

# Characterization of a Bonding Method for High Borosilicate Glass to Kovar Alloy Sealing by Laser Welding

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## Abstract

Glass to metal sealings have many excellent properties and they are extensively used in various industries. Applications for laser glass tube for carbon dioxide laser requires hermetic and mechanically strong seals. With the purpose of fully utilize its effectiveness of glass and metal sealings' properties and enlarge their applications, bond the glass to metal is an important method. Hermetic seals between kovar alloy and borosilicate glass can be produced using a novel laser sealing method when compared to bonding method based on glass pressing at temperatures around the softening point. This paper presents investigation results for the shear tensile bond strength and the waterproof resistance depending on the manufacturing process parameters. The shear bond strength of the glass to metal sealing was amounted to only 9.84 MPa when using traditional high temperature holding process. The maximum shear bond strength can be amounted to 43.68 MPa when sealed by laser welding process. And good adherence with a very high waterproof resistance can ben obtained. The bonding mechanisms was revealed by SEM methods to analyze the interface.

**Keywords:** glass to metal sealing; laser welding; shear strength

## 1 Introduction

With the rapid development of industrial fields, glass to metal sealings are extensively used in the fields which can create vacuum-tight electrical connectors, feedthroughs or optical windows and lenses in electrons or electronic system to meet requirements for harsh environments and high performance. Glass to metal sealings are widely used in industrials such as encapsulated electrical components and systems, semiconductors, electro-chemicals, pyrotechnics, or other substances [1-7]. Glass-to-metal sealing plays a very important role in electronic packaging. Glass to metal sealing can meet a wide variety of specific requirements such as airtight, hermetic packaging applications, whose performance, safety, and durability are very crucial [1-7]. The quite different properties exist between glass and metal, hermetic and reliable bonding between glass and metal is a big challenge for us. Hermetic sealing quality of glass to metal sealings are engineered to deliver uncompromised reliability. Given that the glass material and the metal alloy are quite different materials in physical and chemical properties, the hermetic bonding between glass and metal will be a big challenge when they are used in our actual industrial areas.

Laser functional and electronic devices such as laser

glass tube for carbon dioxide laser are encapsulated with the purpose of protecting the specific gases from leakage and activated into powerful Kovar alloys combine mechanical strength with similar coefficient of thermal expansion (CTE) to borosilicate glass, making them the material of candidate for most high performance containing electronic systems. Figure 1 demonstrates a laser glass tube for carbon dioxide laser electronic system, which is a typical application of borosilicate glass to kovar alloy sealing. Optical windows having function of optical diagnosis systems are other applications for this type glass to metal sealing. High-quality clear-transparent borosilicate glasses offers excellent and uniform optical transmission from the visible to the infrared spectrum and has remarkable thermal capabilities, and retains its mechanical strength. What's more, the hermetic kovar alloy and the glass sealing is a very important component part of the carbon dioxide laser system, as shown in Figure 1 (a).

Bonding of glass and metal is an old technology and has a rich history. What's more, glass-to-metal seals have now been in use for over a hundred years dating back to ancient times. At that time, then on metal surfaces, decorative glass coatings were manufactured. First glass melting and then wetting of the metal surface is the process of this ancient coating technology, and

later, it is called as enameling. Light bulbs and enameled cookware are most industrial applications in our daily life. And they are manufactured by the enameling process, in which using high temperatures to melt the glass, allowing it to wet the metal surface<sup>[1]</sup>. And the construction of vacuum tubes, electric discharge tubes, semiconductor diodes, reed switches, and pressure-tight glass-to-metal windows are the other common use. The basic process is sealed in a temperature-controlled oven, and the assembly is heated to the appropriate temperature for the particular variety of glass to melt and fill the space between the pin and the metal package. Hermetic sealing of glass and metal to meet the requirement of electronics industry is still challenging and is a hotspot in the current days. And researchers pay much attention to study this research area<sup>[8-9]</sup>. The shortcomings of this method is the high temperature heating and long time holding, it will cost large energy and high temperature treatment also may harm the functional properties of glass and/or metals. And it is not easy to achieve automatic production.

Due to the shortcomings of the traditional glass to metal sealing process used high temperature heating process, other low temperature process used for glass to metal sealing have been introduced by many researchers<sup>[10]</sup>. The anodic bonding process is a typical example<sup>[10]</sup>. During this bonding process, the glass remains in its solid state and the glass-to-metal interface is created by static electric fields. In the microfluidics industry, this sealing methods is already get applied<sup>[11]</sup>.

