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Effect of Several Surface Treatments on the Strength of a Glass Ceramic-to-Metal Seal

**Daniel P. Kramer, Robert F. Salerno,
and Eugene E. Egleston**

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Abstract

Test shells of Inconel 625, Inconel 718, 21-6-9 stainless steel, and Hastelloy C-276 were plasma and/or chemically cleaned before sealing with a multi-component glass-ceramic. The effect of the different surface treatments on the strength of the glass ceramic-to-metal seal was evaluated using a hydrostatic burst test. The results show that plasma cleaning can be used to increase the hydrostatic burst strength and hermeticity of a glass ceramic-to-metal seal.

Introduction

The effects of several different surface treatments on the strength of a glass ceramic-to-metal seal were evaluated using a hydrostatic burst test. The shells used for this study were made of Inconel 718, 21-6-9 stainless steel, Inconel 625, and Hastelloy C-276 and were sealed using a multi-component, silicate, glass ceramic. Before sealing, the test shells were treated with a 4-step cleaning process in addition to plasma cleaning using different gases.

Experimental Work

The general purpose of this work was to determine the hydrostatic bursting strength as a function of surface treatment of the glass ceramic-to-metal seal using test shells of different high-strength alloys. Figure 1 shows the experimental design flow sheet that was followed for the study.

After fabrication, the metal test specimens (Figure 2) were treated with the selected surface treatments (Table 1). All three batches of the test specimens were treated with a 4-step cleaning process. In addition to the 4-step cleaning process, two batches of test specimens were plasma cleaned: one batch was plasma cleaned using oxygen gas; the other was cleaned using argon gas.

In plasma cleaning, gas is excited to an ionized state by applying radio frequency power. The radio frequency power is applied externally to the sample chamber, under reduced pressure conditions, to produce a highly reactive plasma at low temperature operating conditions. The complete unit consists of a reaction chamber, RF power supply, and a vacuum system. The choice of gas determines the surface reactions that will occur and hence determines the surface treatment.

After the selected surface treatment had been performed on the test specimen, the shell was sealed using a glass ceramic. The glass ceramic used during this study was a multi-component, silicate glass ceramic known as S-glass. The sealing cycle consisted of a sealing temperature of 1000°C for 9 min, a nucleation temperature of 650°C for 15 min, and a growth temperature of 840°C for 5 min. Total sealing cycle time was about 6 hr, and it was performed in an argon atmosphere. Figure 3 shows a generalized firing cycle for sealing the test specimens.

In order to ensure that the test specimens contained the same thickness of glass ceramic, a grinding operation was necessary. During this operation the top and bottom faces of the glass-ceramic slug were ground flat. The grinding operation continued until the amount of glass ceramic left in the sample was ~0.250 in. thick. Both before and after the grinding operation the test specimens were tested for hermeticity.

At this stage the specimens were ready to be tested using a hydrostatic bursting apparatus. In this equipment, pressurized air drives a fluid pump that is capable of pressurizing the fluid. Each specimen was tested in the same way. The samples were screwed into a test fixture and hydrostatic pressure was applied until failure occurred. That pressure was then recorded.

Results and Discussions

Figures 4 through 7 show the hydrostatic burst test results of the different surface treatments on the strength of the glass ceramic-to-metal seal. Statistical treatment of the burst data was performed by determining the Analysis of Variance (ANOVA) for this experiment. With this statistical analysis it was possible to determine whether there was a significant difference in the seal strength due to the different surface treatments. Additional information about this experiment was obtained by calculating and

FIGURE 1 Experimental design followed during the course of the research.

FIGURE 2 - Outline of the burst test specimens used in the hydrostatic burst test.

plotting the Least Significant Intervals (LSI) in the several figures. In all cases, the Least Significant Intervals were calculated at the 90% probability level (these are the bars plotted on the figures around the mean values). The mean determined from the burst test results for each surface treatment studied is the mean of five observations. Therefore, a total of 60 test specimens were prepared and hydrostatically burst in the course of this work.

