Simulation and experimental study on CO₂ laser polishing quartz glass

Ziruo Dai¹, Tao Chen¹* and Xiao Han²

¹Material and Manufactory Department, Beijing University of Technology, Beijing, 100124, China
²Beijing Space Electromechanical Research Institute, Beijing, 100094, China

*Corresponding author’s e-mail: chentao@bjut.edu.cn

Abstract. High quality surface can be quickly obtained by laser polishing quartz glass. With the improvement of application requirements of quartz glass, the combination of process parameters for optimizing CO₂ laser polishing large area quartz glass sheet is proposed. The untreated quartz ground glass was scanned by CO₂ laser. Taking the roughness as the research index value, the parameters affecting CO₂ laser polishing were studied and analyzed. The transient model was established by COMSOL software to simulate the polishing process, predict the combination of polishing parameters, and obtain the temperature field change in the polishing process and the flow of quartz glass during melting. 300W continuous CO₂ laser was used in the experiment. The effect of surface improvement was the best when the laser power was 300W, the scanning rate was 100~200mm/s. The micro area roughness can reach Ra=10.18nm. Then, the chemical corrosion was combined with CO₂ laser polishing, and the quartz glass was corroded by hydrofluoric acid and sodium hydroxide solution, and then polished to obtain the average minimum roughness Ra=5.63nm. The research results can be put into practical industrial production, which is conducive to improving production efficiency and promote the development of laser polishing quartz glass technology.

1. Introduction
The application of quartz glass in industry is of great significance. It has great application potential in high-tech fields such as aerospace, optoelectronics, semiconductor and nuclear engineering. At present, the surface treatment of transparent brittle and hard materials such as quartz glass by conventional mechanical processing or chemical processing technology is easy to produce crack damage and low processing efficiency. Laser polishing technology is non-contact polishing, which has the advantages of high degree of freedom, high horizontal and vertical resolution and fast processing speed. The infrared band emitted by CO₂ laser is easy to be absorbed by transparent materials, so the efficiency of processing quartz glass with CO₂ laser is high. There are many studies on the mechanism of glass polishing. Laser polishing of quartz glass is mostly based on the flow action theory.

From 2011 to 2012, Jörg Hildebrand[1-2] in Germany studied the factors affecting laser polishing fused quartz through experiment and simulation. The concept of "polishing line" is proposed. It is found that in an ideal temperature range, the rough polishing of quartz glass surface can be completed in one step without removing a large amount of raw materials, and the laser polishing rate is reduced to 4.8s/cm². From 2015 to 2017, Heidrich[3-6] (ILT) in Germany developed the laser polishing manufacturing process chain, the combination of laser polishing and laser correction to improve the speed to 1s/cm².
In recent years, it has been reported in China that [7-9] obtained high-quality super smooth surface by laser polishing fused quartz under different beams, chemical corrosion and laser polishing. The root mean square roughness of the surface is less than 0.5nm, the lowest is 0.156nm, and there is almost no micro defect or damage. It is concluded that the combination of low power density and slow scanning speed can improve the surface quality of laser polishing. The numerical model is used to simulate the laser polishing process. It is found that the surface tension controls the surface fairing process, which is the key factor to obtain the micro roughness of sub nano fused quartz laser polishing.

So far, the existing CO₂ laser polishing of quartz glass is studied after the surface reaches a lower roughness by mechanical polishing or other pretreatment methods. There is no experimental research on the laser polishing of quartz glass with larger original roughness. The research goal of this paper is to apply CO₂ laser polishing to the initial untreated quartz glass surface, and combine acid-base corrosion and laser polishing to treat the quartz glass surface.

2. Establishment of numerical model

Laser polishing involves the process of solid heat transfer and fluid flow. Laser irradiation makes the quartz glass below the evaporation temperature. The increase of temperature reduces the dynamic viscosity and makes the shallow surface of quartz glass melt and flow. Under the action of force, the flow redistribution occurs in the molten part, so that the surface roughness is smoothed.

2.1 temperature field simulation

In order to reduce the workload of repeated experiments, the temperature state in the polishing process can be determined and the polishing process parameters can be optimized through numerical simulation. The surface deformation of quartz glass surface micro bulge melting process under Gaussian beam and the temperature change during laser action are studied.

