



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<p>(21) International Application Number: PCT/US90/06423 (22) International Filing Date: 6 November 1990 (06.11.90) (30) Priority data: 432,390 6 November 1989 (06.11.89) US (71) Applicant: CERAMATEC, INC. [US/US]; 2425 South 900 West, Salt Lake City, UT 84119 (US). (72) Inventors: JOSHI, Ashok, V. ; 3377 South 2410 East, Salt Lake City, UT 84105 (US). NACHLAS, Jesse, A. ; 1324 Westminster Avenue, Salt Lake City, UT 84105 (US). STUFFLE, Kevin ; 223 East Southgate, Salt Lake City, UT 84115 (US).</p>		<p>(74) Agents: BOND, Laurence, B. et al.; Trask, Britt &amp; Rossa, P.O. Box 2550, Salt Lake City, UT 84110 (US). (81) Designated States: AT (European patent), AU, BE (European patent), CA, CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), JP, LU (European patent), NL (European patent), NO, SE (European patent).  <b>Published</b> <i>With international search report.</i></p>
<p>(54) Title: SOLID STATE OXYGEN COMPRESSOR</p> <p>(57) Abstract</p> <p>A solid-state, electrochemical compressor for compressing oxygen and other diatomic, ionizable gases is disclosed. An oxygen ion transport membrane such as a ceramic metal oxide, for example, zirconia, which conducts oxygen ions when subjected to an electrical potential is a component of such a compressor.</p>		

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## SOLID STATE OXYGEN COMPRESSOR

Background of the Invention

5           This invention relates to the electrochemical  
compression of ionizable gases capable of electrochemical  
transport through a solid electrolyte and especially to  
the compression of oxygen. Pure oxygen has numerous  
applications. For instance, oxygen is used in large  
10 quantities in enrichment of blast furnaces, chemical  
synthesis, oxy-acetylene welding, life support and other  
medical uses. U.S. commercial consumption exceeds 18  
metric tons (20 million short tons) per year. Oxygen  
costs about 17.7 cents per cubic meter (5 cents per cubic  
15 foot) in small quantities, and about 10 cents per kilogram  
(\$15/ton) in large quantities. Currently, 99% (percent)  
of such oxygen is prepared by liquefaction of air and  
about 1% (percent) by electrolysis. Efficient storage and  
transport requires that oxygen be prepared at high  
20 pressure by mechanical compression. Mechanical  
compression of pure oxygen is a dangerous and inefficient  
operation due to the large positive Joule Thomson  
coefficient of oxygen. The present invention offers an  
alternative to mechanical compression of oxygen.

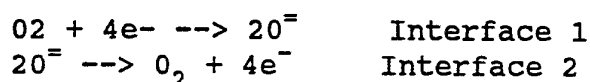
25           The present invention makes use of oxygen-ion  
conductive solids to prepare high pressure oxygen  
electrochemically. It has long been known that various  
solids such as zirconia, ceria, and bismuth-oxide will  
conduct oxygen ions when subjected to an electrical  
30 potential gradient across such solid oxides but will  
remain electronic insulators. Furthermore, some of these  
materials possess high strength.

Oxygen-ion conductive solids are currently used  
in numerous practical devices for power generation, oxygen  
35 partial pressure measurement and oxygen separation. In  
U.S. Patent 4,725,346 an oxygen delivery device is dis-  
closed. The specification of that patent is incorporated  
herein by reference.

An understanding of oxygen-ion transport in  
40 certain metal oxides may be facilitated by considering an

ion-conductive solid that separates oxygen at two different partial pressures which has a porous electronic conductor that is in contact with opposed surfaces of such ionic conductor at the gas-solid interfaces. This situation is depicted schematically in FIG. 1, which is an electrochemical cell. The ion conductive solid is the electrolyte and the electronic conductors are the electrodes. The half reactions occurring at each interface are:

10



The net chemical potential at each interface is the sum of the chemical potentials of the individual species at that interface. The electrolyte will conduct oxygen in order to move the system toward a state of equilibrium.

Equilibrium is achieved when the net chemical potential difference between reactants at one interface and products at the other interface,  $P$ , each times their stoichiometric coefficient, is zero. This is expressed mathematically by the following expression.

$$\sum v_i \mu_i = 0$$

$$2\mu_{\text{e},1} + \mu_{\text{O},1} = 2\mu_{\text{e},2} + \mu_{\text{O},2}$$

The net chemical potential difference of oxygen between interfaces 1 and 2 can be affected by controlling the electronic potential difference of electrons between interfaces 1 and 2. The chemical potential of oxygen at a gas-solid interface is direct function of oxygen partial pressure in the gas. Therefore, the oxygen partial pressure difference in the gas phase at the two interfaces can be affected by controlling the electronic potential difference.

