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April 1984

CERAMIC-TO-METAL BONDING FOR PRESSURE TRANSDUCERS

Principal Investigator: J. D. Mackenzie

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FINAL REPORT

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
NASA-LEWIS RESEARCH CENTER

for

CERAMIC-TO-METAL BONDING FOR PRESSURE TRANSDUCERS
(Contract No. NAG3-295)

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ABSTRACT

A solid-state diffusion technique involving the placement of a gold foil between INCONEL X-750 and a machinable glass-ceramic "MACOR" was shown to be successful in bonding these two materials. This technique was selected after an exhaustive literature search on ceramic-metal bonding methods. Small expansion mismatch between the Inconel and the MACOR resulted in fracture of the MACOR when the bonded body was subjected to tensile stress of 535 psi. The bonded parts were submitted to a cyclic loading test in an air atmosphere at 1 Hz from 0 to 60 KPa. Failure was observed after 700,000 cycles at 650°C. Ceramic-Inconel bonding was not achieved with this method for boron nitride and silica glass.

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1. Introduction and Background

A high temperature transducer capable of continuous operation was designed and tested for NASA in 1977 (NASA Report No. Cr. 135282, Contract No. NAS-3-19556). A key part of the transducer was the diaphragm which was made of a thin plate of silica glass. The maximum use temperature was to be 650°C. It was not certain if silica glass was the ideal material. Subsequently, a one-year research effort was conducted at UCLA under NASA sponsorship (NASA Contract No. NAG-3-69) to evaluate a large number of solids to ascertain their relative merits as high temperature diaphragm materials. It was concluded that a glass-ceramic material called "MACOR," manufactured by Corning Glass Works, was probably the best candidate material rather than silica glass. The major disadvantage of silica glass was its very low coefficient of thermal expansion. Other candidate materials, also probably superior to silica glass, were boron nitride (BN) and silicon nitride (Si_3N_4).

In the original design of the high temperature pressure transducer using the silica glass diaphragm, the sensor was positioned tightly within an Inconel X-750 alloy outer casing via a spring. Sealing was achieved mechanically via a gold O-ring at the top and an alumina washer at the bottom. (See Figure 1, a reproduction of Figure 47d from NASA Report No. Cr-135282). In other words, the sensor was not hermetically sealed, that is, not chemically bonded to the Inconel casing.

The objective of this project was to examine the feasibility of forming a chemically bonded seal between materials such as "MACOR," BN, Si_3N_4 and even silica glass with Inconel X-750 alloy. If this was possible, the gold O-ring and/or the alumina washer of the original design could be eliminated. The main problem in the successful continuous operation of the transducer was considered to be the expansion mismatch between Inconel X-750 and the diaphragm materials such as "MACOR" and BN although the mismatch was very significantly less than that involving silica glass. After repeated heating and cooling of the transducer assembly over a long period of time, the expansion mismatch between the ceramic material and the metal casing must constitute a serious problem. A comparison of the expansion coefficients of some of those materials is shown in Figure 2. The expansion mismatch problem can be minimized by (a) the use of a graded seal, (b) the use of materials with very low elastic modulus, and/or (c) the proper design of a seal, for example, one having a bellow-type structure. The materials used must of course be bondable to Inconel X-750 on the one side and to the ceramic on the other. It must of course be able to withstand prolonged exposures to an oxidizing atmosphere up to 650°C. No such seal design was known in 1982.

This project was initiated in the summer of 1982 and was to be divided into two phases. The first phase was a non-experimental program, mainly consisting of literature search plus some theoretical considerations. The second phase was to be experimental in nature and would be concerned with the preparation of seals between Inconel X-750 and some of the above-mentioned ceramic materials and the testing of the seals. This is the final technical report of the one-year project.

2. Non-Experimental Program

In the ultimate design of the seal, considerations must be given to the materials selected, the behavior of such materials under cyclic treatment at the elevated temperatures and oxidizing atmosphere in question, the manner in which a chemical bond is to be formed, the minimization of stresses arising from expansion mismatch and the optimum design which would provide maximum strength. For instance, a chemical bond could be formed by the use of frit or by the use of electric-field assisted diffusion, etc. Minimization of stress could be achieved if the two materials in question have different expansion coefficients by the use of a graded seal and/or by a bellow-type design. In the first phase of this project, a thorough literature search was made with the help of the computer-aided search facilities of the UCLA Research Library to obtain information on the formation of seals, the design of seals and the best sealing materials available. Some of the most appropriate references are listed in Table 1.

From the literature search and from theoretical considerations, a solid-state diffusion bond using gold foil as the material between Inconel and the ceramic appeared to be a promising approach. The plan was to use gold foil alone as a start. If bonding was achieved and the bond strength was adequate, then cyclic tests would be performed. If the strength was inadequate then other foil materials could be used in addition to the gold so that graded seals could be formed on both sides of the gold. A glass-type frit was originally considered. However, the MACOR" supplier, Corning Glass Works, was unable to supply the proper frit. An independent frit development program for "MACOR" would require a separate research and development project.

Other readily available frits were considered to be unlikely candidate materials because of the 650°C upper temperature of operation of the transducer assembly. At such temperatures, deformation and/or devitrification of the frit was considered to be a major problem. The experimental program carried out thus involved only solid-state bonding utilizing metallic foils, and mainly gold foils.

3. Bonding of Inconel X-750 to Ceramics

A small apparatus was designed and constructed for the formation of the ceramic-metal bond via the gold foil. This apparatus is shown in Figures 3-7. The bonding method involved the placement of an Inconel X-750 rod, 1/4 inch in diameter and 1 - 1/4 inch in length, on top of a ceramic rod of the same dimensions. A gold foil of 12.5 microns thickness was placed between the Inconel and the ceramic. Pressure up to 285 psi was applied for 1 to 1 - 1/2 hours at temperatures between 875° and 950°C in dry nitrogen. This technique enabled the bonding of Inconel to "MACOR." An adapter was designed and constructed to measure the bond strength in tension in an Instron apparatus. Figure 8 shows the assembly mounted in an Instron apparatus. The highest fracture stress at room temperature for the Inconel-MACOR bond was 535 psi (3,700 KPa). The fracture always resulted in a cone-shaped piece of "MACOR" still bonded to the Inconel as shown in Figure 9. It appeared that the bond was fairly strong at the two interfaces but that expansion mismatch between "MACOR" and Inconel had resulted in a lowering of the strength and hence fracture on tensile loading.

Figure 10 shows a cross-section of the interfaces in question. Interdiffusion profiles are shown in Figures 11 and 12. Dissimilar diffusion coefficients had apparently led to the formation of Hartley- Kirkendall voids as seen in the gold layer in Figure 10. Results in Figure 11 are somewhat surprising in that silicon had apparently migrated from the "MACOR" through the gold into the Inconel. Both nickel and chromium had migrated into the gold and a small amount of chromium had even penetrated into the "MACOR." Interdiffusion resulting in bond formation in the present Inconel-Gold-MACOR system is obviously highly complex.

In order to minimize the expansion mismatch between "MACOR" and Inconel, an Ascoloy alloy was inserted between two gold foils. However, bonding was unsuccessful on the "MACOR" interface. Aluminum foil was tried in place of gold. Results were unsatisfactory. No bonding was possible between silica glass and Inconel and between boron nitride and Inconel using the gold foil method and under the same conditions as for "MACOR." Attempts to machine silicon nitride rods to the same dimensions as the Inconel and "MACOR" rods were unsuccessful. Such rods were also not readily available from vendors. Thus no experiments were conducted with the bonding of silicon nitride to Inconel. A summary of the diffusion bonding experiments is shown in Table 2. A summary of the materials tested, their chemical compositions and the suppliers used are shown in Table 3.