In the case of test specimens fabricated of Inconel 625 (Figure 4), test shells that had been 4-step cleaned and treated with the oxygen plasma had significantly stronger seals than shells that had only been 4-step cleaned or 4-step cleaned and treated with argon plasma. Four-step cleaning alone gave stronger seals than a combination of 4-step and argon plasma cleaning.

Figure 5 shows the seal strength measurements for test specimens fabricated of 21-6-9 stainless steel. Again, the experimental results have determined that shells both 4-step chemically cleaned and oxygen plasma treated have significantly stronger glass ceramic-to-metal seals. However, it also shows there is no significant difference in

Table 1 - METAL SURFACE TREATMENTS USED DURING THIS WORK

4-Step Cleaning Process	Plasma Cleaning	
	Method 1	Method 2
1) Ultrasonically clean in methylene chloride (2 min)	Gas: Argon	Oxygen
2) Ultrasonically clean in 4-step cleaning solution (2 min)	Pressure: 0.5 torr	0.5 torr
	Watts: 200	200
3) Rinse in distilled water ultrasonically clean in distilled water (1 min)	Time: 0.5 hr	2 hr
4) Ultrasonically clean in isopropyl alcohol (1 min)		
5) Dry under heat lamp (minimum 15 min)		

seal strength between 4-step cleaning alone and argon plasma cleaning in addition to 4-step cleaning.

When test specimens fabricated of Hastelloy C-276 were burst, the results showed that both 4-step cleaning and 4-step cleaning plus oxygen plasma cleaning produced higher seal strengths than plasma cleaning with argon (Figure 6). But there was no significant difference in seal strength between 4-step cleaning alone and the oxygen plasma surface treatment.

For the study in which test shells were fabricated of Inconel 718 (Figure 7), we found that the only significant difference in seal strength was between 4-step cleaning only and argon plasma treatment. In all other combinations of the three surface treatments, the results showed no significant difference.

Another factor that can determine the eventual application of a particular glass ceramic-to-metal seal is its ability to make and maintain a hermetic seal. This property is, of course, surface dependent and therefore was investigated as part of this study. Figure 8 shows the effect of surface treatment on the hermeticity of the glass ceramic-to-metal seals studied. The most

dramatic effect was found for the case of test shells made of Inconel 718. We determined that plasma cleaning with argon, in addition to 4-step cleaning, produced hermetic seals in all of the Inconel 718 test shells. To a much lesser degree, the same result was found when Inconel 625 was processed in the same way using argon plasma cleaning. Glass ceramic-to-metal seals made with Hastelloy C-276, however, were hermetic only when the test shells were treated with the oxygen plasma process. This ran counter to the results found for 21-6-9 stainless steel which determined that oxygen plasma was the only surface processing technique that did not produce hermetic seals.

Conclusion

This study indicates that a plasma surface treatment can play an important role in improving the overall strength and hermeticity of a glass ceramic-to-metal seal. Adding a plasma surface preparation to test shells that have been chemically cleaned can increase the burst strength of the seal by as much as a third. Plasma surface treatment can also dramatically increase the hermeticity of a glass ceramic-to-metal seal. Of course, the type and degree of improvement was found to be dependent on the materials selected for making the seal.

FIGURE 3 - Generalized firing cycle showing the several temperature regions used for sealing the test specimens.

FIGURE 4 - Effect of the several surface treatments on the burst strength of test specimens fabricated of Inconel 625.

FIGURE 5 - Effect of the several surface treatments on the burst strength of test specimens fabricated of 21-6-9 stainless steel.

FIGURE 6 - Effect of the several surface treatments on the burst strength of test specimens fabricated of Hastelloy C-276.

FIGURE 7 - Effect of the several surface treatments on the burst strength of test specimens fabricated of Inconel 718.

FIGURE 8 - Effect of surface treatment on the hermeticity of the glass ceramic-to-metal seals.

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