35

#### Description of the Drawings

FIG. 1 is a schematic illustration of oxygen transport through an oxygen-ion conductive electrolyte;

FIG. 2 is a cross-sectional view of a solid state oxygen compressor illustrating one embodiment of the invention;

FIG. 3 is a graphical illustration of the rate of oxygen pressure increase achieved by the compressor illustrated in FIG. 2;

FIG. 4 is a cross-sectional view of a solid state compressor similar to that illustrated in FIG. 2 which includes a heater internal to the high pressure chamber;

FIG. 5 is a graphical illustration of the rate of oxygen pressure increase achieved by the compressor illustrated in FIG. 4;

FIG. 6 is a cross-sectional view of a solid state compressor similar to that illustrated in FIG. 2 in which dead volume has been minimized and illustrates another embodiment of the invention;

FIG. 6A is an enlarged sectional view of the end of the solid state compressor shown in Figure 6; and

FIG. 7 is graphical illustration of the rate of oxygen pressure increase achieved by the compressor illustrated in FIG. 6.

#### Description of Invention

An oxygen compressor utilizing a solid state oxygen ion transport membrane which transports oxygen ions when subjected to a voltage differential is disclosed. Particularly useful oxygen ion transport membranes are ceramic metal oxides such as zirconia, ceria, hafnia, bismuth oxide and the like. Such electrolytes are disclosed and described in U.S. Patent 4,725,346 infra, the description thereof being incorporated herein by reference.

The present invention consists of an ion-conductive solid constructed in such a way as to create a mechanical barrier between a low pressure oxygen reservoir and a confined volume. The barrier and confined volume

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-3a-

must be capable of supporting high pressure gas (most particularly oxygen). Each side of the ion-conductive

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solid (electrolyte) is coated with an electronically  
conductive material (electrode) to form a substantially  
continuous coating that is either porous or pervious to  
oxygen, i.e. it has appreciable oxygen solubility and  
5 diffusivity. This material is applied in such a way that  
a uniform electronic potential can be maintained  
throughout each electrode layer and so that a uniform  
electronic potential difference can be maintained across  
the ion- conductive solid. The electronic potential is  
10 maintained by means of an electrical power supply and lead  
wires. This is greatly facilitated if the ion conductor  
is an electronic insulator. Transport of oxygen will  
occur from the reservoir to the confined volume. The rate  
of oxygen transport is directly proportional to the  
15 overall chemical potential difference between each  
interface. High partial pressures of oxygen can be  
achieved in the confined volume since transport of oxygen  
ions will occur until chemical potential difference  
generated by the oxygen between the low pressure reservoir  
20 and the high pressure gas in the confined volume exactly  
cancels the applied electromotive force (EMF).

Most oxygen-ion conductive solids are metal  
oxides, a class of ceramic materials, such as zirconia,  
hafnia, bismuth oxide and the like. The chief solid  
25 material that is used as an oxygen ion conductor is  
stabilized zirconia.

Stabilized zirconia has a combination of  
properties that makes it well suited for application in  
practical devices. These include high oxygen ion  
30 conductivity, especially at temperatures greater than  
800°C, high strength, and sinterability which facilitate  
ease of fabrication of useful shapes, such as tubes, cups,  
cylinders, open-ended cylinders, flat plates and the like.  
The essence of the art in this field of technology is net  
35 shape fabrication techniques, processing parameters,  
microstructure control, type and amounts of dopants to  
control conductivity, electrode materials, electrode

application techniques and processing parameters, electrode morphology and many other details of design.

#### Example 1

5 An electrochemical oxygen compressor was fabricated as depicted in FIG. 2. A closed end stabilized zirconia (92 mole % (percent)  $ZrO_2$  - 4 mole % (percent)  $Y_2O_3$  - 4 mole % (percent)  $Yb_2O_3$ ) tube 0.95 centimeters outer diameter x 0.67 centimeters inner diameter x 24.13 centimeters long  
10 (0.375 inches outer diameter x 0.266 inner diameter x 9.5 inches long) was used as the electrolyte. About 11.4 centimeters (4.5 inches) of the length of the zirconia tube was coated inside and out successively with lanthanum strontium manganite and silver to form the electrodes. A  
15 silver lead was coiled around the external electrode to form the external lead wire. Another silver wire was laid along the internal wall of the tube to form the internal lead wire. The open end of the zirconia tube was secured into the top flange 11 of an Inconel pressure vessel 12  
20 (internal volume of 0.4 liters) using a compression fitting. The top flange 12 was bolted into a fixed flange 12a welded to the pressure vessel 12. The internal diameter of the zirconia tube was exposed to the atmosphere by a hole in the top flange into which the  
25 compression fitting was welded. A teflon ferrule was used to create the seal. The internal surface of the pressure vessel was lined with alumina insulation 13. A heater 14 was placed along the internal diameter of the zirconia tube. This heater was fabricated by coiling nickel-chrome  
30 wire around an 0.31 centimeters outer diameter (0.125 inches outer diameter) alumina tube 15 and coating with alumina cement. The alumina tube 15 also served as an air inlet and preheater. Air was pumped in through the alumina tube and out through the annulus between the  
35 alumina tube and the zirconia tube. The zirconia tube was heated to approximately 800°C. The air inside the pressure vessel reached 137,900 newtons per square meter



(twenty psig) due to heating. When the temperature reached 800°C a potential of one volt was applied across the electrolyte such that the outside surface of the zirconia tube was positive. Oxygen was conducted through the zirconia tube until a pressure of 344,750 newtons per square meter (fifty psig) was reached external to the zirconia tube. Pressure increase developed inside the vessel as a function of time is plotted in FIG. 3.

