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HIGH TEMPERATURE SEALS BETWEEN CERAMIC SEPARATION MEMBRANES AND SUPER-ALLOY HOUSING

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Abstract

One of the concepts for oxygen production from Martian atmospheric carbon dioxide involves the use of tubular electrochemical membranes for oxygen separation. The tubular configuration offers the advantage of being able to separate the oxygen at pressures of up to 500 psi, thereby eliminating the need for a "pre-liquefaction" oxygen compressor. A key technology that has to be developed in order for the electrochemical separator to combine as a compressor is a high temperature static seal between the ceramic separation cell and the nickel-based super-alloy tube. Equipment has been designed and fabricated to test the seals. Efforts are under way to develop a finite element model to study the thermal stresses at the joints and on the seal, and the optimal shape of the seal. The choice of seal materials and the technique to be used to fabricate the seals are also being investigated.

Problem Statement

Preliminary experiments are being conducted to evaluate the feasibility of concentrating O_2 from the electrochemical decomposition of gaseous CO_2 using a solid oxide electrochemical cell (ECC) leased from Ceramatec, Inc, Salt Lake City, Utah. A schematic of the ECC is shown in figure 1. The separation membrane, composed of an electrolyte sandwiched between electrode coatings, has a tubular structure. The membrane tube is enclosed in a concentric nickel-based super-alloy tube. Since ceramics (electrolyte material is ceramic) have excellent compression strength but are weak under tension, the outside of the ceramic tube (oxygen side) can be subjected to fairly high pressures (up to 500 psi). Hence, if the O_2 produced is liquefied and stored for later use, the configuration shown in figure 1 has the advantage over a disk system in that an O_2 compressor can be eliminated. The energy required to compress the O_2 is provided to the system by way of the potential applied across the membrane. The mimimum voltage required is given by:

$$E_{Nernst} = \frac{RT}{nF} \ln (P_2/P_1)$$

where E_{Nernst} = Pumping Voltage

R = Ideal Gas Constant

T = Absolute Temperature

n = number of electrons per mole of the reactant

F = Faraday Constant

P_1 = Partial Pressure of O_2 on the inlet side and

P_2 = Partial Pressure of O_2 on the exit side

In figure 1, the inlet stream is CO_2 flowing at room temperature. This gas is heated as it flows through the feed tube by means of clam-shell heaters that wrap around the super-alloy tube. The reason for not preheating the gas is that the seal between the ceramic membrane and super-alloy tube is rated only for a maximum operating temperature of 150°C. Efficient design of an electrochemical membrane calls for seals between the ceramic cell and the outer metal tube that can withstand the high temperatures. Such seals would permit the design of once-through multi-tube modules that would look similar to a shell and multi-tube heat exchanger in configuration.

State-of-the-Art

Ceramic joining and sealing technology is quite well developed for low-temperature applications. However, the technology for joining and sealing ceramics and metals for high-temperature, high stress, and/or corrosive environments is still in its infancy. In the area of brazing with filler metals, efforts have been made to specially formulate filler metals that will wet and adhere to untreated ceramic surfaces (direct brazing). Another approach has been to vapor coat the ceramic surface with a suitable metal layer prior to brazing (indirect brazing). A totally different approach has been to use specially formulated glasses for brazing. All three techniques have had some success for specific applications. While it is possible to calculate the stresses induced in the joints and the seal due to the differences in coefficients of thermal expansion, no systematic and rigorous analysis has been found in the literature.

<u>Approach</u>

An experimental facility has been designed and fabricated for testing the seals. Figure 2 shows a schematic of the facility. It has the capability of heating the test seal to temperatures ranging from 600$_{o}$C to 1300°C. The seal can be pressurized to 600 psi. A Validyne variable reluctance type differential pressure transducer has been calibrated and installed to monitor pressure drops as low as 0.1 psi. The pressure and temperature sensors and controllers are connected to an IBM compatible personal computer via a Data Translation analog-to-digital conversion board. An electrically activated valve shuts the system down and turns off the supply gas when the loss of pressure (due to a leak in the seal) exceeds a predetermined value.

A PC-based finite element code will be developed to analyze the stresses induced in the seals and joints due to the difference in the coefficients of thermal expansion between the metal, ceramic material, and filler material.