4. Cyclic Experiments with Inconel-MACOR Bonds

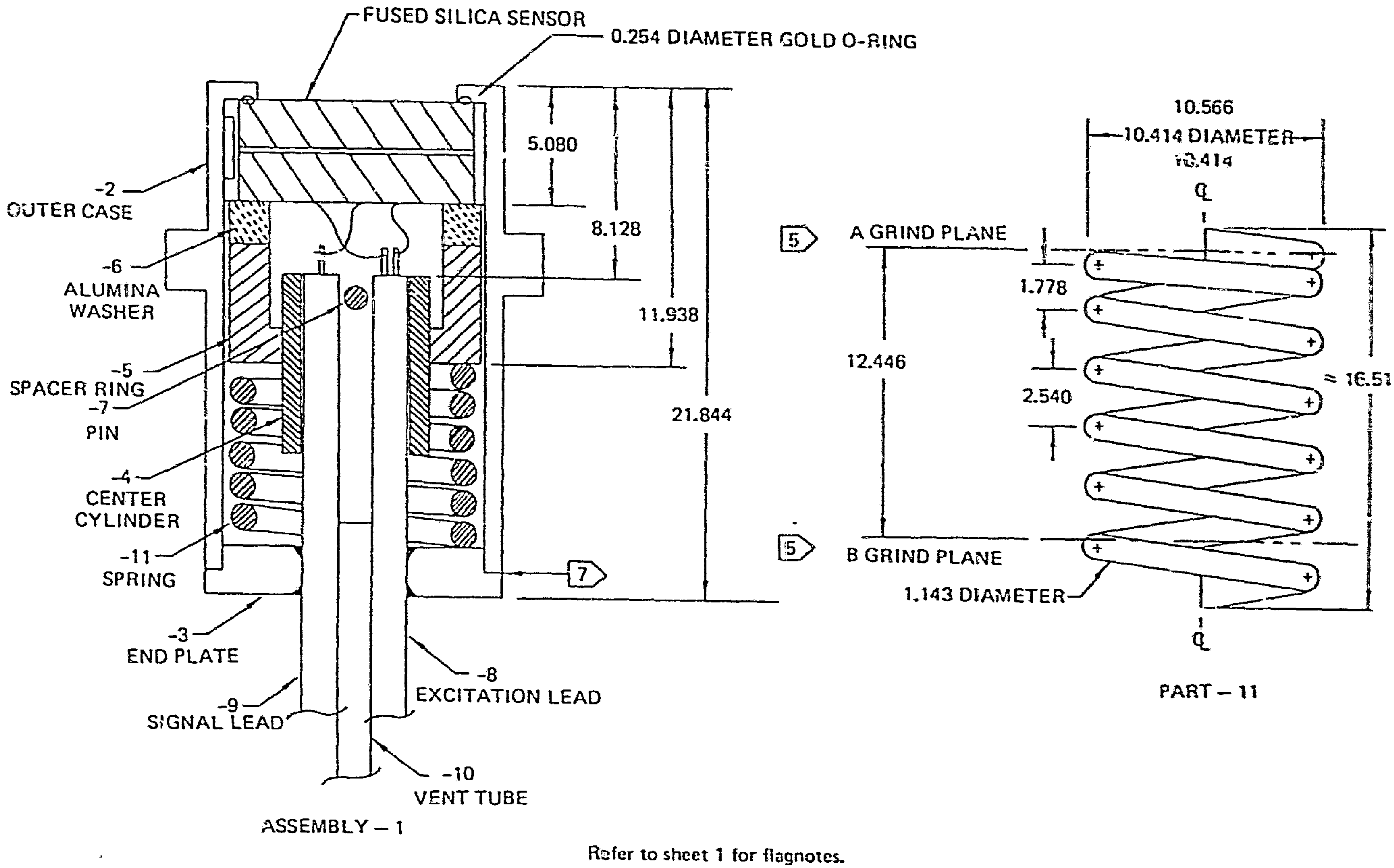
Because the tensile strength of 535 psi observed for the Inconel-MACOR bond was fairly high and the possibility existed that the bond strength could be increased with further experiments, it was considered that fatigue tests under cyclic conditions should be carried out. A fatigue testing system was thus designed and constructed. A schematic representation of this apparatus is shown in Figure 13. Details of the sample holder are shown in Figure 14. Figure 15 and 16 are photographs of the system and the two-way pressure cycle valve respectively. "MACOR"-gold-Inconel samples bonded at 950°C as described above and in the configuration shown in Figure 14 were mounted in the sample holder and placed in an electrically heated furnace at 650°C. Pressure was applied on the gold seal through the Inconel tube as shown. Air was alternately pumped into the Inconel tube and withdrawn from the tube at one cycle per second via the two-way cycle valve. The maximum pressure exerted was 60 KPa. Fracture was observed after an average of 700,000 cycles under these conditions.

5. Conclusions

(a) Inconel X-750 alloy can be bonded to "MACOR" machinable glass-ceramic by the use of a gold-foil diffusion bonding method. The tensile strength of the bond is in excess of 500 psi.

(b) The "MACOR"-gold-Inconel type bonds can be kept at 650°C and subjected to some 700,000 cycles of air pressure from 1 to 60 KPa at 1 Hz before bond failure was observed.

(c) It is probable that the bond strength and the bond life could both be improved with additional developmental efforts.



Reproduced from *Figure 47d. Case Assembly Drawing, Sheet 4* , NASA Report NO. CR-135282