10

## Example 2

The oxygen compressor of Example 1 was modified by installing a heater that was external to the zirconia tube as depicted in FIG. 4. A sterling silver wire was coiled in contact with the internal diameter of the electrolyte tube to serve as the lead wire. The compressor was operated in the same manner as in example 1 except that the pressure caused by heating was released to atmospheric pressure before applying the potential across the electrolyte. The pressure inside the vessel was brought from 0 newtons per square meter (0 psig) to 1,172,000 newtons per square meter (170 psig) in approximately 570 minutes. The pressure increase developed as a function of time is plotted in Figure 5.

25

## Example 3

An electrochemical oxygen compressor was fabricated as depicted in FIG. 6. The design of the compressor was changed to minimize the volume of the design. As in the two previous examples, a closed end stabilized zirconia (92 mole % (percent)  $ZrO_2$  - 4 mole % (percent)  $Y_2O_3$  - 4 mole % (percent)  $Yb_2O_3$ ) tube 0.95 centimeters outer diameter x 0.67 centimeters inner diameter x 24.13 centimeters long (0.375 inches outer diameter x 0.266 inches inner diameter x 9.5 inches long) was used as the electrolyte. The pressure vessel was designed so that the internal wall of the pressure vessel was close fitting to the zirconia tube. In this case a

-7-

furnace was placed around the outside of the pressure vessel in order to heat the electrolyte to 800°C. The bottom 19.1 centimeters (7.5 inches) of the zirconia tube was coated inside and out successively with lanthanum  
5 strontium manganite and silver to form the electrodes. Silver wires were coiled around the electrodes to act as lead wires. The zirconia tube was secured into the top flange of the pressure vessel (internal volume of 0.025 liters) using a compression fitting. The internal  
10 diameter of the zirconia tube was exposed to the atmosphere by a hole in the top flange into which the compression fitting was welded into. A teflon ferrule was used to create the seal. Air was pumped through an 0.31 centimeters outer diameter (0.125 inches outer diameter)  
15 alumina tube and out through the annulus between the alumina tube and zirconia tube. The zirconia tube was heated to 800°C. Internal pressure due to heating was released at this point. A potential of one volt was applied across the electrolyte. The vessel was  
20 pressurized to 1,379,000 newtons per square meter (200 psi) in approximately 18 minutes. The pressure as a function of time is plotted in FIG. 7.

In an oxygen compressor of the invention it is desirable to utilize an ion (oxygen ion) conducting  
25 electrolyte having excellent strength. Zirconia or hafnia are for this reason preferred electrolytes with zirconia especially preferred because of its strength and commercial availability. The zirconia should preferably be in the shape of an elongated cylinder. A cylindrical  
30 tube with one closed end and one open end is especially preferred.

Another feature of the invention is that the high pressure region is external to the electrolyte. Thus, the pressure acts on the external surface of the  
35 tube radially toward the longitudinal axis of the tube. While it is common in metal systems to contain high pressure inside the smallest diameter member, the tendency

of ceramics to fail in tension militates against containing high pressure internally in a ceramic tube.

Placing the high pressure region external to the zirconia may require a heavier walled metal container  
5 (shell), however, this is more desirable than making the electrolyte with a very thick tube wall. Electrical resistance of the electrolyte increases with electrolyte wall thickness, thus diminishing the electrical efficiency of the compressor.

10 Ceramics generally are much stronger in compression than in tension, thus a cylindrical electrolyte tube of a certain diameter and wall thickness will sustain a greater pressure on its external surface. External pressure puts the cylindrical wall under  
15 compression, thus taking advantage of the electrolytes compressive strength which is much greater than its tensile strength.

A feature of the present solid state compressor is a low ratio of the pressure chamber volume to  
20 electrolyte area.

The quantity of oxygen transported through a particular electrolyte is proportional to current for a given temperature. In oxygen compressors it is desired to achieve the operating pressure as rapidly as possible.  
25 Thus, providing a small annular space between the interior of the pressure shell and the exterior of the electrolyte causes a rapid buildup of pressure within the pressure chamber.

The compressor illustrated in FIG. 6 is  
30 particularly effective in rapidly reaching a desired high-pressure output.

The high pressure creates some back EMF which at a pressure of 13,790,000 newtons per square meter (2000 psi) amounts to about 115 millivolts. Thus, at an  
35 operating voltage of one volt or more the back EMF is minimal.

Although the present invention has been described in detail in terms of oxygen compression, the techniques and devices described herein may be utilized generally in the compression of ionizable diatomic gases capable of electrochemical ion transport through a solid electrolyte. Such other transportable gases include hydrogen, chlorine, fluorine and the like. Hydrogen may be readily ionized and transported through proton conductors such as barium cerate, hydrogen uranyl phosphate, strontium cerate, etc.