Governing equation of transient solid heat transfer:

\[
\rho C_p \frac{\partial T}{\partial t} + \nabla \cdot (-k \nabla T + q_r) = Q
\]  

(1)

Where parameters \( \rho \) is the density (kg/m³), \( C_p \) is constant pressure heat capacity (J/(kg∙K)), \( T \) is temperature (K), \( t \) is time (s), \( k \) is thermal conductivity (W/(m ∙K)), \( q_r \) is the radiant heat flux (W/m²), and \( Q \) on the right side of the equation is the heat source (W/m²).

Due to the melting of the shallow surface of quartz glass, the convective heat transfer with air should be considered. The convective heat transfer is calculated by Newton formula:

\[
Q = h \cdot (T_{\infty} - T_f) \cdot A
\]  

(2)

The convective heat transfer coefficient of gas is determined by:

\[
h = \left( \frac{k}{\alpha} \right) \cdot C_0 \cdot \left( \frac{G_r}{P_r} \right)
\]  

(3)

calculation. The above formula \( Q \) is the heat source (W/m²), \( h \) is the surface convective heat transfer coefficient (W/(m²K)), \( T_{\infty} \) is the temperature of the gas (K), \( T_f \) is the variable surface temperature (K) in contact with the gas, \( k \) is the thermal conductivity of the gas, \( a \) is the surface area (m²), \( L \) is the plate height (m), and \( C_0 \) is the experimental constant, which is obtained from the literature, where 0.54, \( G_r \) is Grashov constant, \( P_r \) is Prandtl constant, and all are empirical constants. The basic physicochemical properties of quartz glass change with temperature. The curves of density, thermal conductivity, viscosity, constant pressure heat capacity and temperature are obtained by consulting the literature.

In order to study the smoothing effect of laser scanning process on surface micro protrusion, COMSOL software is used to draw square 30mm×30mm, 3mm thick quartz glass sheet, draw a semi ellipsoid with a half axis of 0.04mm, B half axis of 0.12mm and C half axis of 0.01mm on the surface as micro protrusion, and the height of micro protrusion is \( \Delta h=40\mu m \) (as shown in Figure.1). The laser light source is approximately Gaussian intensity, the bottom surface of the quartz glass sheet model and the four sides a, b, c, d (as shown in Figure.2) are set as adiabatic, and the upper surface is subject to the incidence of the laser heat source and the natural convection and radiation with the air.
When the laser is scanned at full power of 300W, the surface of quartz glass melts, and its melting effect is related to the scanning rate. When the scanning rate is 100~200mm/s, the surface micro bulge is obviously smooth.

The relationship between the falling height of micro bulge and the scanning rate when the beam diameter is 1.2mm and the laser power is 300W is obtained, as shown in Figure.3. The Figureure shows that the falling height of micro protrusion decreases with the increase of scanning rate. When the scanning rate is large, the height of melting deformation of quartz glass surface is less than the height of micro protrusion. The melting degree of the surface is not enough and the flow of surface thin layer is not enough, which will increase the surface roughness. Therefore, the smaller the scanning rate within a certain range, the better the polishing effect.

As can be seen from Figure.4, the smoothing effect is the best when the scanning rate is about 100mm/s. In the pre experiment, we found that at full power (300W), when the scanning rate is more than 200mm/s, the molten pool depth on the surface of quartz glass is small, the surface roughness is more than 90nm, and the polishing quality is poor. Combined with the simulation results, we choose the scanning rate to be about 100mm/s, and we will continue to try finer parameters in the future.

Large area quartz glass is polished by laser to make the shallow surface part in the molten state, and the surface is smooth after remelting. However, there is overlap of molten pool, which will increase the surface roughness after cooling. In order to solve the problem of lap joint, it is proposed that if the whole surface is scanned before solidification, and the whole quartz glass sheet is remelted at the same time, the whole surface can be smooth. In order to prove the feasibility of this assumption, the three-dimensional model (as shown in Figure.5) is proposed to start from small area melting and gradually increase the scanning area. The simulation is still looking for the optimal parameter combination.
2.2 Melt flow simulation

Further considering the melting flow in the shallow surface of quartz glass, the micro mechanism of polishing process is studied. Combined with fluid heat transfer and level set two-phase flow module, the two-dimensional temperature and velocity model of quartz glass melting flow is established. German Kerstin Hecht[10] concluded that the surface removal of quartz glass material can be controlled and low surface roughness can be obtained in the range of 2000~2100°C. Therefore, it is better to keep the surface temperature within this range.