Figure 1. Original design of transducer assembly

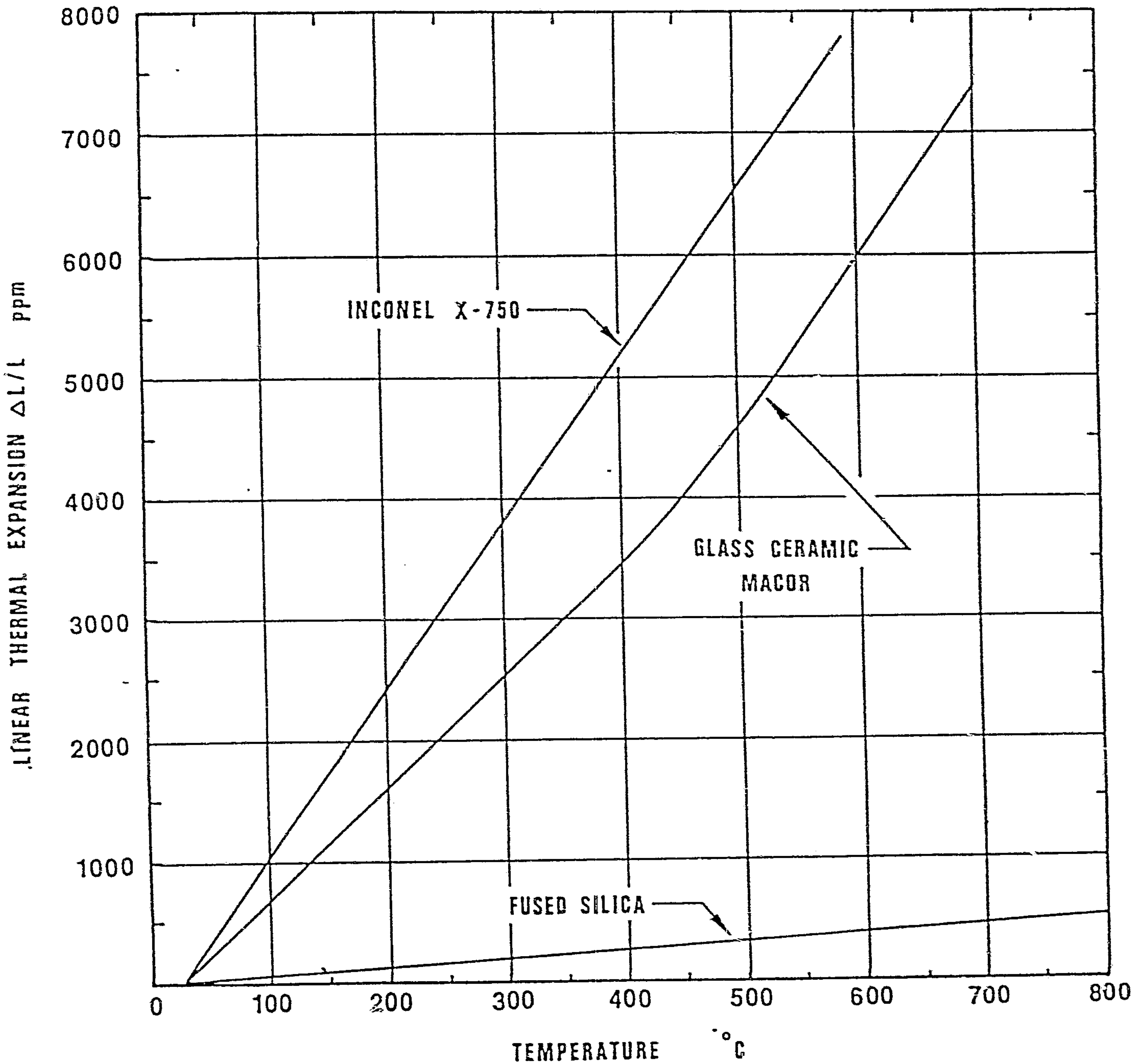


Fig. 2 Thermal Expansion of Fused Silica, Glass Ceramic Macor and Inconel X-750 as a Function of Temperature

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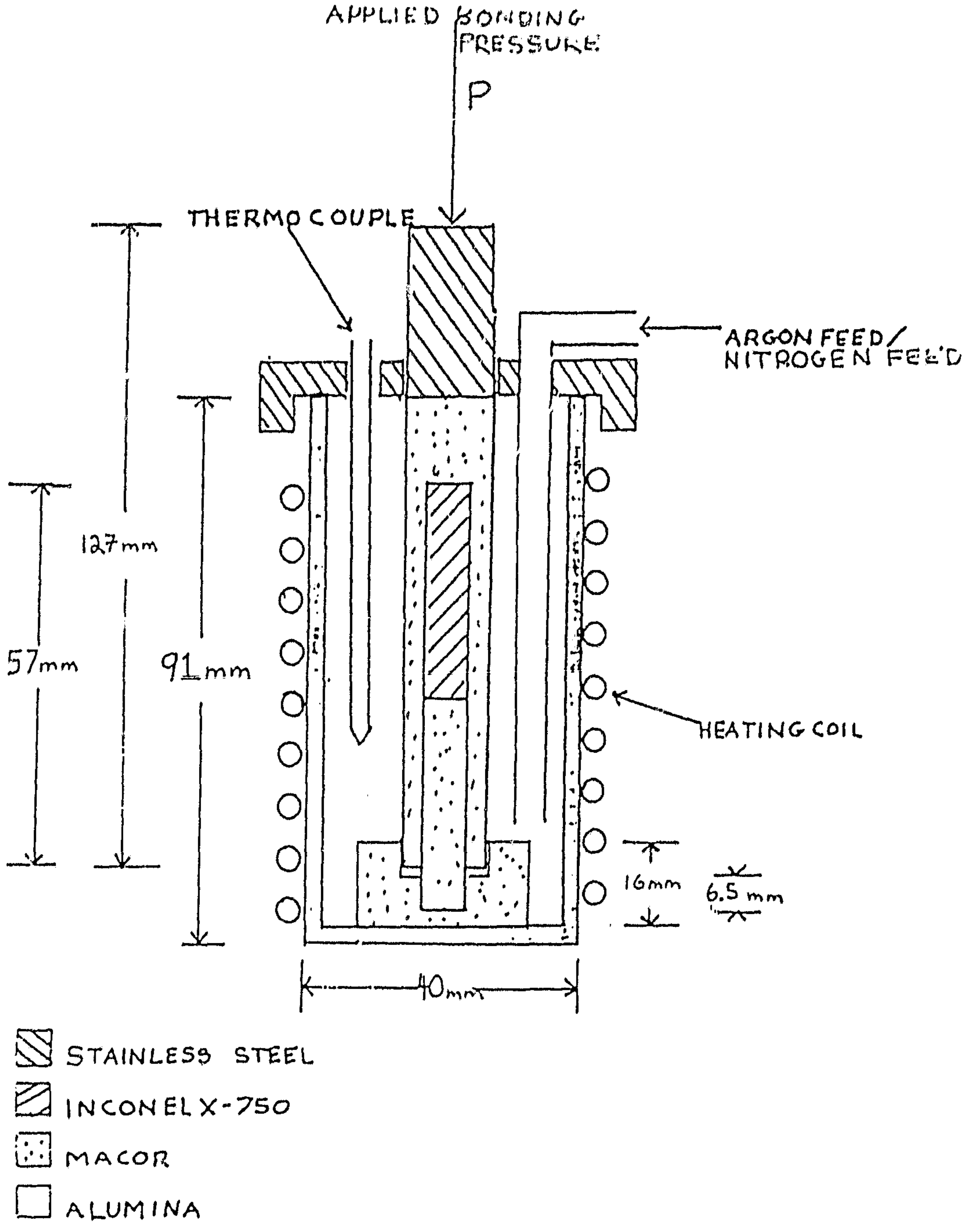

Fig. 3 - HIGH TEMPERATURE, INERT ATMOSPHERE, DIFFUSION SAMPLE PRESS.

