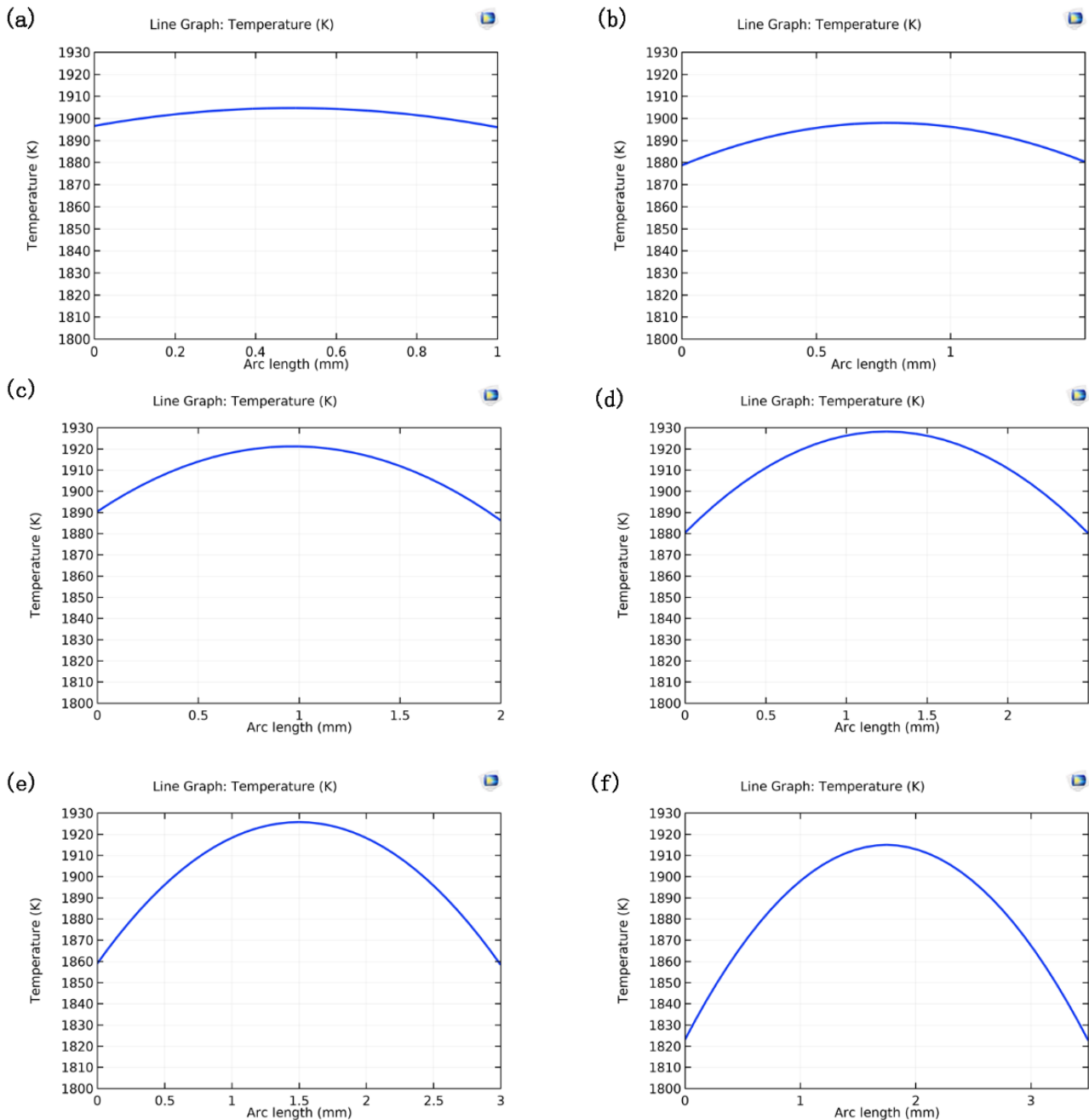


Figure 8. The temperature distribution of undersurface of fillers with different fillers width; (a) 1mm; (b) 1.5mm; (c) 2mm; (d) 2.5mm; (e) 3mm; (f) 3.5mm.