Governing equation of transient fluid heat transfer:

\[ \rho C_p \frac{dT}{dt} + \rho C_p u \cdot \nabla T + \nabla \cdot (-k \nabla T) = Q \]  
(4)

This model follows both Navier Stokes equation (5) and continuity equation (6):

\[ \rho \frac{du}{dt} + \rho (u \cdot \nabla u) = -\nabla p + \mu \Delta u + F \]  
(5)

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0 \]  
(6)

Where parameters \( \rho \) is the fluid density (kg/m\(^3\)), \( C_p \) is constant pressure heat capacity (J/(kg\(\cdot\)K)), \( T \) is temperature (K), \( u \) is fluid flow rate (m/s), \( k \) is thermal conductivity (W/(m\(\cdot\)K)), \( p \) is pressure (Pa), \( \mu \) is the dynamic viscosity (Pa\(\cdot\)s), and \( F \) is the volume force acting on the fluid (N). Set the laser power of 300W, the beam diameter of 1.2mm, and the scanning rates of 100, 150 and 200mm/s respectively. The total height of the two-dimensional model is 4mm, the upper part of 1mm is air, and the lower part of 3mm is the thickness of quartz glass sheet. Considering the effect of convective heat transfer with air.

The surface temperature variation of quartz glass and the melt flow rate in the molten part under different process parameters are obtained by the model. In general, the higher the laser scanning speed, the higher the surface temperature of quartz glass, the smaller the molten pool depth, and the smaller the absolute value of flow velocity in the molten part. In the calculation result of scanning rate of 100mm/s, we take two points scanned on the surface of quartz glass when \( t=0.1s \) (Figure.6), and obtain the variation diagram of their temperature and flow rate with time (Figure.7,8).

It can be seen from Figure 7,8 that the point time when the temperature reaches 2000~2100°C is about 0.05s, so the interaction time between laser and surface is 0.05s. Comparing the two Figure, it is found that the point 1 closer to the starting point of laser scanning melts after heating, and the melt flows in the opposite direction to the laser scanning, because its position is in the front of the spot, this point flows outward after heating, and heat conduction occurs with the interior of the material; As the scanning continues, the temperature of the spot is the highest, the surface tension gradient is generated with point 1, and the Marangoni flow occurs, so the flow direction of point 1 changes. Point 2 is behind the spot, and there will be no reverse flow. The main control factor of flow is the Marangoni effect caused by surface tension gradient.
3. Experimental method

3.1 Laser direct polishing

RF CO₂ laser with 300W output power of spurs laser is used in the experiment. The schematic diagram of laser polishing system is shown in Figure.9. The sample is 50mm prepared with 80#~100# a diamond grinding wheel on a plane milling mill50mm×50mm quartz glass sheet, 3mm thick, with an original roughness Ra average of about 700nm. Polishing experiments were carried out by adjusting the scanning rate, processing power, scanning spacing and defocus of CO₂ laser.

The machined surface is measured by Wyko NT1100 optical profilometer, and the surface roughness Ra value is observed, as shown in Table 1. The sample surface is untreated and polished by laser, which is later called direct polishing (Abbreviated as DP). The average surface roughness of directly polished quartz glass is less than 20nm, and the lowest is 10.18nm. At this time, the parameter combination is laser power 300W (full power), scanning rate 100mm/s, beam diameter...
1.2mm. The two-dimensional and three-dimensional models of the directly polished surface under this condition are observed, as shown in Figure.10.

### 3.2 Chemical corrosion assisted laser polishing

As mentioned in zuohuajifu's glass manual[11], hydrofluoric acid and hot concentrated alkali have strong corrosion effect on quartz glass. When etching the glass with acid, the glass surface tends to soften with the erosion, and the difficulty of polishing depends on the hardness of the hydrolyzed layer. Quartz glass (mainly composed of SiO₂) reacts with HF as follows:

\[
\text{SiO}_2 + 4HF \rightarrow SiF_4 \uparrow + 2H_2O
\]

SiF₄ is not only easy to hydrolyze, but also does not exist in water. The following reactions will occur:

\[
\text{SiF}_4 + 4H_2O \rightarrow H_4SiO_4 \downarrow + 4HF
\]

