Welding of 304 stainless steel and glass using high-repetition-frequency femtosecond laser

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Abstract
In this study, a femtosecond laser with a repetition frequency of 0–400 kHz was used to join soda lime glass and 304 stainless steel. The effects of single-pulse power, repetition frequency, welding speed, and defocusing on the weld quality were investigated. The joining mechanism and fracture surface morphologies were studied using scanning electron microscopy and x-ray diffraction analysis. The results show that no new phases were formed between the glass and stainless steel, and that the joining mechanism consisted mainly of mechanical mixing between the two materials. Using a suitable combination of process parameters, a good weld with a strength of 8.79 MPa was obtained. The weld strength was influenced mainly by the amount of glass that adhered to the stainless steel, the bonding strength between the glass base material and the remelted glass, and the wetting of the stainless steel by the molten glass.

1. Introduction

Glass is oxidation-resistant, corrosion-resistant, and high-temperature-resistant, with high hardness and wear resistance; however, the low ductility and impact toughness of glass limit its application in engineering structures.

Connectors consisting of glass and metal with high plastic toughness and strong impact resistance have been widely used in industrial fields including solar cell, medical device, and sensor manufacturing [1–3]. The study of glass and metal connections has scientific significance and great engineering value.

With expanded application of glass and metal connectors in industry, connection methods have emerged including anodic bonding, brazing, and adhesive bonding. These bonding methods usually have critical defects, such as aging, degassing, a large thermal effect, and low accuracy, which cannot meet increasing requirements in terms of environmental effectiveness and miniaturization [4, 5]. Compared with these methods, laser welding has the advantages of non-contact connection, pleasing connection appearance, high precision, and no intermediate layer. Laser welding has broad application prospects in metal and glass connections. Femtosecond pulse lasers have a small thermal effect, nonlinear absorption, and high precision, and are suitable for welding transparent materials. They have been successfully used in high-precision welding of microparts such as electronic chips, and have become a popular research topic in the field of glass micromachining. When glass is welded with metal, the coefficient of thermal expansion changes greatly. The heat-affected zone is very small for welding similar optically transparent materials. Yasuyuki [13] used a femtosecond laser with a pulse duration of 130 fs and a repetition rate of 1 kHz in micro-welding glass and copper. It was found that femtosecond laser micro-welding can suppress the thermal effect in the welding process and accurately control the welding area, expanding its scope to include transparent materials and metals. Richard and Itoh [14] explored direct welding of transparent materials and metals, welding glass and a variety of metals using a femtosecond laser with a low repetition rate. Carter [15] found that an ultrashort pulse laser with a high repetition rate (on the order of 0.1–1 MHz) achieved faster and more efficient
melting or micro-plasma generation in these materials through heat accumulation (heat accumulation rate greater than cooling rate). Matsuyoshi [16] used a 1 MHz femtosecond laser to weld copper and glass without pressure assistance. At present, the research on metal and glass welding by high repetition rate femtosecond laser is still in the stage of feasibility study, and there is little research on its connection mechanism. In this study, high-repetition-rate femtosecond fibre laser welding of glass and austenitic stainless steel was investigated. The influences of laser pulse energy, defocusing, repetition rate, and welding speed on the weld quality and the metal–glass connection mechanism using a femtosecond laser were studied.

2. Materials and methods

Soda lime silicate glass with dimensions of 15 mm × 15 mm × 1 mm and 304 austenitic stainless steel with dimensions of 40 mm × 15 mm × 1 mm were used in the experiment. The chemical compositions of 304
austenitic stainless steel and soda lime glass are shown in tables 1 and 2, respectively. Before welding, the 304 stainless steel surface was ground with 400#, 800#, 1000#, 1500#, and 2000# sandpaper to improve contact between the stainless steel and soda lime glass. The soda lime glass and 304 stainless steel samples were ultrasonically cleaned in alcohol, dried, and fixed on the processing platform.

The femtosecond laser system experimental setup is shown in figure 1. The system consisted of femtosecond laser focusing, imaging, and processing of positioning parts. The light source was a Yb-doped photonic crystal fibre chirped pulse amplifier producing a femtosecond laser of 0–400 kHz, 270 fs, and \( \leq 25 \mu \text{J} \), with a centre wavelength of 1040 nm. The spot diameter was approximately 10 \( \mu \text{m} \). A half-wave plate and a polarising beam splitter (PBS) were used to adjust the femtosecond pulse energy. An objective lens (NA = 0.4) placed on a computer-controlled 3D translation stage was used to focus the laser pulse on the sample. Laser processing was monitored using a confocal microscope. The 3D platform was a Newport ESP300 with a minimum moving distance of 0.1 \( \mu \text{m} \).