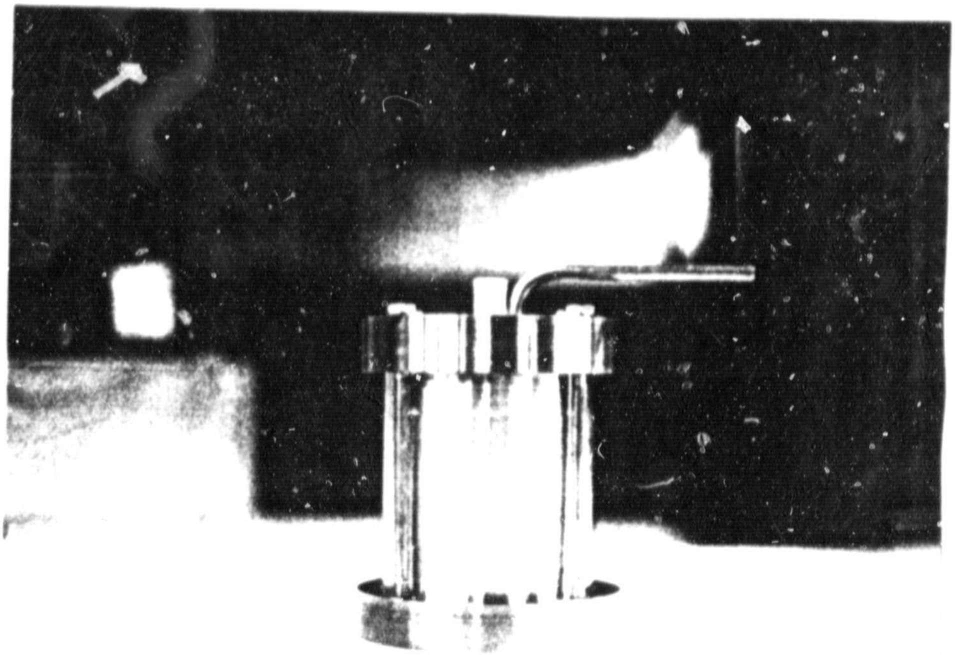

Fig. 4 - High Temperature, Inert Atmosphere, Diffusion Sample Press, side view

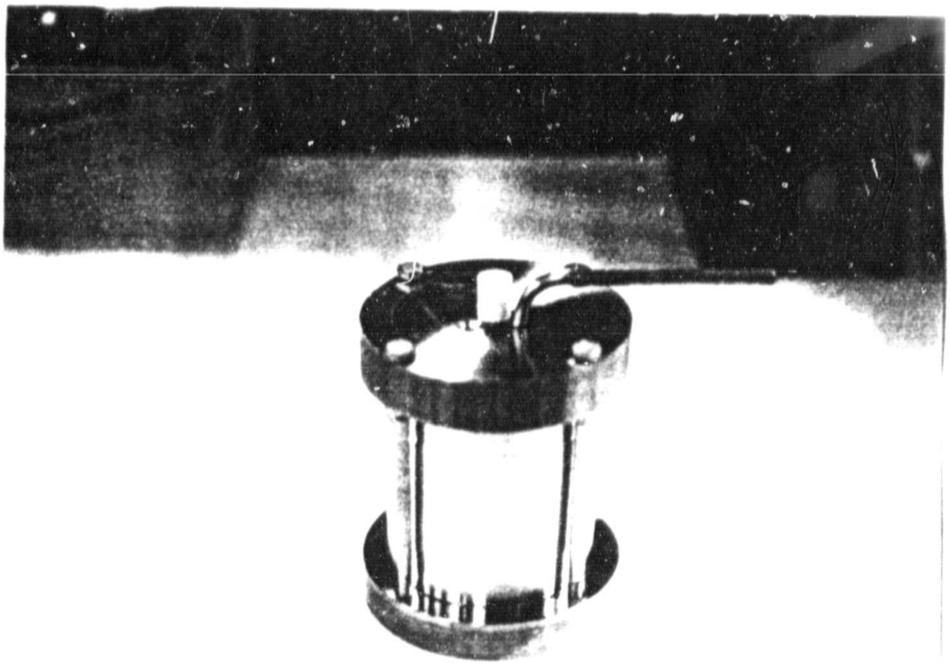

Fig. 5 - Diffusion Sample Press, perspective view.

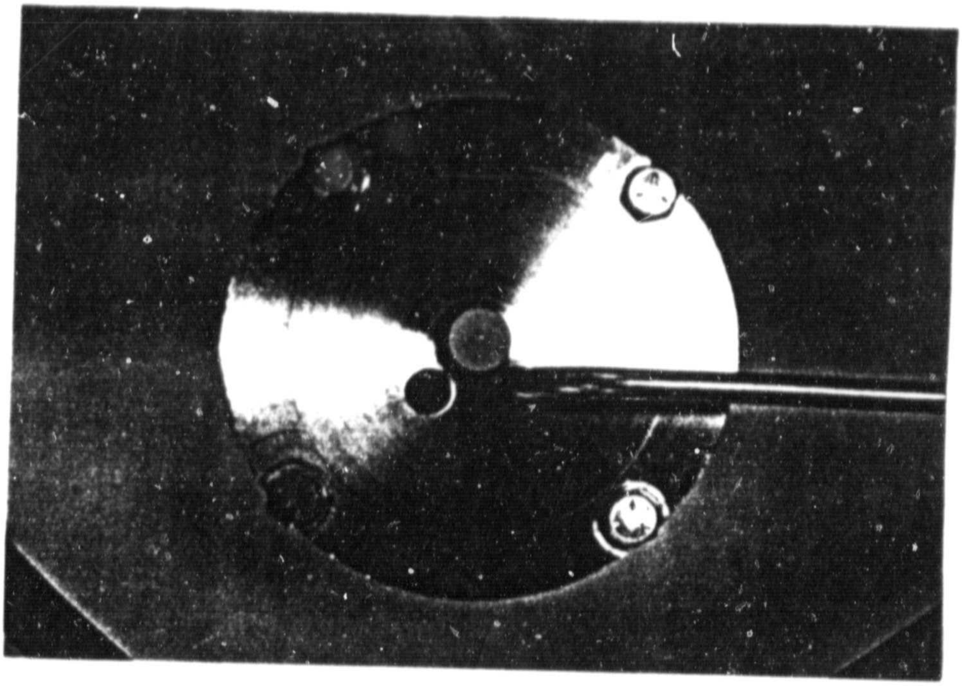

Fig. 6 - Diffusion Sample Press, top view

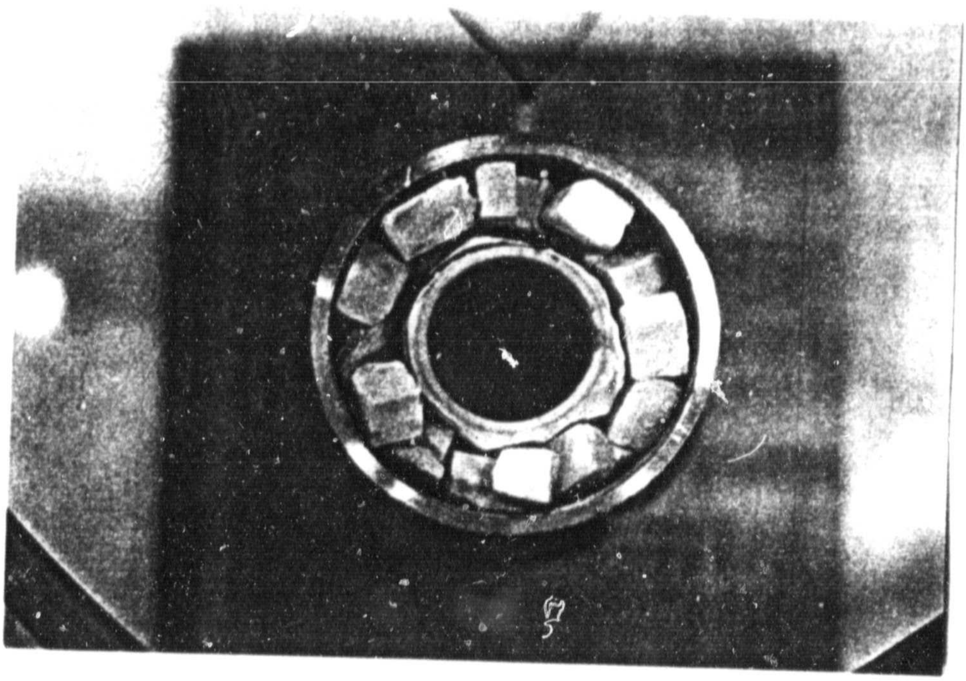

Fig. 7 - Diffusion Sample Press, internal view. The heating chamber and high temperature insulation are exposed.