With the purpose of trying to find an alternative solution, low temperature heat process used for glass-to-metal bonding technology was once introduced in references<sup>[12-19]</sup>, has emerged as an important research in advancing welding technology. Laser welding technology has the advantages that has low process temperature and also it can be operated under atmosphere.

The interface created at highly focused laser energy and formed on localize zone, which allows an interim transitional zone contained both glass and metal. The main research objectives of the present paper is to investigate the laser process parameters on the sealing quality of the glass to metal sealing.

Kovar alloy, a nickel-cobalt-ferrous alloy, is one of the most commonly used glass sealing alloys. While borosilicate glass has constitutions of silicon oxide, boron oxide, and sodium oxide<sup>[20]</sup>, which has outstanding properties such as low CTE, good thermal stability, high mechanical property. Due to borosilicate glass's excellent properties, it is widely used in nuclear filed, electronic devices and electronics, optical window, vacuum-tight electric connectors and so on<sup>[21]</sup>.

Success bonding between Kovar alloy and borosilicate glass was achieved by Satoru Nishikawa and Makoto Takahashi<sup>[22]</sup> using anodic bonding. In their study, the authors attempted to study the influence of

voltage on the sealing joint and they analyzed the element distribution feature on the bond surface. And it can be inferred that the remaining contact area will be influenced by changing the bonding temperature. The mechanical properties and the microstructure characteristics of the joint between borosilicate glass and stainless steel once reported by authors Li et al.<sup>[21]</sup>. They analyzed the influence factors of shear strength of joint, such as bonding temperature, surface pretreatment, and reaction time were analyzed in their study. The maximum shear strength 27 MPa can be obtained at the conditions of holding at 1050 °C for 45 min, and the 304 SS was pre-oxidized.

The present study examined the quality of glass to metal sealing by a modern laser welding technology. First, the Kovar alloy was underwent high-temperature pre-oxidization treatment with the purpose to producing an oxide layer with a certain thickness and adhesion on kovar alloy surface. Previous research recommended that the optimum oxide layer formed and the alloy surface, which can be bonded with a high quality junction is about several  $\mu\text{m}$ <sup>[23]</sup>. According to the published literature and the authors previous published paper<sup>[1,23]</sup>, the optimum oxidizing temperature and holding time is easily determined in the present paper. Then the glass to metal sealing is sealed by laser welding technology. The interface structure is examined and the shear tension strength and leakage properties are determined.

## 2 Experimental Procedure

### 2.1 Preparation of samples

The laser bonding of kovar alloy and borosilicate glass, as an alternative sealing methods is used in the present study. Kovar alloy has a composition of 29% Nickel, 17% Cobalt, Balance Iron (in wt %) and its Coefficient of Thermal Expansion (CTE) is  $50 \times 10^{-7} / ^\circ\text{C}$ . High borosilicate glass has excellent chemical stability, thermal stability and good mechanical properties, and has been widely used in instrument glass, fireproof glass, flat panel display and solar cells and other fields.

The high borosilicate glass chosen for the present investigation has a coefficient of thermal expansion ( $10.3 \times 10^{-6} \text{ K}^{-1}$ ). It is similar to the coefficient of thermal expansion of kovar alloy ( $50 \times 10^{-7} / ^\circ\text{C}$ ). It has a composition of  $\text{SiO}_2$  (80.4%),  $\text{B}_2\text{O}_3$  (12.7%),  $\text{Al}_2\text{O}_3$  (2.4),  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  (4.2%), others  $\text{BaO}$  (0.3%), all in weight percent.

### 2.2 Preoxidation treatment

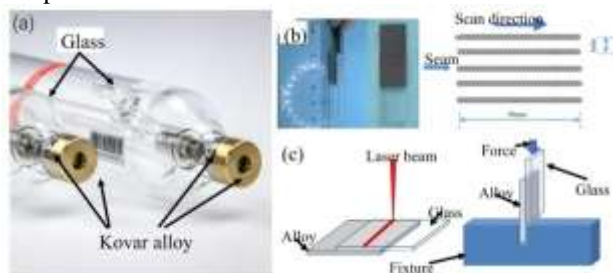
The Kovar samples are flat shapes with dimensions  $10\text{mm} \times 10\text{mm} \times 3\text{mm}$ . Filled with atmospheric air, the samples are heated in a muffle furnace at a heating rate of  $40 \text{ }^\circ\text{C min}^{-1}$  to three different temperatures, i.e 700, 800 and 900 °C, for seven different isothermal holding periods of 0, 3, 5, 10, 15, 30 and 60 min. After heat

treatment, all the samples are cooled to room temperature by cooling at  $20\text{ }^{\circ}\text{C min}^{-1}$ . According to previous published papers [1, 23], the optimal process parameters was selected as following:  $800\text{ }^{\circ}\text{C}$  and holding 5 minutes.

