Influence of Free-Electron Density Distribution on Mechanical Strength in Micro-Welding of Glass by Picosecond Pulsed Laser

Zhiyong OUYANG*,**, Yasuhiro OKAMOTO*, Kazuki IHORIYA* and Akira OKADA*

(Received 12 May 2021, Accepted 15 July 2021)

The free-electron density is very important to investigate the nonlinear absorption phenomenon, which strongly relates to the joining characteristics such as mechanical strength in fusion micro-welding of glass material by an ultrashort pulsed laser. In this study, the free-electron density distributions for different numerical apertures (N.A.) were simulated, and photoelasticity observation and the mechanical strength in micro-welding of glass was experimentally investigated. N.A. 0.45 has a large difference of free-electron density in beam axis, and the area of large principle stress is generated. On the other hand, N.A. 0.65 can obtain small difference of peak electron density in the upper and lower parts of molten area, which led to continuous molten area formation. This joining characteristic resulted in small area of large principal stress difference and high mechanical strength.

Key Words: Ultrashort Pulsed Laser, Glass Material, Free-Electron Density, Molten Area, Numerical Aperture

1. Introduction

The reliable and stable joining of glass materials is required owing to the demand for glass products precise and sophisticated shapes in the field of micro electro mechanical systems (MEMS)1,2. In the past several years, ultrashort pulsed laser has been proven to be a powerful and reliable tool for fusion micro-welding of glass material3,4. This joining method has the capability for space selective welding, and it can be accomplished by local heating without any intermediate layer and mechanical contact5. In this method, ultrashort pulsed laser with high repetition rate leads to the melting glass material at the vicinity of focal region due to heat accumulation6. In glass material, the laser energy is absorbed by nonlinear absorption phenomenon, which consists of nonlinear photo-ionization and avalanche ionization7,8. In the nonlinear photo-ionization process, the electrons in the valence band are excited as free electrons, which are then promoted to the conduction band9. The free electrons in conduction band can impact and ionize the bound electron in the valence band, and the process can repeat itself to realize the avalanche ionization. When ultrashort pulsed laser was tightly focused inside the glass material, the laser energy was absorbed by nonlinear absorption phenomenon. The free electrons were generated, and played an important role during nonlinear absorption. The free electron density affected the temperature distribution and thus the melting of glass material. During welding process, the absorption point moved in upper direction along the laser axis, and the accumulation of the laser energy absorbed by avalanche ionization led to the expansion of absorption area. Then, the absorption point dropped down and shifted to complete the first cycle of molten area formation. The formation and characteristics of molten area affect the mechanical strength. Therefore, the free-electron density is very important to investigate the nonlinear absorption phenomenon, which is strongly related to the mechanical strength. Control of focusing situation of laser beam inside glass is necessary for obtaining the reliable welding process, and the numerical aperture (N.A.) of objective lens is an important condition that determines the focusing situation in glass. In micro-welding of glass by ultrashort pulsed lasers, nonlinear absorption is necessary to ignite the absorption of laser energy, and two kinds of numerical apertures N.A. 0.45 and 0.65, which can perform the nonlinear absorption of laser energy, were employed in this study. These N.A.s can change the appearance of molten area10, which would be caused by the different incidence and refraction angles of ray in glass.

In this study, the distribution of free-electron density for different numerical apertures was simulated, and photoelasticity observation and the mechanical strength in micro-welding of glass was experimentally investigated. N.A. 0.45 has a large difference of free-electron density in beam axis, and the area of large principle stress is generated. On the other hand, N.A. 0.65 can obtain small difference of peak electron density in the upper and lower parts of molten area, which led to continuous molten area formation. This joining characteristic resulted in small area of large principal stress difference and high mechanical strength.