Figs. 9(a)-(d) represent the radial distribution of stress field of the Al_2O_3 liners adjacent to the laser spot after 10s laser scanning process when the fillers width was 2mm-3.5mm. The arc length represents the width of liners in Fig. 9. As shown in Fig. 9, with the increase of fillers width, the thermal stress of ceramic liners near the joining zone decreased gradually. The stress concentration at the interface between fillers and liners was mainly caused by the thermal expansion coefficient mismatch between fillers and liners. The residual thermal stress would weaken the joint quality. When the fillers width was 3mm and 3.5mm, the thermal stress at the interface significantly decreased. As can be seen from Figs. 8(e), the penetration depth, and the utilization rate of laser energy when the fillers width was 3mm is greater than that when the fillers width was 3.5mm. As a result, 3mm width of the fillers is preferred.

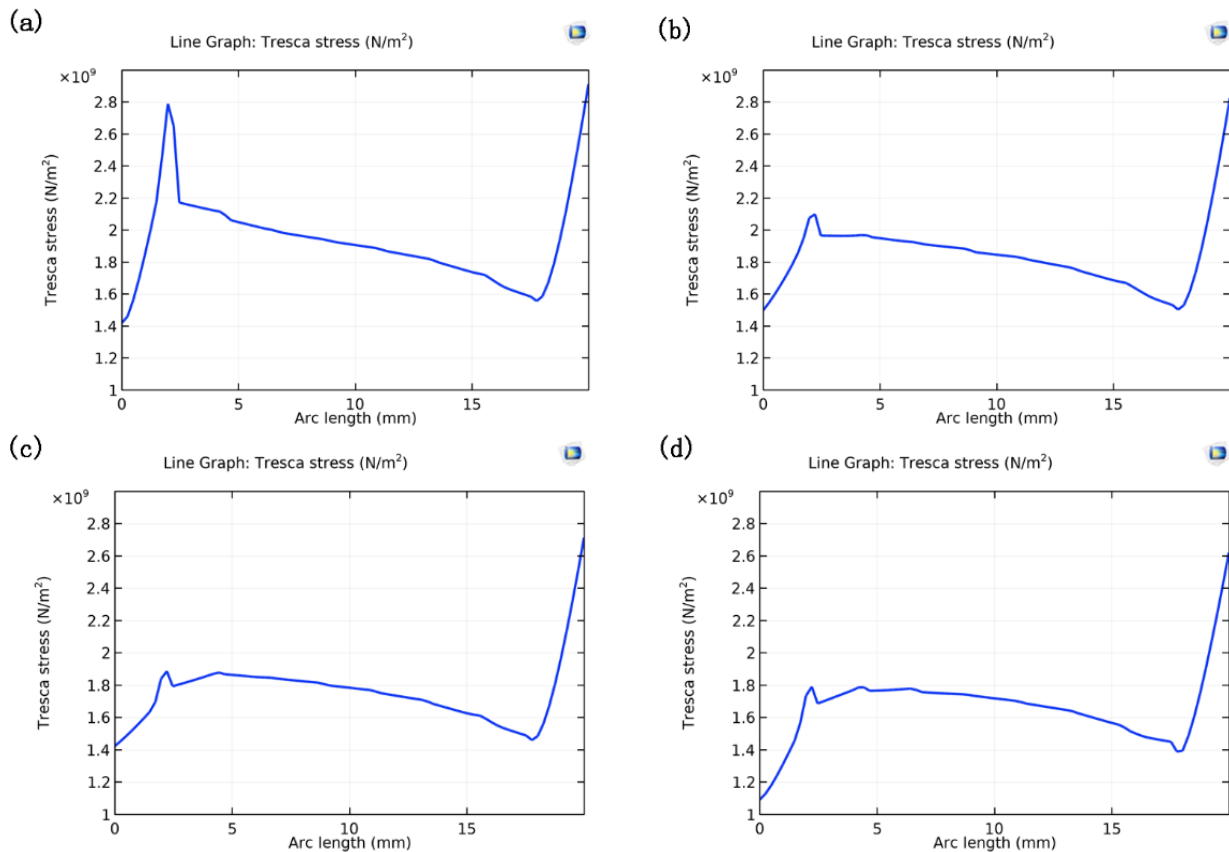


Figure 9. The stress field along the width of the Al_2O_3 liners adjacent to the laser spot with different fillers width; (a) 2mm; (b) 2.5mm; (c) 3mm; (d) 3.5mm.

4.1.2. The effect of laser energy density on the stress field and temperature field of the models

On the premise that the fillers width and the laser absorbance of the samples remain unchanged, the changes of laser power and scanning speed ultimately affect the laser energy density on the samples. In this model, the fillers width and the scanning speed of the laser was set as 3mm and 1mm/s, respectively. The influence of laser radiation on joining quality was illustrated.

Fig. 10 showed the temperature distribution along the thickness of the fillers (5mm) at the laser spot center after 10s, where (a)-(f) represented the laser power of 400W-900W respectively. The arc length represents the fillers thickness in Fig. 10. As can be seen from Fig. 10, with the increase of laser power, the penetration depth in the joining process increased gradually. When the laser power was 400W-600W, the penetration depth could not reach to 5mm of fillers thickness. When the power of laser reached to 700W, the minimum temperature of the fillers underside where the laser spot located was about 1900K, which meant that the penetration depth reached 5mm. Therefore, the joining thermal stress distribution of the weld zone was calculated when the laser power was 700W, 800W and 900W respectively.