Where H₄SiO₄↓ is flocculent precipitation. The HF produced by the above reaction is coordinated with the remaining non hydrolyzed SiF₄:

\[
\text{SiF}_4 + 2HF \rightarrow H_2SiF_6
\]

The general equation of HF etching quartz glass can be obtained:

\[
2\text{SiO}_2 + 6HF + 4H_2O \rightarrow H_4SiO_4 \downarrow + H_2SiF_6 + 4H_2O
\]

General equation of sodium hydroxide etching quartz glass:

\[
2\text{NaOH} + \text{SiO}_2 \xrightarrow{\Delta} \text{Na}_2\text{SiO}_3 + H_2O
\]

The generated sodium silicate is a viscous material with high strength, which will increase the surface hardness of quartz glass.

In the experiment, 100ml of 20% HF and 40% NaOH solution (heated and kept constant at 100°C) were soaked in quartz glass for 2h, half of the samples were soaked, and the other half was used as the blank control group. Then polish both sides of the sample with the same parameters, and observe the surface roughness on both sides respectively.

### 4. Data analysis

It can be seen from table 1 that under the condition of constant laser power in direct polishing and chemical corrosion combined with laser polishing, the smaller the scanning rate of laser polishing, the lower the surface roughness of quartz glass, which is roughly consistent with the simulation results. When the laser power is 300W, the scanning rate is 100mm/s, the spot diameter is 1.2mm, the micro roughness of HF etching polishing (HF-P) is the lowest, Ra=4.82nm, and the average value is about 7.57nm. Under the same conditions, the roughness of polishing after NaOH corrosion (NaOH-P) is the lowest, Ra=1.83nm, with an average of about 5.63nm, which is greatly improved compared with direct laser polishing of quartz glass.

| sample          | DP1 | DP2 | DP3 | DP4 | DP5 | DP6 | DP7 | DP8 | DP9 |
|-----------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| laser power (W) | 300 | 300 | 300 | 300 | 300 | 300 | 300 | 300 | 300 |
| scanning rate (mm/s) | 100 | 150 | 200 | 100 | 150 | 200 | 100 | 150 | 200 |
| surface roughness result Ra (nm) | 16.04 | 17.7 | 18.07 | 7.57 | 8.79 | 9.91 | 5.63 | 5.93 | 6.35 |

The surface of the sample magnified 5000 times under the scanning electron microscope is shown in
Figure.11, in which (a) is the initial quartz glass surface microstructure, and there are obvious gullies, which is the main reason for the large roughness; After laser polishing, the surface (b) is smooth and the gully is basically eliminated; After HF immersion, the surface (c) gullies disappeared and spherical reticular microstructure appeared; After HF immersion, the laser polished surface (d) is obviously smooth and the spherical network structure is eliminated; After NaOH corrosion, the surface (e) gully disappeared and a semipore structure was formed on the shallow surface; After NaOH corrosion, the laser polished surface (f) is smooth with a small amount of pitting on the surface. Figure.12 shows the two-dimensional and three-dimensional morphology of the sample.

Figure11. Sem magnified surface microstructure 5000 times. (a) Initial quartz ground glass surface (b) Laser direct polishing surface (c) Quartz glass surface after HF immersion (d) Quartz glass surface after HF immersion and laser polish (e) Quartz glass surface after NaOH immersion (f) Quartz glass surface after NaOH immersion and laser polish

The Vickers hardness tester is used to measure the random multi-point surface hardness of quartz glass, after direct laser polishing, after HF corrosion, after HF corrosion polishing, after NaOH corrosion and after NaOH corrosion polishing, and calculate the average value. It is found that laser polishing (mean value 654.05HV0.2) or HF corrosion (607.50HV0.2), HF post corrosion polishing (629.78HV0.2) and NaOH post corrosion polishing (645.6HV0.2) have little effect on the surface hardness of quartz glass (about 600~700HV0.2). Among them, only the Vickers hardness value after NaOH corrosion reaches 776.3HV0.2, indicating that the generated sodium silicate increases the surface hardness of quartz glass.