2. Experimental methods

Fig. 1 shows the schematic illustration of laser irradiation setup

* Graduate School of Natural Science and Technology, Okayama University (3-1-1 Tsushima-naka, Kita-ku, Okayama 700-8530, Japan)
** Japan Society for the Promotion of Science (5-3-1 Kojimachi, Chiyoda-ku, Tokyo 102-0083, Japan)
in bead-on-plate welding experiment. A picosecond pulsed laser with wavelength of 1064 nm, pulse repetition rate of 1.0 MHz and pulse duration of 12.5 ps was used as a laser source, and the laser beam was focused by objective lenses of N.A. 0.45 and 0.65 with spherical aberration correction. The borosilicate glass (Schott, D263) specimen of 1.1 mm thickness was fixed on a clamping jig and moved linearly by using several combinations of processing parameters. After welding, the molten area was observed by an optical microscope from the cross-sectional view.

In order to investigate the formation of molten area, the laser beam irradiation area was observed from the side using a high-speed video camera (Shimadzu Corporation HPV2A). The frame rate was 64,000 fps, and the exposure time was a quarter of frame interval (4 μs). The laser beam was tightly focused inside the borosilicate glass specimen by objective lenses of N.A. 0.45 and 0.65, which enabled the nonlinear absorption that resulted in the creation of a free electron plasma. In this observation, some laser pulses were captured in one image, but the molten area was formed by not only one laser shot but also the sum of multiple laser shots. Thus, the movement of absorption point could be detected, and a series of the plasma light movement could be captured to observe the absorption point movement, which was calculated by using the feed rate of specimen. In addition, two achromatic lenses with focal lengths of 60 mm and 100 mm were combined in the optical system of observation, as shown in Fig. 2.

Since the stress distribution can indicate the stability of welding process, photoelasticity method was carried out to obtain the distribution of relatively principal stress difference during the creation of molten area under different focusing situations.

The experimental setup was shown in Fig. 3. In this study, isochromatic fringes were observed to obtain the distribution of relatively principal stress difference for N.A. 0.45 and 0.65, respectively.

The mechanical strength was evaluated by breaking test, which was carried out by a universal test machine (Shimadzu EZ-L), and the glass specimens were clamped as shown in Fig. 4(a). The glass specimen was fixed by these clamping jigs, and the shapes and dimensions of clamping device were shown in Fig. 4(b). The pulling load was slowly added to separate the welded specimen, which was in perpendicular direction of weld area to avoid the influence of optical contact force. In addition, clamping jigs were moved by using linear guides to avoid twist of test specimens during the pulling process, and speed of cross head was set to 0.5 mm/min. The pulling load was divided by the welding area to calculate the perpendicular tensile stress, which was recorded as the breaking stress.

3. Simulation model of free-electron density during one laser shot

In this simulation model, the depth inside glass along the laser beam axis was defined as the coordinate of “z”, and z = 0 is the top surface of glass, as shown in Fig. 5. The variation of free-electron density was calculated by using the feed rate of specimen.
Influence of Free-Electron Density Distribution on Mechanical Strength in Micro-Welding of Glass by Picosecond Pulsed Laser

OUYANG, OKAMOTO, IHORIYA, OKADA

density $\rho(z, t)$ at the time $t$ is described by the rate equation (1) \(11\): \[ \frac{\partial \rho(z, t)}{\partial t} = \eta_p I(z, t) + \eta_c I(z, t) \rho(z, t) - \eta_r \rho(z, t)^2 \] where $\eta_p$ is the photoionization coefficient, $I(z, t)$ is the laser intensity in glass, $K$ is the number of photons for photoionization, $\eta_c$ is the cascade ionization coefficient, and $\eta_r$ is the recombination coefficient. The laser intensity $I(z, t)$ is calculated by the Eq. (2) \(3\): \[ I(z, t) = \frac{2Q}{1.06 \times \pi w(z)^2} \exp \left[ -\frac{2z^2}{w(z)^2} \right] \exp \left[ -\frac{t}{\tau_p} - \frac{m_0 (z-z_f)}{c} \right] \exp \left[ \int_0^t a(z, t) \, dz \right] \] where $Q$ is the pulse energy, $\tau_p$ is the duration of laser pulse, $w(z)$ is the laser spot size in glass, $n_0$ is the refractive index of glass, $z_f$ is the distance from the top surface of glass to the focusing position, $c$ is the speed of light, and $a(z, t)$ is the absorption coefficient of laser beam to glass. The values of constants used in this simulation are shown in Table 1.