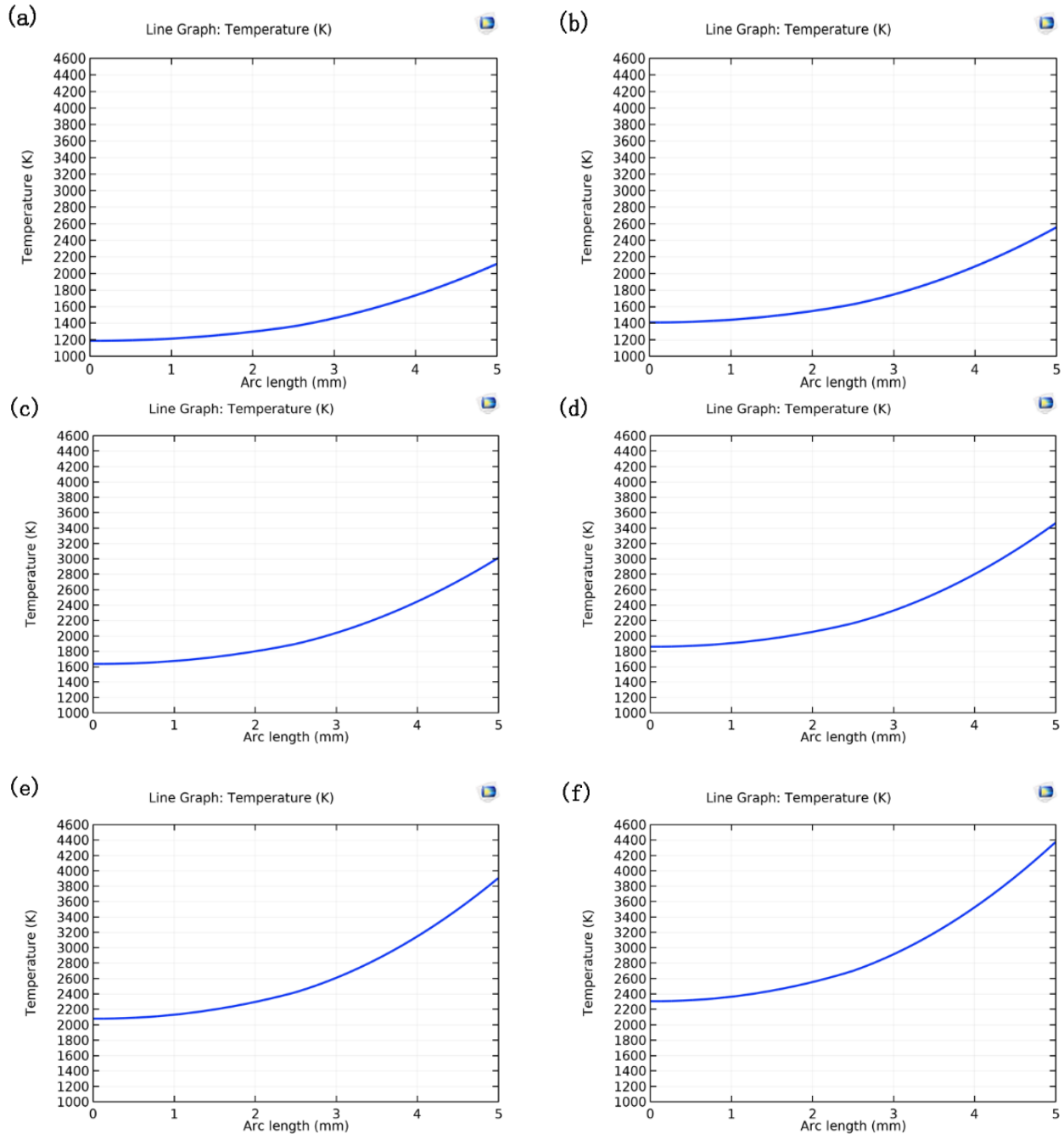


Figure 10. the temperature distribution along the thickness of the fillers at the laser spot center under different laser power;(a) 400W; (b) 500W; (c) 600W; (d) 700W; (e) 800W; (f) 900W.

Figs. 11(a)-(c) represented the stress field along the width of ceramic liners near the fillers where the laser spot was located after 10s laser radiation with power of 700W, 800W and 900W, respectively. The arc length represented the width of liners in Fig. 11. It can be elicited from Fig. 11(a)-(c) that the thermal stress of the liners was mainly concentrated around 2mm away from the fillers, and the maximum thermal stress gradually increased with the increase of the laser power. According to Fig.10(d), full penetration could be achieved when the laser power was 700W, and the laser utilization rate was the highest in comparison to the laser power of 800W and 900W. At this point, the laser energy density is about $23333 \text{ W}\cdot\text{s}/\text{cm}^2$.