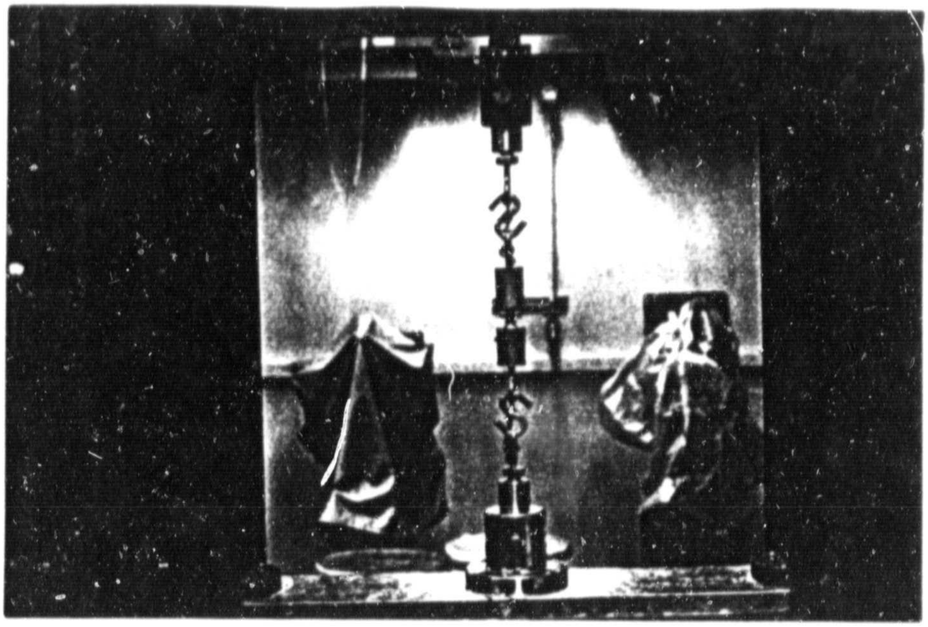

Figure 8. Assembly to measure tensile strength of ceramic-metal bond in tension

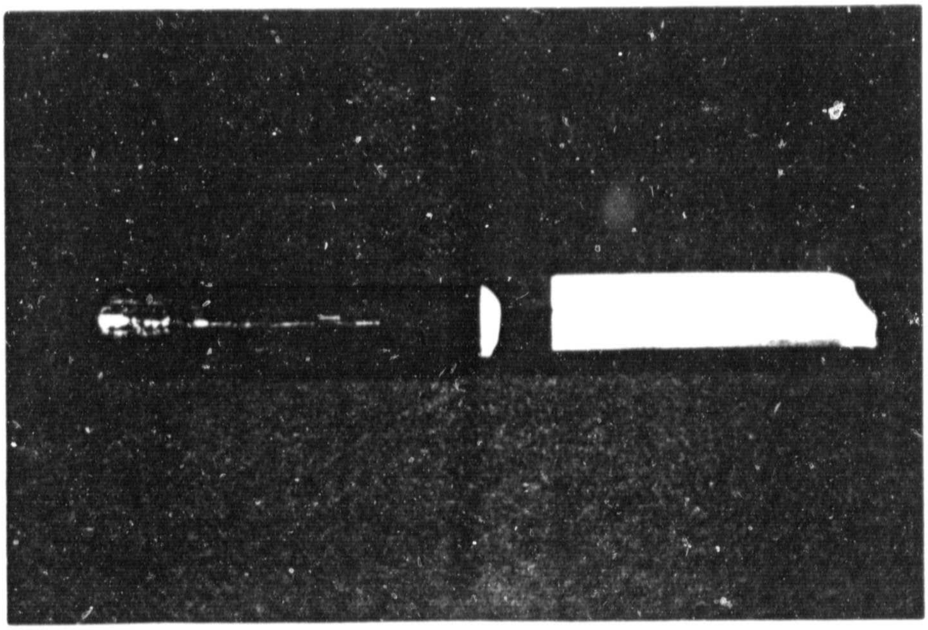

Figure 9. "MACOR" - Inconel bonded sample broken in tension

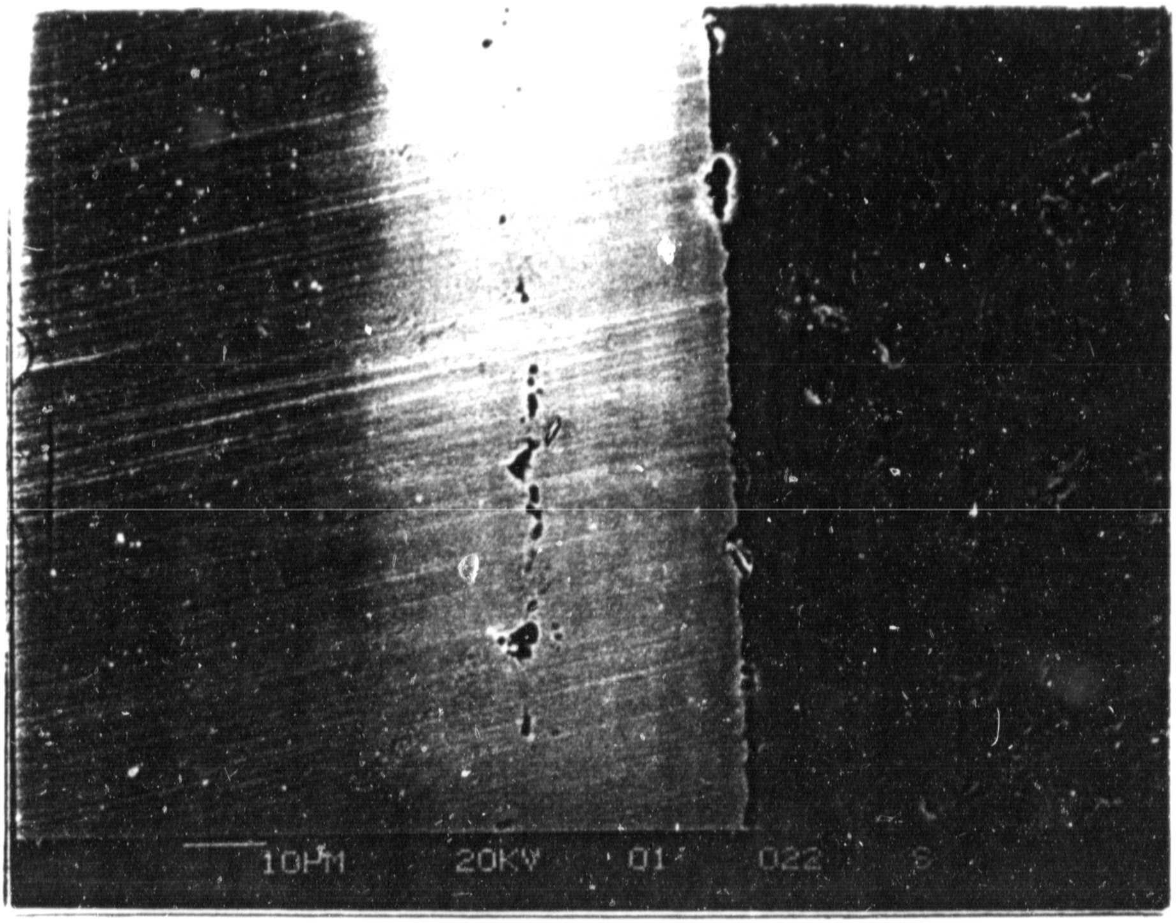

Figure 10. Inconel-gold-MACOR bonded regions showing Kirkendall voids in the gold layer