| Number | \( e (\mu \text{J}) \) | \( f (\text{kHz}) \) | \( h (\mu \text{m}) \) | \( v (\text{mm s}^{-1}) \) |
|--------|-----------------|-----------------|-----------------|-----------------|
| 1      | 8               | 400             | 0               | 0.2             |
| 2      | 10              | 400             | 0               | 0.2             |
| 3      | 12              | 400             | 0               | 0.2             |
| 4      | 14              | 400             | 0               | 0.2             |
| 5      | 16              | 400             | 0               | 0.2             |
| 6      | 14              | 400             | -20             | 0.2             |
| 7      | 14              | 400             | +20             | 0.2             |
| 8      | 14              | 400             | +40             | 0.2             |
| 9      | 14              | 100             | 0               | 0.2             |
| 10     | 14              | 200             | 0               | 0.2             |
| 11     | 14              | 400             | 0               | 0.4             |
| 12     | 14              | 400             | 0               | 0.6             |
| 13     | 14              | 400             | 0               | 0.8             |

Figure 3. Effects of (a) laser pulse energy; (b) defocusing; (c) repetition frequency; (d) welding speed on weld strength.
To investigate the influence of the laser parameters on the weld quality, the welding speed \((v)\), laser pulse energy \((e)\), pulse repetition frequency \((f)\), and defocusing \((h)\) were varied, as shown in table 3. The laser was focused on the glass–stainless steel interface. After setting the laser parameters, light was emitted by the laser, and welding was performed by scanning along a straight line. The length of the welding line was 2 mm; the line spacing was 50 μm, and the number of scanning lines was 100, as shown in figure 2(a).

To study the interface morphology, samples were cut from the cross-section of the joint, ground, and polished. The interface morphology was observed using an environmental field emission scanning electron microscope (FEI-Quanta FEG 250). The shear strength test was conducted using the XYZTEC Condor 150 multifunctional push–pull tester (diagram of sample shear is shown in figure 2(b)) to investigate the influence of the process parameters on the welding strength. A shear force tester was used to gradually apply force to the welded samples until fracture occurred. The maximum force at which fracture occurred was recorded. The average of three welding patterns for each process parameter was recorded as \(F_{\text{max}}\). The weld strength \(\sigma\) can be calculated using equation (1) [17].

\[
\sigma = \frac{F_{\text{max}}}{S}
\]

where \(S\) is the welding area (as shown in figure 2(a)).

To understand the fracture mode of the joints, the glass and stainless steel fracture surfaces were characterised using SEM. The phases on both fracture surfaces were studied using D8 advanced XRD analysis.

3. Results and discussion

3.1. Mechanical properties

The weld strength is used to characterise the quality of welded joints. Figure 3(a) shows the relationship between weld strength and laser pulse energy. With a pulse energy of 8 μJ, a weld strength of 2.38 MPa was obtained. The pulse energy density is approximately 10.2 J cm\(^{-2}\), much greater than the ablation threshold of stainless steel (0.19 J cm\(^{-2}\)) [18]. Thus, reaching the ablation threshold of materials is not a sufficient condition for femtosecond laser welding. The femtosecond laser energy required for macro connection of metal and glass is...
much greater than the ablation threshold of the substrate metal material, causing a large area of glass to melt and fill the gap between the two materials. As the pulse energy increased, the weld strength between the two materials gradually increased. However, when the pulse energy reached 16 $\mu$J, macroscopic cracks formed on the glass base material, and the weld strength decreased sharply.

Figure 3(b) shows the relationship between the weld strength and the amount of defocusing. The greatest strength was obtained with no defocusing. As the focus position gradually shifts to both sides, the weld strength gradually decreases; when the laser focus moves downward, the stainless steel is ablated on the surface because it is opaque. With defocusing, the laser energy density on the stainless steel surface is not as strong as with focusing, and the thermal effect is small. When the focus moves upward, nonlinear absorption occurs inside the glass, with
an absorption rate less than that of stainless steel \([19]\). The distance between the focus position and the interface leads to a reduction in molten glass adhering to the stainless steel. Thus, the welding effect is best when the defocusing is zero.

Figure 3(c) shows the relationship between weld strength and repetition frequency. With an increase in repetition frequency, the thermal accumulation of the femtosecond laser is significantly improved, and the ablation threshold of the materials decreases \([20]\). With the same single-pulse power, the welding strength is significantly improved. The repetition frequency has a significant influence on joint strength. When the repetition frequency was decreased to 100 kHz, the joining was extremely weak; the weld strength was 0.89 MPa.