Similarly, chlorine and fluorine gases may be readily ionized and transported through chlorine ion conductors such as  $\text{SrCl}_2\text{-Al}_2\text{O}_3$  and fluorine ion conductors such as  $\text{LaF}_3$  or  $\text{PbF}_2$ .

These proton conductors may be readily substituted for the oxygen ion transporting electrolytes described hereinabove and illustrated in the attached drawings.

Although water is generally not considered a gas, both oxygen and hydrogen may be extracted from water vapor. Under appropriate conditions, water vapors disassociates into hydrogen and oxygen. In the presence of an oxygen ion-transporting electrolyte, oxygen may be separated and using the techniques of the instant invention, readily compressed. For example, serial compression of oxygen and hydrogen may be readily accomplished. Water vapor at an elevated temperature may be introduced into an oxygen compressor of the type described herein to produce high pressure oxygen and a by-product water vapor stream rich in hydrogen. This by-product stream may then be introduced into a hydrogen compressor utilizing a proton conducting electrolyte to yield high pressure hydrogen and vent gas of water vapor. Alternatively, the byproduct stream from the oxygen compressor could be fed to a condenser to condense the water vapor, recover pure hydrogen which could be

mechanically compressed, especially after the hydrogen is passed through a drier to remove any residual moisture.

Hydrogen compression by disassociation of hydrogen, either in pure form or in combination with other  
5 gases, is preferably done at temperatures in the range of 50° to about 1000°C for the following proton conductors: phosphate (50-100°C), barium cerate (500-1000°C).  
Electrodes suitably used in conjunction with such proton  
conductors are palladium, lanthanum strontium chromite,  
10 platinum, silver and copper, lanthanum strontium manganite.

Although various electrodes may be utilized on the present invention such as porous platinum, porous or non-porous silver and similar metallic compositions, the  
15 preferred electrode system is composed of lanthanum strontium manganite (LSM). Multiple layers are preferably applied until an electrode thickness of about 20 microns to about 200 microns is achieved. These LSM electrodes are especially adherent to the electrolyte, have a  
20 coefficient of thermal expansion similar to that of the electrolyte and are generally unaffected by the oxidizing condition to which the electrodes are subjected.

Conductive ceramic electrodes other than LSM that are useful in the instant invention are lanthanum  
25 strontium chromite, strontium iron cobaltite and the like.

The sheet resistance of LSM electrodes tends to be higher than that of metal electrodes such as silver. An effective manner of distributing current throughout the whole area of the LSM electrode is to use a metal wire  
30 mesh in intimate contact with the electrode. One technique useful in the instant invention is to form a cylinder of nickel or Inconel wire mesh then force an LSM coated tubular electrolyte into the interior of the mesh cylinder. The metal mesh may be heated to an elevated  
35 temperature to cause expansion of the mesh before it is put over the electrolyte. Upon cooling, the mesh achieves intimate contact between the electrode and electrolyte by

means of a shrink-fit. An overcoat of electrode material such as silver or LSM is preferably added to ensure intimate secure contact between the mesh and the electrode. The internal electrode of a tubular  
5 electrolyte may be similarly structured by forcing a wire mesh cylinder into the electrolyte tube and then coating the mesh in a similar fashion to that done on the external electrode.

Ceria, especially ceria stabilized with calcia,  
10 strontia or yttria, may be readily substituted for zirconia. Lanthanum strontium cobaltite is an especially effective electrode for use with ceria.

Claims

We claim:

- 5           1.    A solid state electrochemical compressor  
for compressing an ionizable diatomic gas comprising:  
a solid tubular electrolyte which conducts ions of said  
                  gas through the electrolyte when said  
                  electrolyte is subjected to an applied  
10            electromotive force;  
a low pressure gas inlet communicating with the interior  
                  of said tubular electrolyte;  
electrodes pervious and/or porous to said ionizable  
                  diatomic gas, said electrodes being intimately  
15            in contact with opposed surfaces of said  
                  electrolyte;  
a high pressure chamber external to said tubular  
                  electrolyte;  
a high pressure gas outlet communicating with said high  
20            pressure chamber; and  
pressure restriction means in said high pressure gas  
                  outlet.
2.    The compressor of claim 1 wherein said gas  
25            is oxygen.
3.    The compressor according to claim 2 wherein  
the ion conductive electrolyte contains zirconia.
- 30            4.    The compressor of claim 1 according to  
claim 1, wherein the low pressure gas is air.
5.    The compressor according to claim 1,  
wherein the low pressure gas is essentially pure oxygen.
- 35            6.    The compressor according to claim 1,  
wherein the high pressure gas is essentially pure oxygen.

7. The compressor according to claim 1, where the oxygen-ion conductive solid electrolyte contains at least one additive selected from any of the following:

HfO<sub>2</sub>, CeO<sub>2</sub>, Bi<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, ThO<sub>2</sub>, Y<sub>2</sub>O<sub>3</sub>, Yb<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>.

5

8. The compressor according to claim 1, where the oxygen ion electrolyte is in proximity to a heater capable of maintaining said electrolyte in a temperature range from about 500 to 1300°C.

10

9. The compressor according to claim 1, where the electrodes contain any of the following lanthanum strontium manganite, lanthanum-strontium chromite, platinum, silver, gold, nickel, Inconel, nickel oxide,

15 palladium, copper.