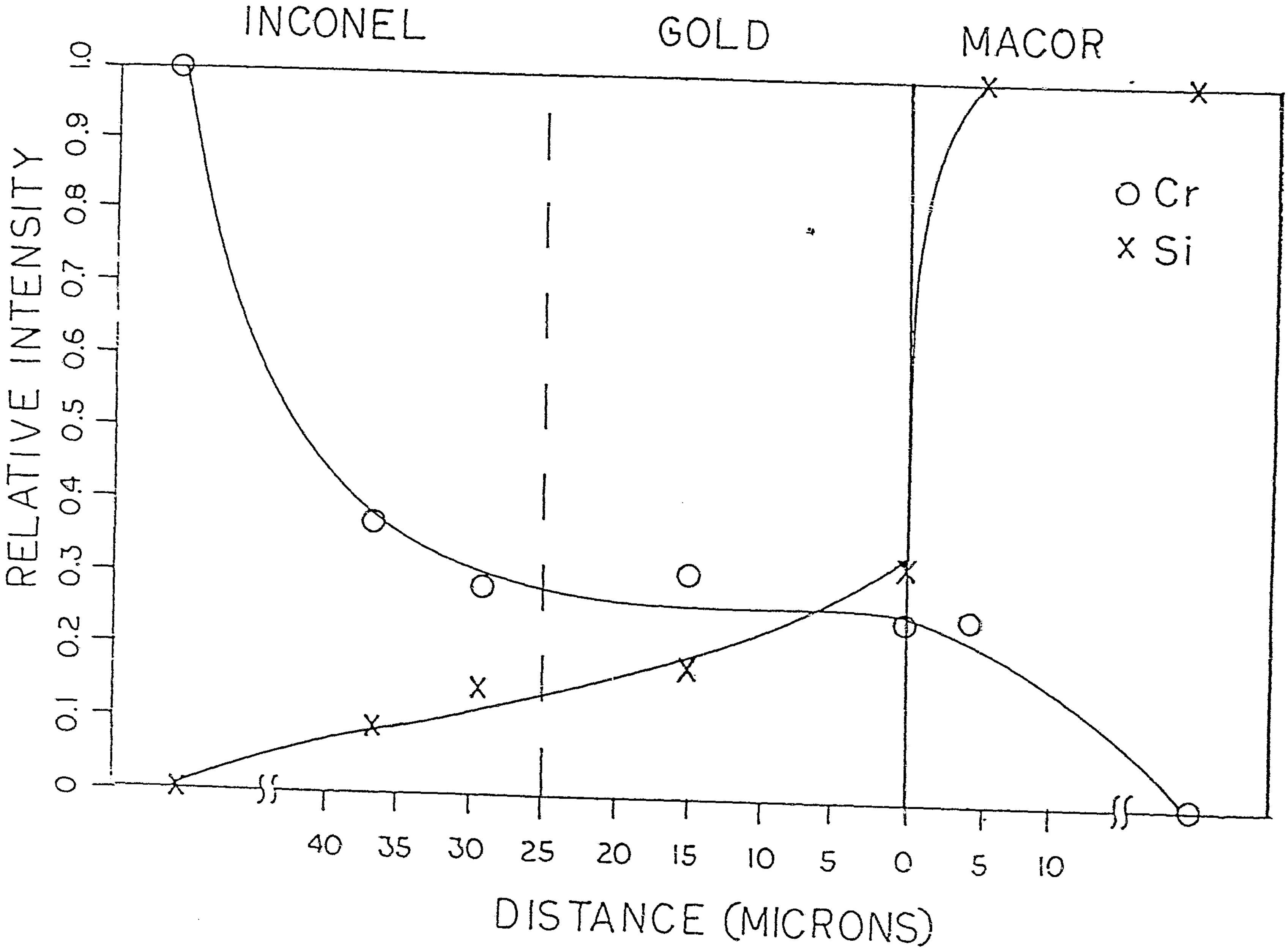

Figure 11. Profiles of Cr and Si concentrations in bonded region after 90 minutes at 950°C