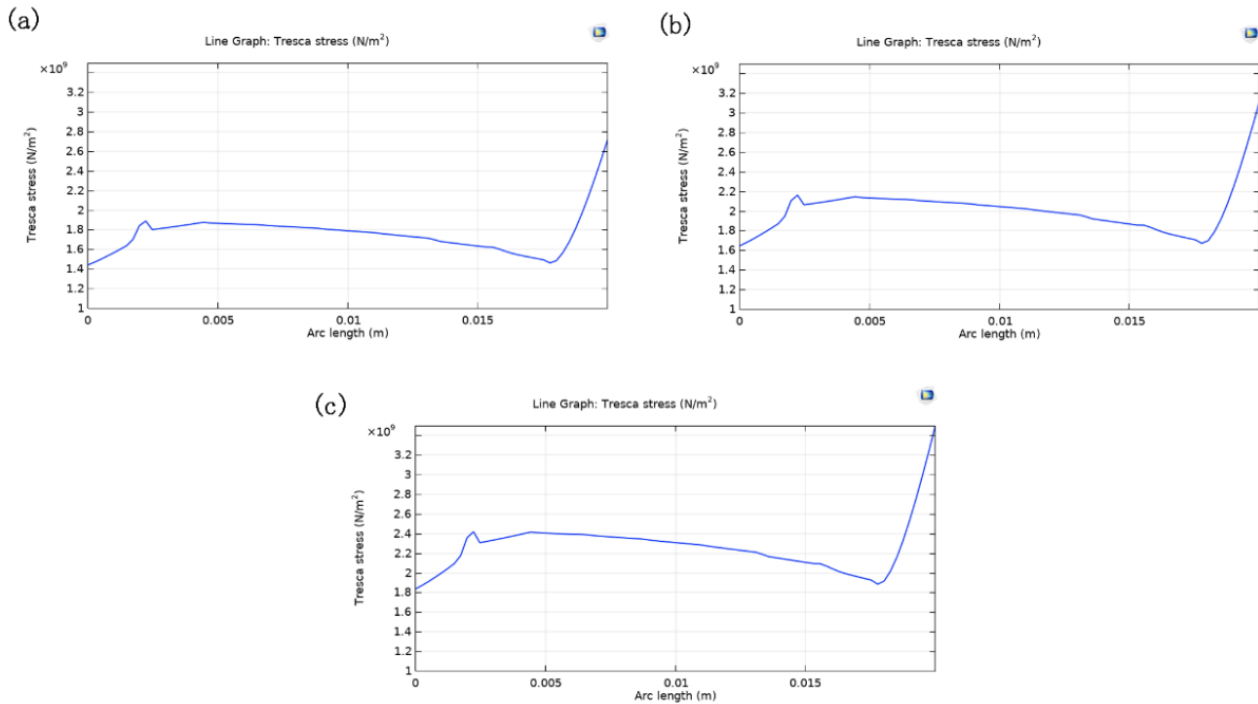


Figure 11. The stress field along the width of the ceramic liners near the fillers under different laser power; (a) 700W; (b) 800W; (c) 900W.

4.2. Experimental studies on the joint quality under different joining process parameters

The power density of laser is expressed as

$$E = P \cdot t/S \quad (6)$$

Where E is the power density, P is the laser output power, S is the laser radiation area, and the value of S is 0.3cm^2 ; t is the laser output time, $t=L/v$, L (10mm) is the length of the fillers, the value of L is 10mm, v is the scanning speed of the diode laser.

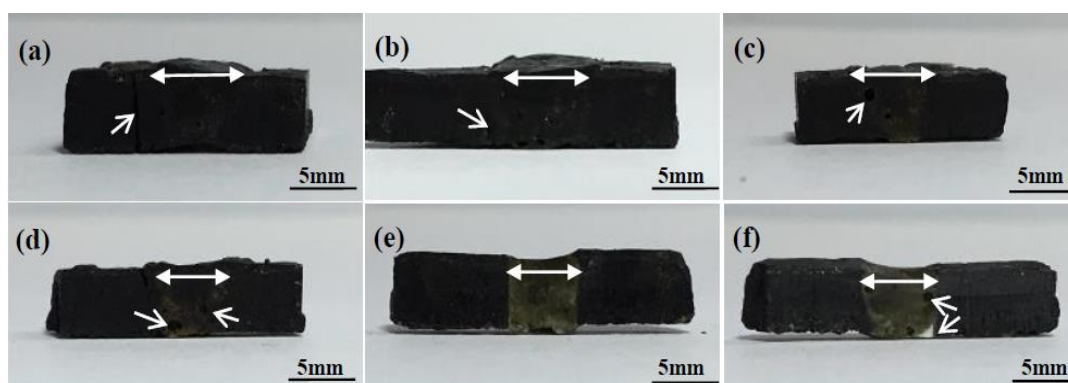
As can be seen from table 1, six groups of fillers with widths ranging from 1-3.5mm/s all achieved full penetration of weld zone under the minimum laser power density.