Direct laser polishing can eliminate micro defects, obviously smooth the surface and reduce the roughness; The roughness is lower by chemical etching and laser polishing. Under acid corrosion, SiO2 reacts with HF to produce new substances. The hydrolyzed layer is soft and easy to be polished; Under alkali corrosion, sodium silicate with high viscosity and strength is generated by surface reaction, which destroys the Si-O bond and is easy to polish. However, the melt flow is weakened and the surface fluctuates greatly. It is found that the laser polishing effect is good after HF corrosion.

Figure12. Quartz glass surface under 3D profilometer. (a) HF etched and polished 2D surface (b) HF
etched and polished 3D surface (c) NaOH etched and polished 2D surface (d) NaOH etched and polished 3D surface

Figure 13. X, Y-axis height fluctuation curve of laser polished surface after HF etching

Figure 14. X, Y-axis height fluctuation curve of laser polished surface after NaOH etching

Figure 15. Sample of laser polishing

We take a 20μm×20μm area randomly on the direct laser polished surface. its surface fluctuation height is within 30nm. The polished surface after HF immersion takes the same area, and its surface fluctuation is within 4nm (as shown in Figure 13), which is higher than that of direct polishing; Take a 80μm×80μm area on the surface after NaOH corrosion and polishing, there is a large fluctuation in the y-axis (as shown in Figure 14), so the NaOH corrosion laser polishing method reduces the micro roughness and causes the waviness of large wavelength. The real object is obtained by combining laser polishing and HF corrosion, as shown in Figure 15. The tabletop grain can be clearly seen through the polished part, and there is obvious clarity change between the polished and unpolished parts.

5. Conclusion

Combined with the simulation results and the actual laser polishing experiment, the optimal scanning rate at full power (300W) is 100mm/s, the average surface roughness Ra of quartz glass is less than 20nm, and the lowest is 10.18nm; The method of laser polishing quartz glass to make the surface reach the molten state at the same time is feasible, which needs more simulation and experimental research; Acid base corrosion combined with laser polishing generates new substances. The average roughness Ra of polishing after HF corrosion is 7.57 nm, and the lowest Ra=4.82 nm, which is lower than that of direct polishing without treatment. After NaOH corrosion, the average Ra of polishing roughness is 5.63nm and the minimum Ra is 1.83nm, resulting in large waviness on the surface, which affects the polishing quality.

References

[1] Hildebrand, J., Hecht, K. et al. (2011) Laser Beam Polishing of Quartz Glass Surfaces. J. Physics Procedia, 12:452-461.
[2] Hildebrand, J., Wittor, B., Hecht, K., Bliedtner, J. et al. (2013) Experimental and Numerical Investigations of Laser Beam Polishing of Quartz Glass Surfaces. J. Ceram. Sci. Tech.,4(1):25-32.
[3] Christian, W. Emrah, U. Andreas, S. et al. (2017) Glass processing with pulsed CO₂ laser radiation. J. Applied optics,56(4):777-783.
[4] Heidrich, S., Willenborg, E., Weingarten, et al. (2015) Laser polishing and laser form correction of fused silica optics. J. Mat. wiss. u. Werkstofftech, 46(7):668-674.
[5] Christian, W., Sebastian, H., Wu, Y., et al. (2015) Laser polishing of glass. C. Optifab, Rochester, New York, Oct 11, Proc. SPIE 9633, 963303.

[6] Christian, W., Andreas, S., Edgar, W., et al. (2017) Laser polishing and laser shape correction of optical glass. J. Journal of laser applications, 29(1):011702.

[7] He, T., Wei, C., Jiang, Z., et al. (2018) Super-smooth surface demonstration and the physical mechanism of CO2 laser polishing of fused silica. J. Optics letters, 43(23):5777-5780.

[8] Wang, D., Fan, F., Liu, M., et al. (2020) Top-hat and Gaussian laser beam smoothing of ground fused silica surface. J. Optics & Laser Technology, 127: 106141.

[9] Cao, Z., Wei, C., et al. (2020) Ground fused silica processed by combined chemical etching and CO2 laser polishing with super-smooth surface and high damage resistance. J. Optics Letters, 45:6014~6017.

[10] Hildebrand, J., Wittor, B., Hecht, K., (2013) Experimental and Numerical Investigations of Laser Beam Polishing of Quartz Glass Surfaces. J. Journal of Ceramic Science and Technology, 4(1):25-31.

[11] Zuo Hua, J., et al. (1985) Glass manual. M. Beijing, China Construction Industry Press, China.