10. The compressor according to claim 1, where the lead wires contain any of the following: silver, gold, palladium, platinum, Inconel, nichrome, nickel and

20 copper.



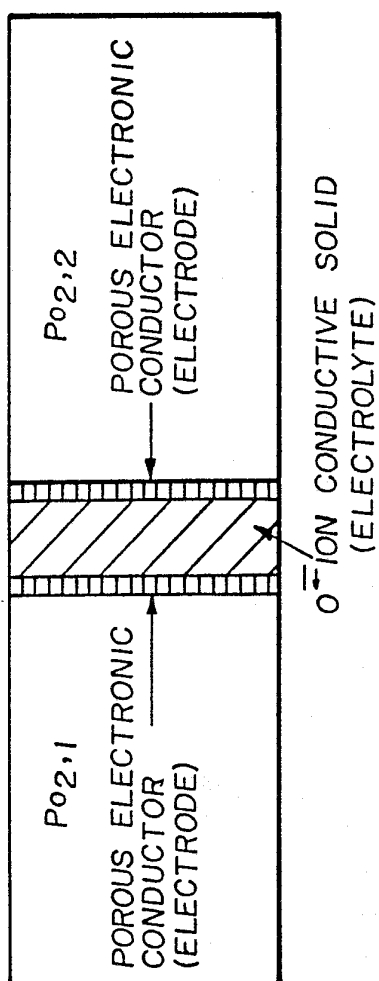


Fig. 1

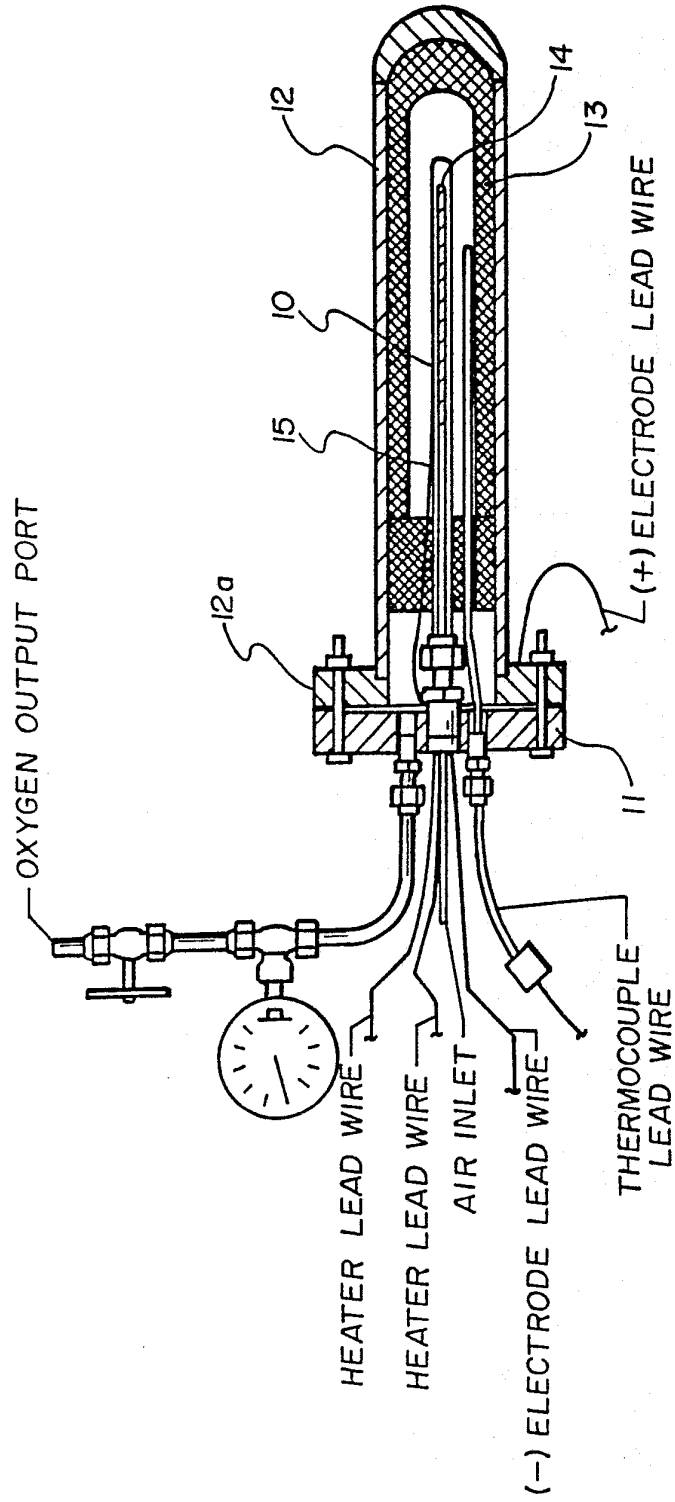
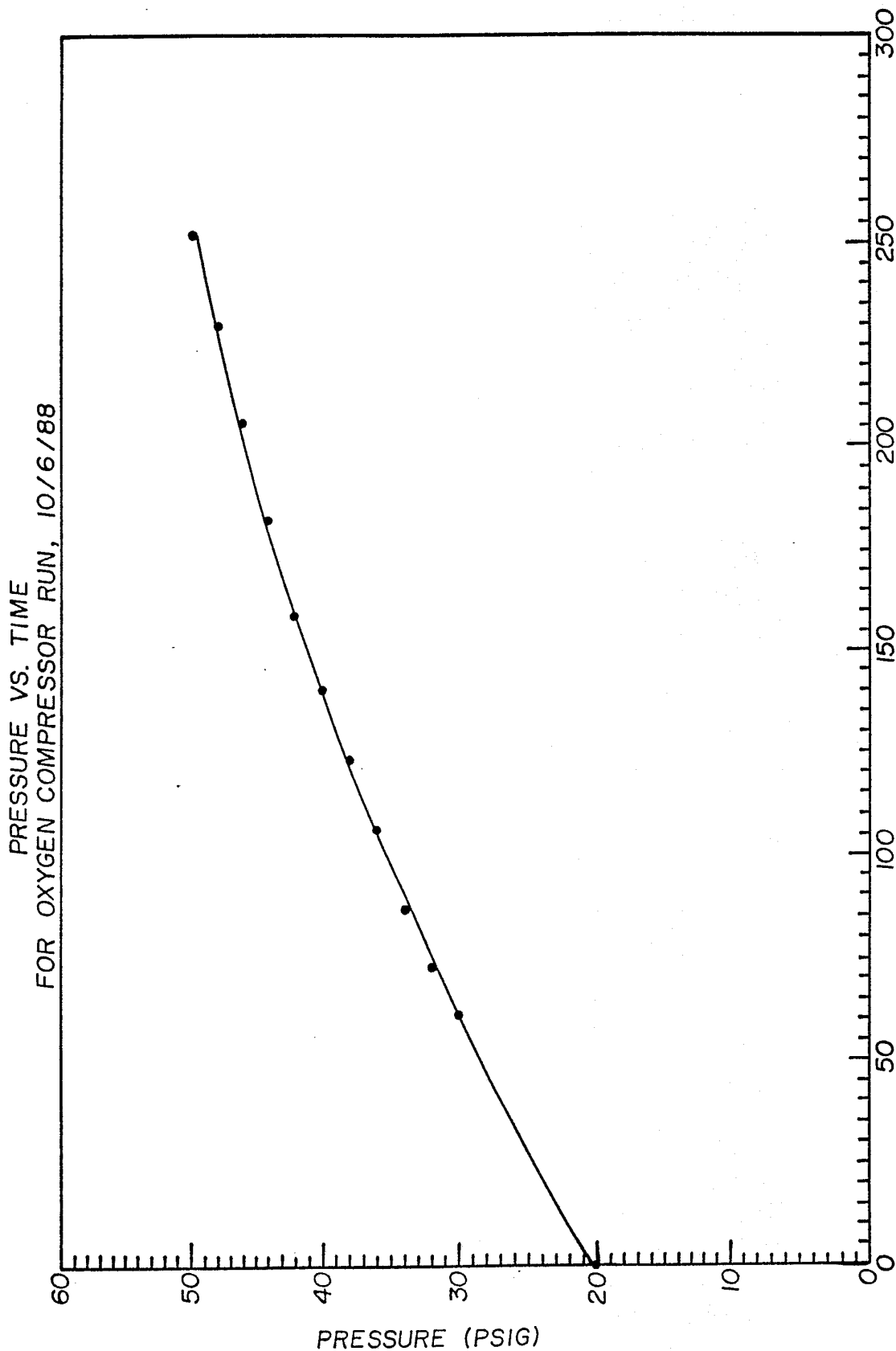


Fig. 2



TIME (MIN)