The code will permit the selection of the optimum shape and thickness of the seal. Since the specialty filler materials used for brazing are generally expensive, such an optimization would reduce the cost of the seals significantly. Presently, efforts are also under way to identify the appropriate filler materials between the two tubes and the fabrication shops that have the technical expertise to do the brazing.

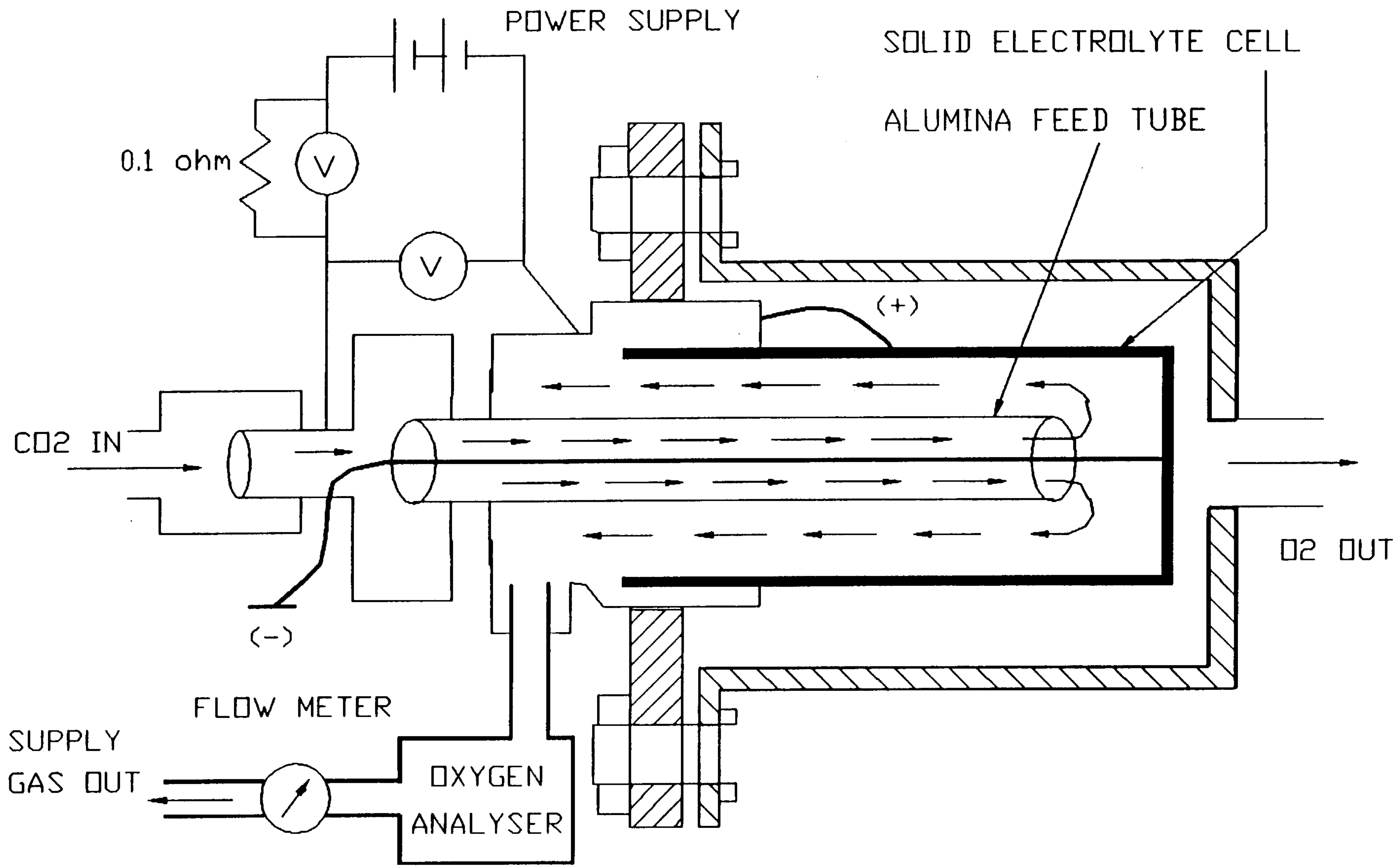

Fig 1: Schematic of The CERAMATEC Tubular Device

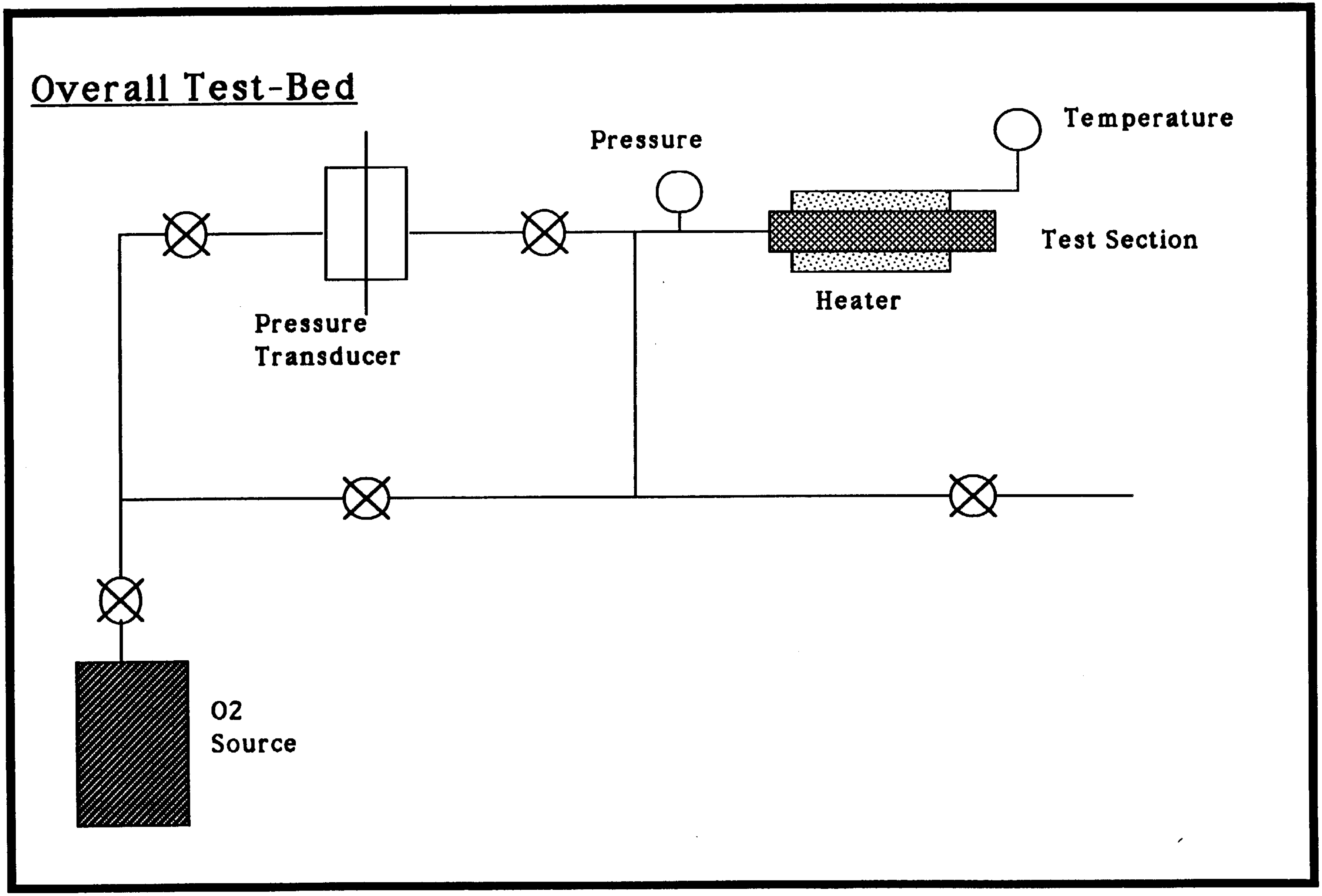

Fig 2: Schematic of The Experimental Facility for Testing High Temperature Seals

III. RESOURCE DISCOVERY AND CHARACTERIZATION