The calculation results of laser intensity for various numerical apertures are shown in Fig. 6. Fig. 6(a) shows the pulse waveform with Gaussian shape, and Fig. 6(b) shows the spatial distribution of laser intensity. It can be seen that the spatial distribution of laser intensity is symmetrical about the focusing position, in which peak intensity is obtained at the focusing point. Although N.A. 0.45 show higher peak intensity in the focusing position, N.A. 0.65 can maintain high intensity in a long distance along the beam axis.

### Table 1 Symbols and constants used in this simulation.

| Symbol | Description                      | Value            |
|--------|----------------------------------|------------------|
| $\eta_p$ | Photoionization coefficient      | $4.4 \times 10^{-28}$ |
| $K$    | Number of photons for photoionization | 4               |
| $\eta_c$ | Cascade ionization coefficient   | $3.1 \times 10^{-4}$           |
| $\eta_r$ | Recombination coefficient        | $2.0 \times 10^{-15}$          |
| $\tau_p$ | Duration of laser pulse          | 12.5 ps                      |
| $n_0$  | Refractive index of glass        | 1.52             |
| $c$    | Speed of light                   | $3.0 \times 10^8$ m/s         |

4. Results and discussion

4.1 Absorption point movement

When picosecond pulsed laser was tightly focused inside the glass, the focusing point was considered as the starting point for laser absorption, which was defined as the absorption point. High-speed observation was carried out to observe the absorption point movement, which is important to understand the formation of molten area. In this observation, the location of lowest point of absorption movement was set to 0 $\mu$m, and the direction to the laser source was considered as positive value. Objective lenses of N.A. 0.45 and 0.65 were used to cause different focusing situations. The results were arranged as the relationship between location of absorption point $H_t$ and time for different numerical apertures in Fig. 7. It can be seen that N.A. 0.45 obtained a large movement of absorption point and long cycle of absorption point movement, which caused long cycle of heating and cooling in the upper and lower parts of molten area. Thus, the imbalance of temperature distribution would result in a large thermal stress and unstable phenomena. N.A. 0.65 obtained smaller location of absorption point and shorter cycle of absorption point movement compared with that of N.A. 0.45.

4.2 Distribution of relatively principal stress difference observed by photoelasticity method

It is considered that different focusing situations caused by different numerical apertures may result in different stress distribution inside glass. In order to confirm this point, photoelasticity observation was carried out in the cases of...
objective lenses of N.A. 0.45 and 0.65. The pulse repetition rate was 250 kHz, laser scanning speed was 25 mm/s, and the number of pulse was $10^7$ pulse/m. The observation results of isochromatic fringes are shown in Fig. 8. It can be seen that at the vicinity of laser irradiated area, there was the bright part, which indicated the relatively principal stress difference. High brightness are a indicated large principal stress difference. Moreover, for each numerical aperture, the area in front of laser irradiation direction showed large principal stress difference. The area of large principal stress difference in the case of N.A. 0.65 was obviously smaller than that of N.A. 0.45. Moreover, the variation of relatively principal stress difference was also smaller in the case of N.A. 0.65, although absorbed laser intensity was equivalent. Therefore, it is expected that stable welding of glass can be achieved by using N.A. 0.65.

4.3 Evaluation of mechanical strength

The measurement results of breaking stress for N.A. 0.45 and 0.65 were shown in Fig. 9. Since the mechanical strength mainly depends on the characteristics of molten area, the optical microscope photographs of molten area from side view were also shown in the upper row of this figure. It can be seen that, in the case of N.A. 0.45, the bottom of molten area showed a remarkable spike shape, which was considered to reduce the mechanical strength. On the other hand, N.A. 0.65 could form continuous molten areas, which resulted in high mechanical strength. Therefore, the mechanical strength of molten area in the case of N.A. 0.45 was lower than that of N.A. 0.65. This result also proved that the superior focusing characteristics of N.A. 0.65 could achieve strong and stable joining of glass.