Figure 3(d) shows the relationship between weld strength and welding speed. As the stable operation speed of the platform was relatively low, the welding speed had little influence on the weld strength in the investigated range.

Figure 3 indicates that the strength of glass and stainless steel femtosecond laser-welded joints can reach 8.79 MPa with optimal process parameters. The weld strength is greater and the efficiency is higher than with traditional glass and metal sealing in an air atmosphere (3.6 MPa) \([21]\).

### 3.2. Weld cross-section

Parameters were selected for the best mechanical properties, and a sample was welded to observe the cross-sectional morphology. A cross-sectional view of the glass and stainless steel weld is shown in figure 4; the upper part is glass and the lower part is stainless steel. Only a small portion of the stainless steel melted, and there was almost no heat-affected zone. The melting zone was much larger on the glass side. The remelting zone underwent rapid cooling and shrinkage after high-temperature melting. Obvious defects were formed between the remelting zone and the glass base material, producing a weak link in the glass and stainless steel joint that was prone to fracture when subjected to force. In figure 4(b), it is observed that the molten glass entered the pit formed in the stainless steel, and the stainless steel also entered the glass, forming a mechanical mixture at the interface. It can be concluded that when the junction is irradiated by a laser, the material splashes from the molten pool in the form of gas or liquid, forming cylinders and nanowires greater than ten microns long after cooling and curing. An uneven mosaic structure was formed. The scanning results of the interface between the two materials are shown in figure 4(c). The results show that there is almost no element penetration at the junction of the two materials. Thus, there may be no metallurgical bonding between the stainless steel and glass, only mechanical bonding.

### 3.3. Fracture surface morphology

Figures 5(a) and (b) show the fracture morphology of the welded area on the glass side and the stainless steel side, respectively, with a pulse energy of 8 \(\mu\)J. A large amount of glass adhered to the stainless steel side, connecting the materials. In figures 5(c) and (d), it is observed that with an increase in single-pulse power, the amount of glass adhered to the stainless steel side during fracture increases. In figures 5(a) and (b), when the pulse energy is low, there is a broken area between the stainless steel substrate and the remelted glass; only some of the molten glass fills the gap and wets the stainless steel surface, due to the unevenness of the stainless steel surface. Comparing figures 5(c)–(f), it is observed that when the laser focus position moves upward, the femtosecond laser radiation
depends mainly on the nonlinear absorption inside the glass, and the absorption rate is less than the linear absorption on the stainless steel side. In addition, the melting zone is far from the interface; only a small amount of glass can fill the gap and wet the stainless steel side. Thus, the weld strength with this combination of process parameters is low. Figures 5(g) and (h) show that the amount of glass adhered to the stainless steel surface is small, and the damage on the glass side is small. Comparing figures 5(c) and (d), the repetition frequency of the femtosecond laser affects the material threshold. The laser repetition rate was low, the heat storage effect was poor, the ablation threshold was high \[20\], the ablation effect was poor, and it was difficult for the molten material to fill the gap to form a reliable connection. According to the analysis of figure 5, the main factors affecting the joint strength are: (1) the amount of glass filling the gap between the materials to wet the stainless steel side; (2) the bonding strength between the glass substrate and the remelted glass; (3) the wettability of the molten glass on the stainless steel.

The strength of femtosecond laser welding between glass and stainless steel is limited mainly by the characteristics of the glass. When the laser pulse energy is low, the melt is not sufficient to fill the gap between the two materials, making a connection difficult. When the laser pulse energy is high, large defects are formed in the glass due to poor toughness and rapid temperature changes; these defects can expand and fracture under stress.

Figure 6 shows the red-framed portion of figure 5 at higher magnification. As most of the stainless steel side of the section is covered by thick glass, it is difficult to see the shape of the glass–stainless steel interface; the area not completely covered by glass was amplified. The results show that when the femtosecond laser was focused on the interface, aperiodic holes appeared on the glass side, and many convex particles were formed on the stainless steel side. The scanning results of the point energy spectrum on the stainless steel side are shown in figure 7. Most of the stainless steel was covered with glass, but the prominent particles were stainless steel, confirming the results in sections 3.2 and 3.3.