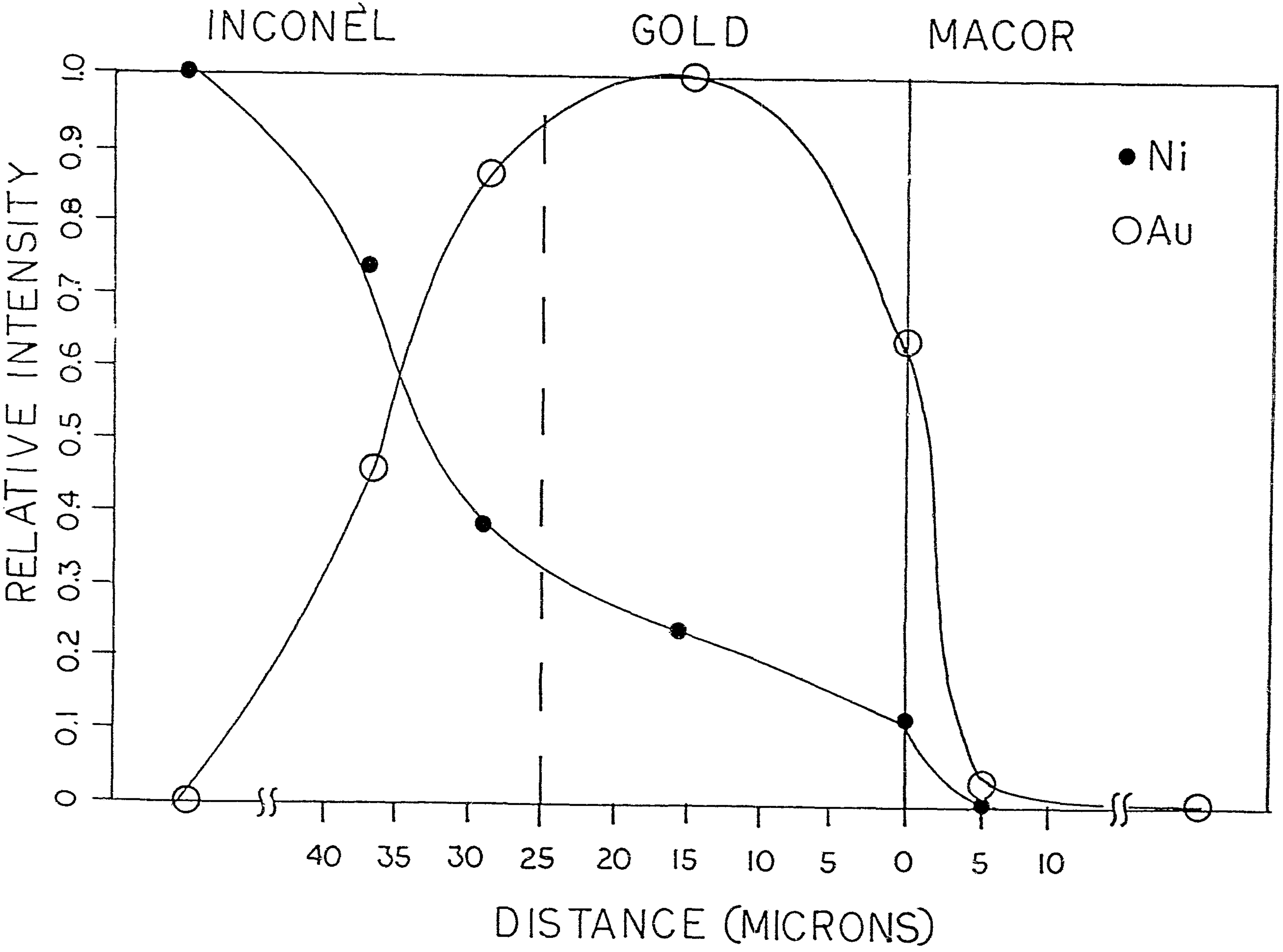

Figure 12. Profiles of Ni and Au concentrations in bonded ;ion after 90 minutes at 950°C

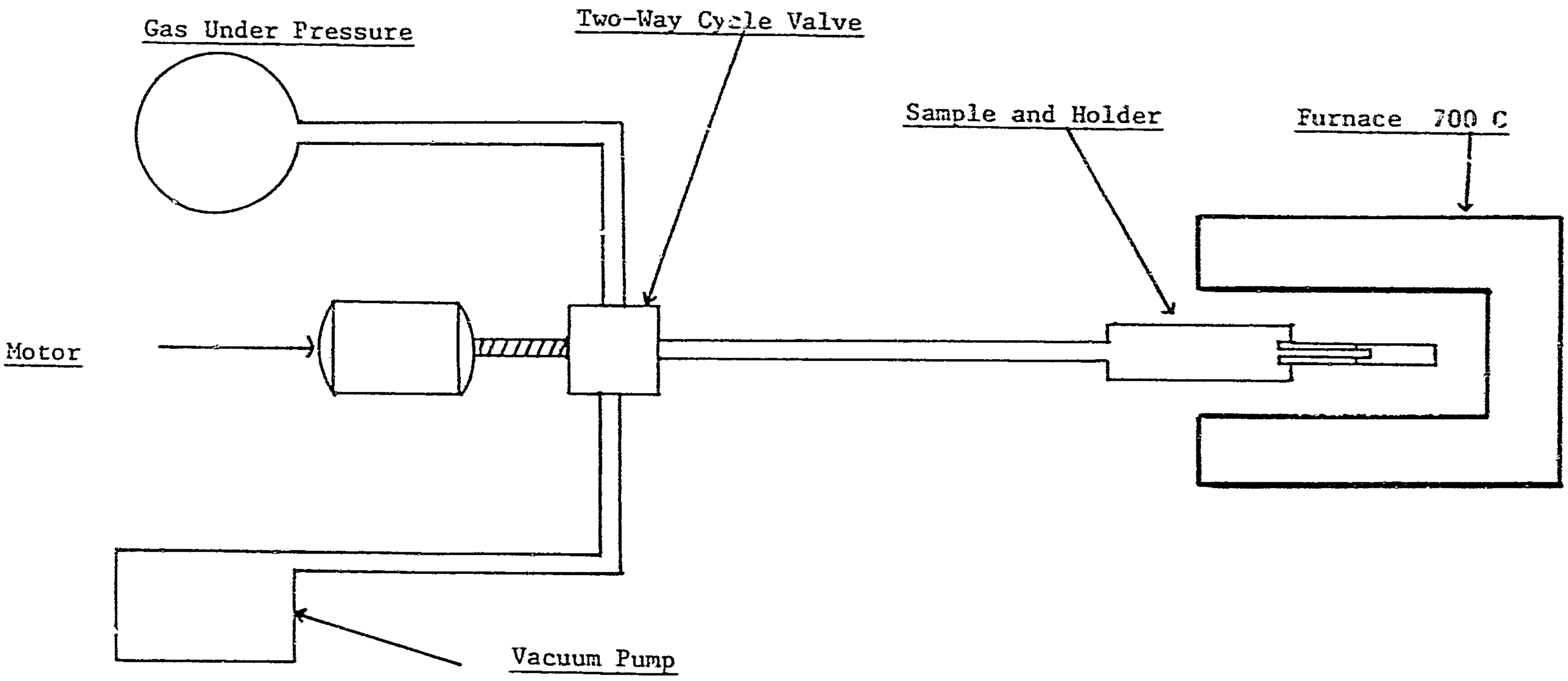

Fig. 13 : SCHEMATIC OF FATIGUE TESTING SYSTEM

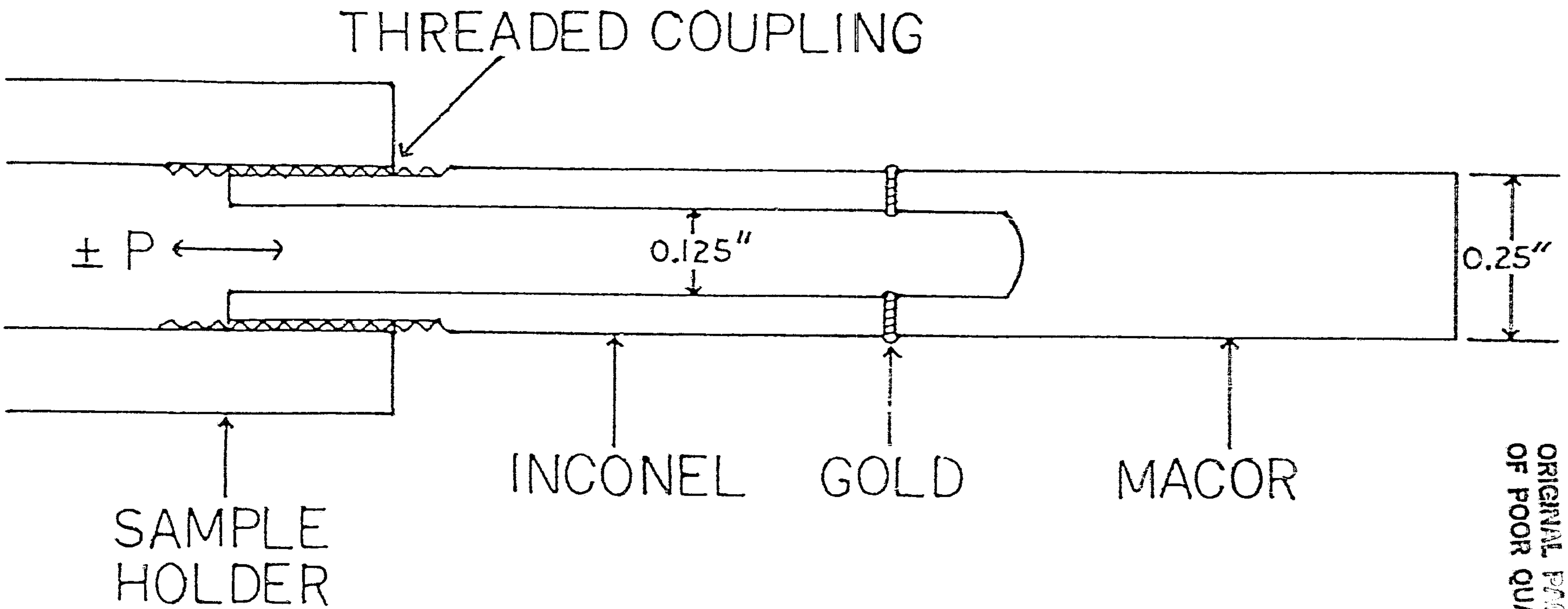

Figure 14. Details of the sample holder in the fatigue testing system of Figure 13.