4.4 Free-electron density distribution

As mentioned above, different focusing situations cause different absorption point movement, which results in different stress distribution and mechanical strength. However, it is still not clear how the focusing situation affects the absorption point movement. Therefore, in order to investigate the absorption of laser energy for different focusing situations, free-electron density distribution in beam axis is simulated during one laser shot. Figs. 10(a) and (b) show the optical microscope photographs of molten area from cross sectional view in the case of N.A. 0.45 and 0.65, respectively. It can be seen that molten area typically shows a dual-structure, which consisted of an elliptical outer structure and a teardrop-shaped inner structure. The value of $H_z$ is defined as the distance from the focusing position to the evaluation point. The focusing position along the laser beam axis is defined as $H_z = 0 \mu$m, and the positive value of $H_z$ means that the points locate above the focusing position in the beam axis. The location of each evaluation point along the beam axis is defined as top point, center point and bottom point of molten area, respectively. In this study, top and bottom points are determined as the midpoints between outer and inner structures along laser beam axis of upper and lower parts, respectively. In addition, center point is determined as the point located at geometrical
Influence of Free-Electron Density Distribution on Mechanical Strength in Micro-Welding of Glass by Picosecond Pulsed Laser (OUYANG, OKAMOTO, IHORIYA, OKADA)

center of molten area along laser beam axis.

Figs. 10(c) and (d) show the calculation results of free-electron density at different points in the case of N.A. 0.45 and 0.65. The free-electron density increases drastically in a short time interval of about 30 ps. When the laser beam with high intensity is tightly focused inside glass, the electrons in the valence band are excited as free-electrons via photo-ionization, and these free-electrons are heated to even higher energy levels to initialize the cascade ionization. However, there must be a limitation of increase in the free-electron density due to the variation of peak intensity in Gaussian waveform of time-dependent laser intensity. Therefore, it takes a maximum value, which is defined as the peak electron density, in the variation of free-electron density. Then, the free-electron density begins to decrease due to the electron-hole recombination loss. Finally, the variation of free-electron density reaches the steady state, while it still keeps high value. On the other hand, there is a similar tendency as for the variation of free-electron density among the top, the center and the bottom points inside the molten area as for each numerical aperture. The obvious difference is the maximum value of free-electron density. The bottom, the center and the top points inside molten area take the highest, middle and the lowest value of peak electron density, respectively.

It can be seen that N.A. 0.45 can achieve higher peak electron density and larger height of molten area than that of N.A. 0.65. In addition, the difference of peak electron density between the top point and the bottom point is also larger by N.A. 0.45, which indicates that the laser energy is not absorbed simultaneously in the upper and lower parts of molten area. On the contrary, there is a small difference of peak electron density between the top point and the bottom point in the case of N.A. 0.65, which shows simultaneous absorption of laser energy. This phenomenon can contribute to both appropriate laser absorption and stable molten area formation.

In the discussion described above, the size of molten area is different with numerical apertures, which results in different values of $H_z$. However, the difference of $H_z$ may play a significant role during the calculation of electron density, which would affect the conclusion drawn from the results shown in Fig. 10. Therefore, to confirm the correctness of the above conclusion, the free-electron density was investigated under the similar height of molten area and the same points of $H_z$ by different numerical apertures. Fig. 11(a) shows the optical microscope photographs of molten area, and different pulse energies of 2 μJ and 3 μJ are used to obtain similar height of molten area for NA. 0.45 and 0.65, respectively. The values of $H_z$ for the top point, the center point and the bottom point inside molten area for N.A. 0.45 are set to the same as that of N.A. 0.65. Figs. 11(b) and (c) show the variations of free-electron density at different points for N.A. 0.45 and N.A. 0.65, respectively. The peak electron density of bottom point is 35 times higher than that of top point in the case of N.A. 0.45. On the other hand, the peak electron density of bottom point
The absorption point is formed and overlapped at the same time. Small area of large principal stress difference was obtained, and the variation of relatively principal stress difference was also smaller. Then, continuous molten areas are formed, which resulted in high mechanical strength. Therefore, multiple absorption points of laser energy in beam axis would contribute to a reliable joining of glass, because sufficient molten area can be obtained with relatively small stress.