XRD analysis was conducted on the glass and stainless steel sides of the fracture surfaces of the joint; the results are shown in figure 8. Figure 8(a) shows the XRD pattern of the glass side. The broadened diffraction peak
is the amorphous glass diffraction peak; there is almost no austenite or martensite phase of 304 stainless steel on the glass side. The XRD pattern of the stainless steel side in figure 8(b) shows that the austenite phase is the main phase, and iron oxide is formed as a result of air in the gap between the glass and stainless steel. The stainless steel was heated and melted by the laser. With the rapid heating and cooling of the laser, a martensite (110) phase appeared in the stainless steel. Upon fracture of the joint, most of the material in the welding zone adhered to the stainless steel. Thus, the stainless steel phases were hardly detected on the glass side fracture surface, and a large amount of amorphous glass adhered to the stainless steel surface did not form diffraction peaks. The XRD pattern and the line scanning results in section 3.2 show that there was no chemical reaction between the glass and stainless steel to form new substances. Only mechanical bonding occurred between the two materials.

3.4. Joining mechanism between metal and glass
A diagram of the femtosecond laser welding metal and glass is shown in figure 9. In the figure, +h refers to photon, black sphere refers to atom and e- refers to free electron. The surface roughness of the stainless steel was 3 μm. The glass was ordinary glass, and did not reach optical contact (the sample gap was less than a quarter of the wavelength [6]). Thus, the process was a large-spacing welding process. The material action area was maximised by multi-pulse laser heat accumulation, such that a large molten pool was formed after the material melted, and the material gap was filled by the ejected melt and plasma to form a weld. When the femtosecond laser was focused by the lens, a high peak power density irradiated the interface, and the upper glass was ionised by nonlinear absorption effects such as multiphoton absorption (MPA). The conduction band free electrons formed by ionization further absorb photons, and then produce more conduction band free electrons through collision ionization and avalanche ionization [22]. These electrons gradually form a plasma by collision ionization. The high-temperature plasma transferred the electron energy to the material lattice through electron–phonon coupling, and the material around the thermal diffusion focus melted or even vaporised. The lower metal contained a large number of conduction band free electrons that absorbed the laser through inverse Bremsstrahlung [23]. Thus, the metal mainly exhibits linear absorption of the laser through conduction band electrons; collision ionisation leads to avalanche ionisation, and eventually forms a high-density plasma state. Plasma enhances the absorption of the laser. Electron–electron and electron–phonon scattering, and lattice heat conduction processes occur. When the lattice temperature reaches the thermodynamic critical temperature, a phase change explosion occurs, resulting in a large number of high-temperature and high-pressure gases and liquids. The liquid and gas ejected near the focus are mixed. In the welding process, under laser irradiation with peak power, a large amount of plasma is generated by photoionisation and avalanche ionisation. The plasma gradually moves closer to the laser source, and reflects, refracts, and absorbs the laser, resulting in uneven absorption of longitudinal energy [24]. Until the plasma rises to a certain height, the laser power density cannot maintain the formation of plasma, and the process ends. In this process, the upper glass absorbs most of the laser energy. The high-temperature plasma produced by ablation of the lower metal can effectively reduce the threshold of the glass [25]. The glass used in the experiment (softening point of approximately 500 °C) melts more easily than stainless steel; thus, the melting zone and heat-affected zone of the glass are much larger. In addition to the blending of the two materials within the focus range, thermal expansion of the glass, wetting, and filling the gap between the two materials on the metal surface after melting are required to form a reliable connection. With the poor toughness of the glass, cracks are easily formed after rapid thermal expansion and contraction, representing a weak link that can fracture when stressed.
4. Conclusions

In this study, a femtosecond laser with a high repetition frequency was used to weld 304 stainless steel and soda lime glass. The influences of the laser process parameters on weld strength were investigated. The joining mechanism between the metal and glass was discussed. The following conclusions can be drawn.

(1) A femtosecond laser with a high repetition frequency can produce an effective connection between glass and stainless steel without optical contact. However, with the characteristics of the glass base material, the welding process window is relatively narrow. There are certain requirements on the surfaces of the two samples. When the gap is too large, the molten material formed by heating has difficulty filling, leading to welding failure.

(2) When the laser repetition frequency was maintained at 400 kHz, as the single-pulse power increased, the welding area became larger, and the weld strength first increased and then decreased. With a suitable combination of process parameters, the weld strength can reach 8.79 MPa.

(3) It was found that the welding strength depends mainly on the amount of glass wetted on the surface of the stainless steel and produce bonding strength between the glass substrate and remelted glass.

(4) No new phase is formed between the glass and stainless steel, indicating that the bonding mechanism is based mainly on mechanical mixing, wetting, and adsorption. The interface between the stainless steel and glass is ablated by an ultrashort pulse laser to form a mixture of gas, liquid, and plasma. The molten glass fills the gap between the two materials and wets the surface of the stainless steel; an adhesive welding of the two materials is achieved.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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