Fig. 3

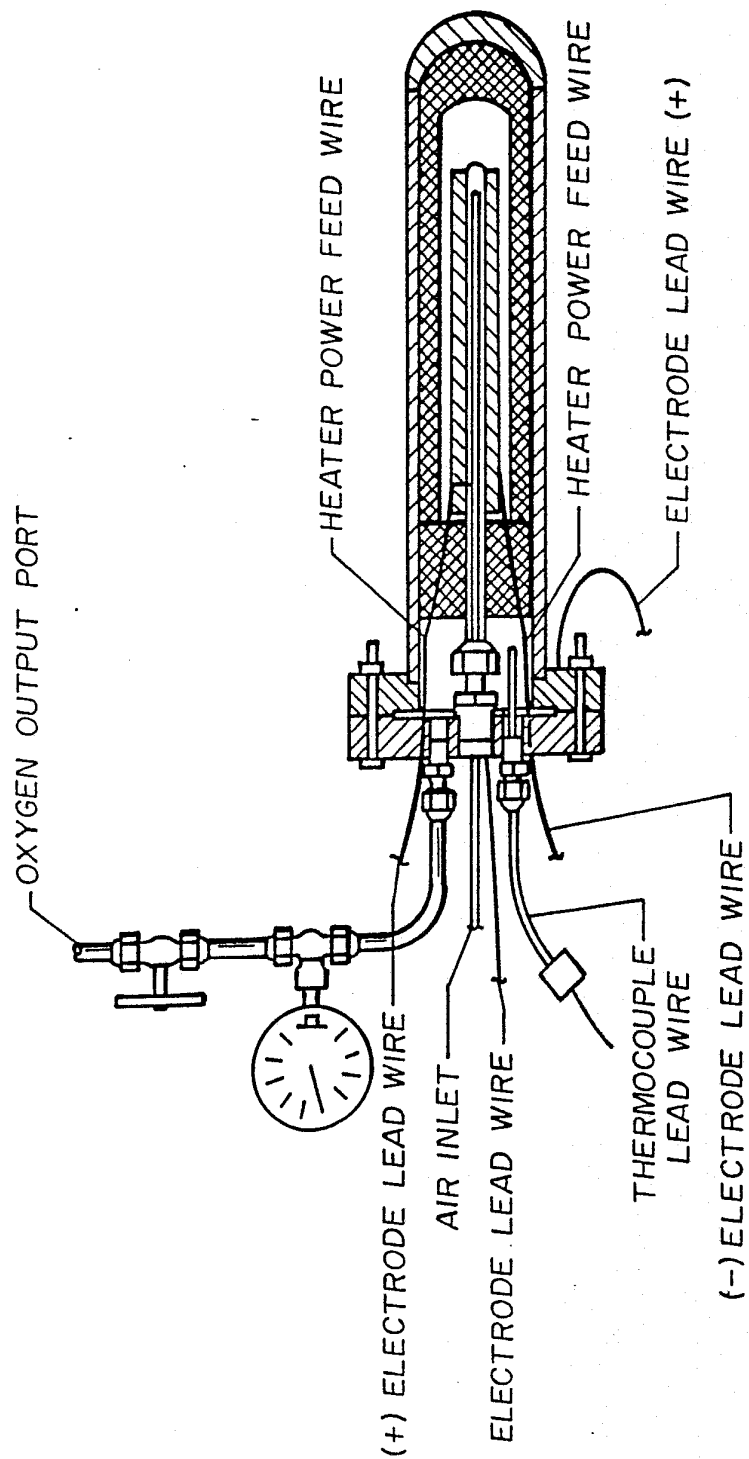


Fig. 4

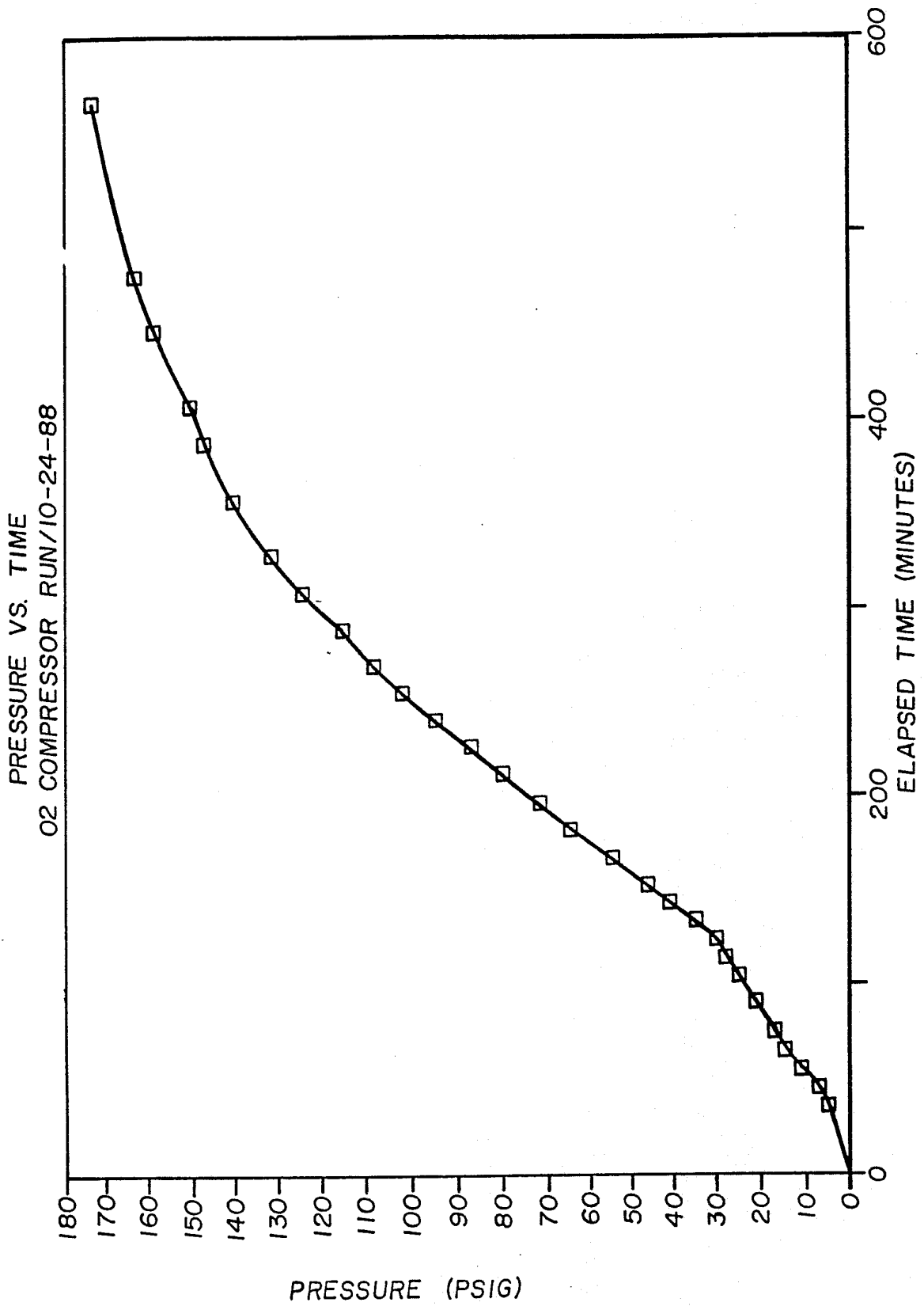