### 2.3 Bonding process: laser welding

The experimental setup for laser transmission bonding and weld strength measurement was shown in Figure 2. And Figure 2 (a) illustrates the welding process and Figure 2 (c) shows a picture of the real clamp for welding sample. Laser processing parameters are selected as following: 150 W, 200 W, 250 W, 300 W, 350 W and scan speed 0.3 mm/min. The focus distance are selected as 0 mm, 0.5 mm (focus beam is located in the glass) and -0.5 mm (focus beam is located below the metal surface). The pulse width is set as 2.8 ms and the laser frequency is selected as 10 Hz.

The Glass to metal welding experiment was conducted using an MD-Focus 500 NdYAG pulsed laser. This type of laser has wavelength of 1064 nm and it has a maximum average power of 500 W. This laser also has a maximum frequency of 500 Hz and its pulse width can be varied from 0.1 to 20 ms. A lap joint form was used for the glass to metal welding as shown in Figure 1(c). A fixture was designed to provide pre-tightening force to reduce the gap between the glass to metal, as shown in Figure 2 (c). Figure 2(a) shows the schematic diagram of the fixture and Figure 2 (b) demonstrate the welding state during laser welding process. Glass to metal sealing samples are laid on the fixture's workbench.



**Figure 1** Laser glass tube for carbon dioxide laser (a), glass to metal sealing samples formed by laser welding (left sample) and its scan direction (right picture), a typical example for glass to metal sealing (b), and schematic illustration of the glass to metal sealing sample welded by laser welding process (left picture) and the shear test (right picture) (c) (drawings not to scale)

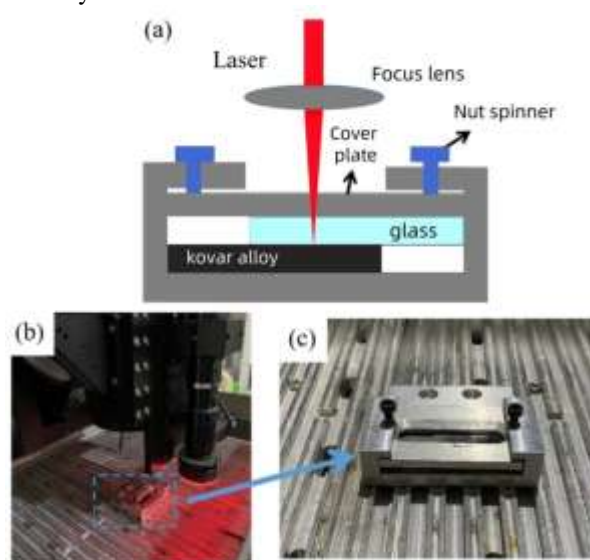
### 2.4 Microscopic observation and measurement of tensile strength and waterproof test

The glass to metal sealing samples are cross-sectioned, mounted and then they are polished into mirror surface and etched using 10% Nital (10% nitric acid +90% alcohol). Samples (joint interface and oxide layer formed on metal surface) for microstructure observation are then observed using optical microscope and scanning electron microscope (SEM, JOEL,

JSM-7000F).

The shear strength of the obtained glass to metal sealing is indicated by modified the traditional tensile test machine. The shear tensile testing machine used in the present study was the QC-548M2F-Computerized Tensile/Compression Testing from Cometech Testing Machines Co., in a line rate of 20 N/s up to 1000 N and capable of applying a maximum tensile force of 5 kN and with a feeding speed ranging from 0.0002 to 3 mm/min. During the sample separation process, recorded the load as a function of position with an accuracy of 0.001% was recorded the load by a digital dynamometer. And the shear strength is calculated by the applied load to the sample weld seam area. Figure 1(c) shows the schematic diagram of the shear tests.

The sealed samples were directly subjected to water bath furnace, heating at a higher temperature  $150\text{ }^{\circ}\text{C}$  holding for 30 minutes. And they were taken from the bath, cooled to room temperature. And after cleaning and dried by hot wind, the samples were tested by PET Colorant to see there exists obvious crack or not by naked eyes.