Table 1. The optimum laser process parameters for different fillers width

Fillers width (mm)	1	1.5	2		2.5		3	3.5
Scanning speed (mm/s)	0.8	0.6	1.2	0.8	1.2	0.8	1	1.4
Power of laser (W)	700	500	900	500	900	500	700	900
Power density ($\text{W}\cdot\text{s}/\text{cm}^2$)	<u>29167</u>	<u>27783</u>	25000		<u>25000</u>		<u>23333</u>	<u>21426</u>

Fig. 12 shows the joint morphology with full penetration under the optimized parameters for each group of fillers. When the width of fillers was 1 mm, more than 50% of the laser energy was directly irradiated on the liners. The instant warming and cooling in the laser joining and post-joining process resulted in a large thermal deformation and macro thermal crack in the weld zone, as can be seen in Fig. 12 (a). When the width of fillers reached to 1.5mm, more laser energy was applied to the fillers, while Al_2O_3 liners adjacent to the filler would still absorb excessive laser energy and generated thermal deformation, resulting in the joining thermal crack in weld zone shown in Fig. 12(b). When the width of fillers increased to 2 mm and 2.5 mm, the energy was mostly absorbed by fillers, and there was no macroscopic crack found in the ceramic liners. However, there still existed defects such

as pores (as shown in Fig. 13(c) and (d) in a single arrow) in the joint. When the width of fillers was 3 mm, the laser energy was totally irradiated on the fillers, the temperature rise at the liners only depended on the heat conduction from high-temperature filler to the liners. The fillers could completely melt and fill the gap of liners to form a dense joint after absorbing enough energy of the laser radiation, and there was no macroscopic crack and pores on the joining zone as shown in Fig. 12(e) due to the residual relatively low thermal stress in brittle liners. When the width of fillers increased to 3.5 mm, only the center part of the filler could absorb the energy of laser radiation and translated into heat, the edge region of the fillers failed to absorb the laser energy directly. The fillers failed to fully formed molten pool in the process of welding, the gap between the fillers and Al_2O_3 liners couldn't be completely filled by the molten pool (as shown in Fig. 13(f) in a single arrow). The experimental results are in good agreement with the numerical simulation results.



(a) 1mm; (b) 1.5mm; (c) 2mm; (d) 2.5mm; (e) 3mm; (f) 3.5mm

Figure 12. The interface of the joint

5. Conclusion

The Al_2O_3 - SiO_2 glass ceramics were determined as fillers for the diode laser joining of Al_2O_3 liners. To obtain the process parameters and verify the simulation results, the laser joining experiment was conducted using a 3 kW DILAS diode laser. The main conclusions are drawn as follows:

The mullite filler with the quality ratio of 70wt.% Al_2O_3 -30wt.% SiO_2 was determined as the initial filler for the joining of liners according to the phase composition of the Al_2O_3 liners and the physical/chemical compatibility between the fillers and liners. The thermodynamic properties of the Al_2O_3 liners and fillers were analyzed and tested. The temperature and stress field during the process of diode laser joining Al_2O_3 ceramic liners was analyzed by transient multiple physical model of based on COMSOL. According to the simulation results, 5mm penetration depth could be achieved under the relatively small thermal stress when the fillers width was 3mm, the power of the laser was 700W and the scanning speed was 1mm/s.

The joint quality under different width of fillers was studied. It can be concluded that when the width of fillers was 3 mm, the energy of laser was totally irradiated on the fillers, the rise of temperature at the liners only depended on the heat conduction from high-temperature filler to the liners. The fillers could completely melted and fill the gap of liners to form a dense joint after absorbing enough energy of the laser radiation, and there was no macroscopic crack and pores on the welding zone. The highest utilization of the laser radiation could be achieved while the power and scanning speed of the laser was 700W and 1mm/s, respectively. The experimental results are in good agreement with the numerical ones.

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