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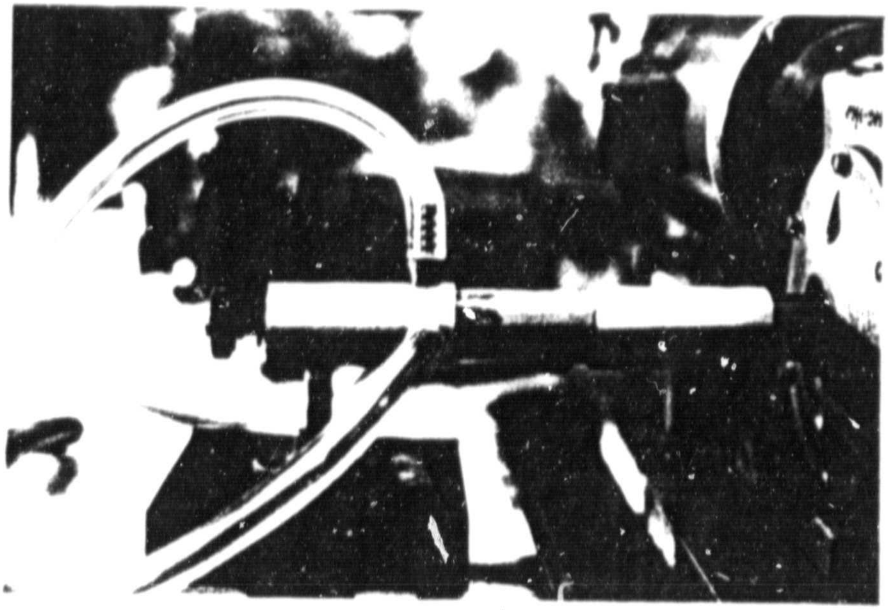

Figure 15. Photograph of fatigue testing system

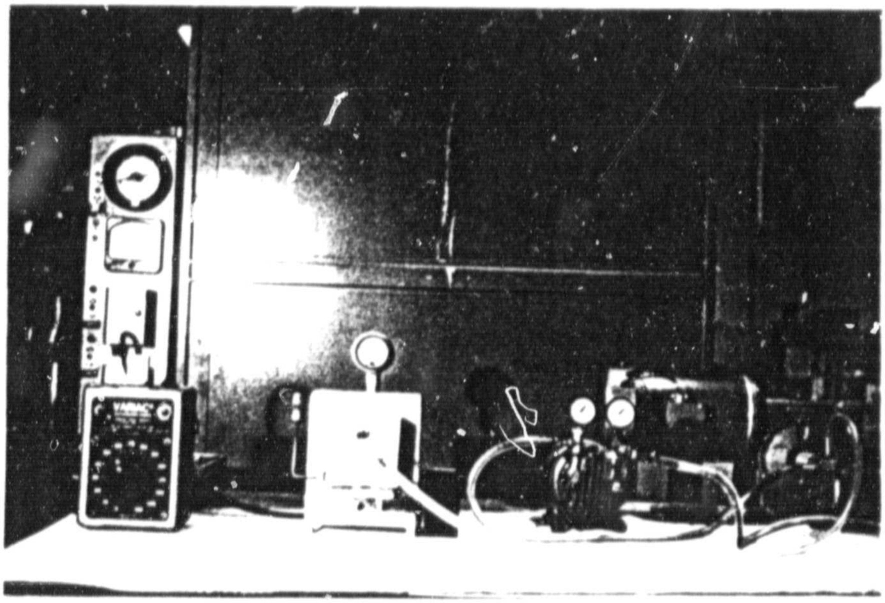

Figure 16. Two-way pressure valve of the fatigue testing equipment

Table 1. Appropriate references on Ceramic-Metal seals

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Table 2.

SUMMARY OF DIFFUSION BONDING TESTS WITH VARIOUS CANDIDATE SYSTEMS

SYSTEM	BONDING CONDITIONS	RESULT
MACOR / GOLD / INCONEL	950°C - 90 Mins.	MACOR SUCCESSFULLY BONDED TO INCONEL. DOME-SHAPED FRACTURE OBSERVED. STRENGTH OF 535 PSI.
SILICA / GOLD/ INCONEL	950°C - 60 Mins.	GOLD BONDED TO INCONEL. SILICA DID NOT BOND TO GOLD.
MACOR / ALUMINUM / INCONEL	650°C - 105 Mins.	NO BONDING BETWEEN ALUMINUM AND MACOR.
BORON NITRIDE / GOLD / INCONEL	920°C - 60 Mins.	NO BOND BETWEEN GOLD AND BORON NITRIDE.
GRADED DIFFUSION BOND: MACOR / GOLD / ASCOLOY / GOLD / INCONEL	850°C - 30 Mins. 900°C - 30 Mins.	ASCOLOY BONDED TO INCONEL. NO BOND BETWEEN ASCOLOY AND MACOR.

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Table 3. Summary of materials tested, their composition and supplies used

MATERIAL	SUPPLIER/MANUFACTURER	COMPOSITION
Silica	Heraeus Amersil 650 Jernees Mill Rd. Sayerville, NJ 08872	Impurities: Al 200 to 500 ppm; Sb 0.06 to 0.3 ppm; Ca 30 ppm; Cu 0.4 ppm; Fe 7.0 to 100 ppm; Ti 10 to 160 ppm
MACOR(tm)	Corning Glass Works Corning, NY 14830	Composition Proprietary
Inconel X-750	Fry Steel 13325 Molette St. Santa Fe Springs, CA 90670	Ni 71.82%; Cr 15.40%; Fe 7.99%; Ti 2.56%; Al 0.82%; Cb+Ta 0.81%; Mn 0.09%; Si 0.07%; C 0.03%; Co 0.02%; Cu l.t. 0.01%
Greek Ascoloy	High Temp. Metals 11541 Bradley Ave. San Fernando,CA 91340	Fe 81.19%; Cr 12.79%; W 2.68%; Ni 2.00%; Mn 0.42%; Mo 0.34%; Si 0.31%; C 0.16%; Cu 0.060%; P 0.018%; S 0.013%; Al 0.010%; Sn 0.010%; Pb 0.0003%
Gold Foil	Alfa Products Thiokol/Ventron Div. 152 Andover St. Danvers, Mass. 01923	Gold Foil 0.025 mm thick Metal Purity = 99.99%
Boron Nitride	Cerac Inc. Box 1178 Milwaukee, Wis. 53201	BN Purity = 97.5%
Silicon Nitride	Cerac Inc. Box 1178 Milwaukee, Wis. 53201	Si 60.4%; N 36-38%; Fe 0.30%; O 0.5-0.9%; Ca 0.1-1.0%; Total Others l.t. 0.5%

NOTE: All percentages expressed in weight percent.