**Figure 2** Schematic diagram of the picture shown the welding: schematic diagram of laser welding with a clamp, (b)real welding photo and (c)its real clamp (drawings not to scale)

## 3 Results and discussion

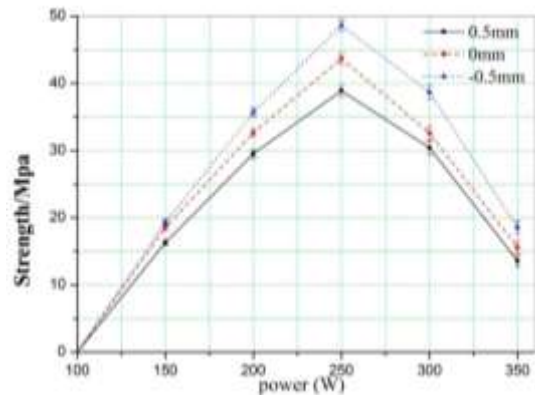
### 3.1 Tensile bond strength and waterproof resistance

The shear strength plays a critical role in determining the quality of glass to metal sealing. During laser welding process, laser processing energy is one of the most important processing parameters, which can significantly affect the final joint quality. Figure 3 demonstrates the shear strength (SS) as a function of laser power for three different focal plane positions. The laser power was varied by adjusting the laser output

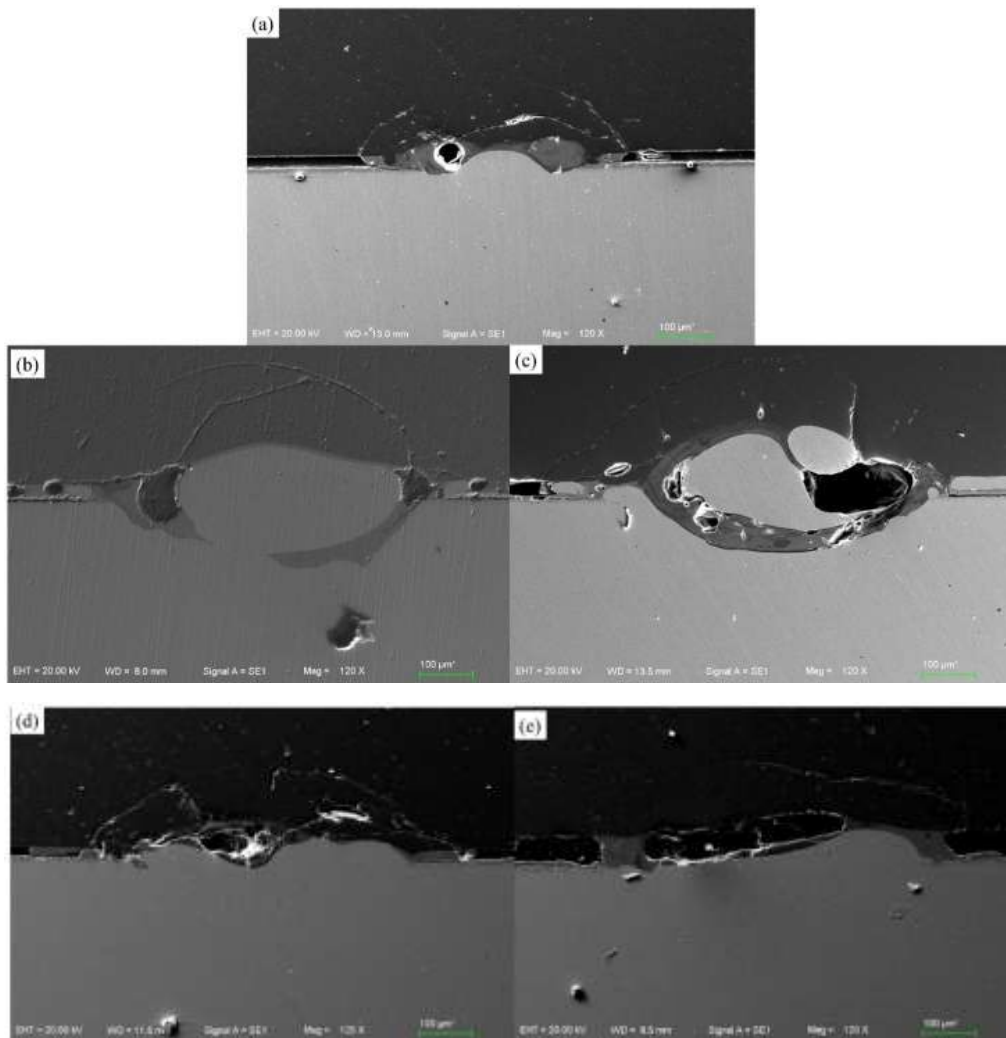
power ranged from 100 W to 350 W, i.e., 100 W, 150 W, 200 W, 250 W, 300 W and 350 W. The three focal plane positions were negative (0.5 mm below the interface, i.e. below the metal surface), at the interface, and positive (0.5 mm above the interface, i.e. in the glass), respectively. During laser welding processing, a fixed sample jig clamping force of 60 N was applied to provide pressure assistance and ensure the close contact.