5. Conclusions

The main conclusions obtained in this study are as follows:

1) In the case of N.A. 0.45, the difference of peak electron density between the top point and the bottom point is larger than those of N.A. 0.65, and the absorption point moved, which led to enlarging the area of large principal stress difference. Then, the remarkable spike shape was generated, which reduced the mechanical strength.

2) In the case of N.A. 0.65, the difference of peak electron density between the top point and the bottom point is small, and small area of large principal stress difference was obtained. Then, continuous molten areas are formed, which resulted in high mechanical strength.

3) Small difference of peak electron density between the top point and the bottom point in the absorption area of laser energy can create continuous formation of molten area, which leads to the reliable joining of glass with high mechanical strength.

Acknowledgment

This work was supported by Grant-in-Aid for JSPS Fellows, JSPS KAKENHI (Grant Number: JP21J13688).

References

1) I. Miyamoto, A. Horn, J. Gottmann, D. Wortmann and F. Yoshino: “Fusion welding of glass using femtosecond laser pulses with high-repetition rates”, Journal of Laser Micro/Nanoengineering, 2 (2007), 57-63.

2) K. Sugioka: “Progress in ultrafast laser processing and future prospects”, Nanophotonics, 5 (2016), 393-413.

3) I. Miyamoto, Y. Okamoto, R. Tanabe, Y. Ito, K. Cvecek and M. Schmidt: “Mechanism of dynamic plasma motion in internal modification of glass by fs-laser pulses at high pulse repetition rate”, Optics Express, 24 (2016), 25718-25731.

4) E. Mottay, X. Liu, H. Zhang, E. Mazur, R. Sanatinia and W. Pfleger: “Industrial applications of ultrafast laser processing”, MRS Bull, 41 (2016), 984-992.

5) W. Watanabe, S. Onda, T. Tamaki, K. Itoh and J. Nishii: “Space-selective laser joining of dissimilar transparent materials using femtosecond laser pulses”, Applied Physics Letters, 89 (2006), 021106.

6) S. Richter, S. Döring, A. Tünnermann and S. Nolte: “Bonding of glass with femtosecond laser pulses at high repetition rates”, Applied
7) I. Miyamoto, Y. Okamoto, R. Tanabe and Y. Ito: “Characterization of plasma in microwelding of glass using ultrashort laser pulse at high pulse repetition rates”, Physics Procedia, 56 (2014), 973-982.
8) M. Sun, U. Eppelt, S. Russ, C. Hartmann, C. Siebert, J. Zhu and W. Schulz: “Numerical analysis of laser ablation and damage in glass with multiple picosecond laser pulses”, Optics Express, 21 (2013), 7858-7867.
9) B. Chimier, O. Uteza, N. Sanner, M. Sentis, T. Itina, P. Lassonde, F. Legare, F. Vidal and J. C. Kieffer: “Damage and ablation thresholds of fused-silica in femtosecond regime”, Physical Review B, 84 (2011), 094104.
10) Z. Ouyang, Y. Okamoto, Y. Ogino, T. Sakagawa and A. Okada: “Influence of numerical aperture on molten area formation in fusion micro-welding of glass by picosecond pulsed laser”, Applied Sciences, 9 (2019), 1-15.
11) A. Vogel, J. Noack, G. Hüttman and G. Paltauf: “Mechanisms of femtosecond laser nanosurgery of cells and tissues”, Applied Physics B, 81 (2005), 1015-1047.

Mail Address
Zhiyong OUYANG  po0x5g7k@s.okayama-u.ac.jp