Traditional glass to metal sealing strength once was reported that the shear strength value can be reached 9.84 MPa [1,24]. So the present study achieved higher strength when compared to tradition furnace sealing process. During the present study, the shear strength is shown in Figure 3. It can be seen that the strength is increased first. When it is amounted to maximum value and then decreased. Strength results showed that the focus plane and laser power have significant effects on the seal strength. Maximum strength can be reached to 43.68 MPa when the laser power is 250 W under the conditions of the focus plane was set as -0.5 mm. And it can be seen

that all the strength results obtained under -0.5mm are higher than that those obtained under 0 mm and + 0.5 mm focus plane.



**Figure 3** Experimental results demonstrating the variation of shear strength with incident laser pulse energy varied from 100 W to 350 W under three different laser focus plane



**Figure 4** Cross-sectional SEM pictures showing the cross-section interface between glass to kovar sealing under different laser process parameters: (a); 150 W; (b)200 W; (c)250 W; (d) 300 W; (e)350 W;

And according to the waterproof testing results, the waterproof level can be reached to IPX7. After PET Colorant test, there exists no obvious crack by naked eyes.

Five of the glass to kovar alloy sealing samples with chosen laser processing parameters were cross-sectioned perpendicular to the glass to metal sealings interface, then they were mounted, grounded, polished for the preparation of microscope observation by SEM. Figure 4 demonstrates the SEM pictures for the selected laser process parameters observed for the interface. These images depict the internal structure of the welds before separation. These images depict the internal structure of the welds before separation. And it can be clearly seen the bonding zone between glass and kovar alloy.

### 3.2 Bonding mechanism

Glass to metal sealing mechanism can be roughly divided into mechanical bonding and metallurgical adhesion [25]. And interlocking is the base for the mechanical bonding. In mechanical bonding, one material will fill voids, pores of the another material. While, metallurgical bonding interface successfully forms through electrostatic and molecular forces. And the commons forms are chemical forces or van der Waals forces.

The oxide layer was obtained by pre-oxidation of Kovar alloy and then the sealing of the glass to the pre-oxidized metal can be obtained. For mechanical and hermetic sealing also requirements were comprised of an adequate wetting of the pre-oxidized alloy surface by the glass and an appropriate dissolution of the surficial metal oxide in the glass, without formation of detrimental crystal phases [1, 24]. These requirements were shown to depend on the values of temperature, time and atmosphere of sealing under optimized with the values mentioned above.

In general, optimal oxide film formed on the kovar alloy surface and proper laser processing parameters are the fundament reasons to obtain both good shear strength and hermetic seal quality defined by waterproof test.

In the SEM images in Figure 4, the materials are bonded together after laser welding. And no visible gap between the glass to metal sealing interface can be clearly seen under the present magnification. The observed seal quality is verified by the high shear strength and high waterproof measured in samples produced with the same process parameters. Accordingly, the measured waterproof is high. However, the parameter combination resulted in a relatively high shear strength amounted to 43.68 N.

The SEM images in Figure 4 show that the shapes are spherical cavities in the glass sample that were exposed during the preparation of the SEM samples. Due

to the fact that not all the sample have cavities, it can be inferred that they must be formed during the bonding process. Gas is the main reason for the formation of cavities. And there are two main sources of gas formation, i.e., one comes from already existed gas and the another originates from chemical reaction during the laser welding process at higher temperatures. Till to now, such phenomenon were not reported in the literature references. And the higher strength can be obtained by -0.5 mm focus plane means that the bonding is achieved by melted metal alloy surface first and then mixed with glass.

## 4 Conclusion and Outlook

Results with high shear strength and high waterproof resistance glass to metal sealing occur under optimal proxidation kovar alloy and laser processing parameters. Good mechanical joint can be obtained and the maximum shear strength can be amounted to as high as 43.68 MPa in the present study.

Bubble formation at the interface can be seen in some laser process parameters. Shear strength can be amounted to 43.68 MPa. And selected parameters can be selected by analyzed the glass to metal sealing interface using SEM.

It is very important that deep understanding the bonding mechanisms to obtain a better sealing performance with the optimal process parameters.

What's more, finite element method can be used and studied on the stress evolution and forming to obtain process parameter optimization.

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