Fig. 5

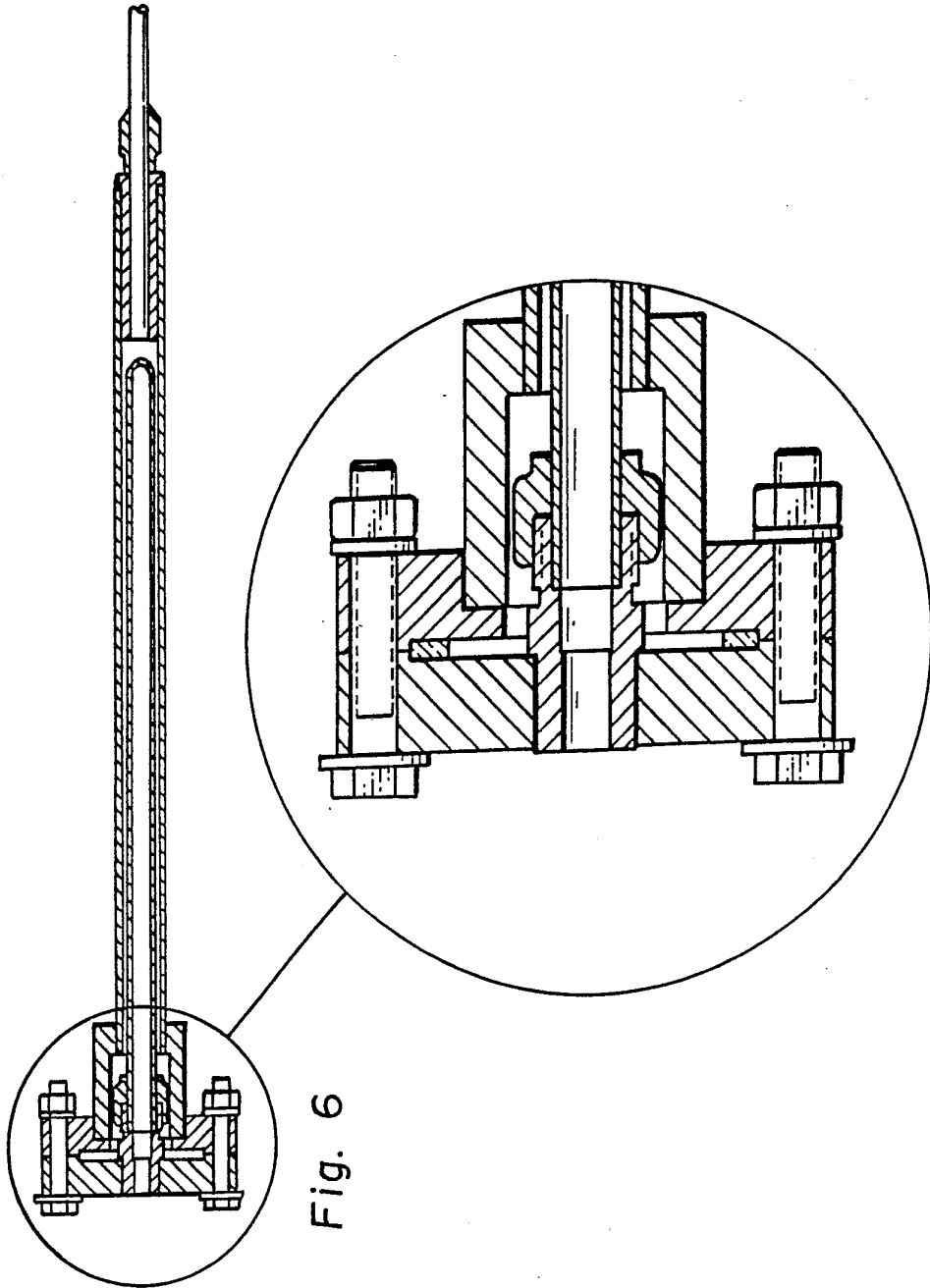


Fig. 6A

Fig. 6

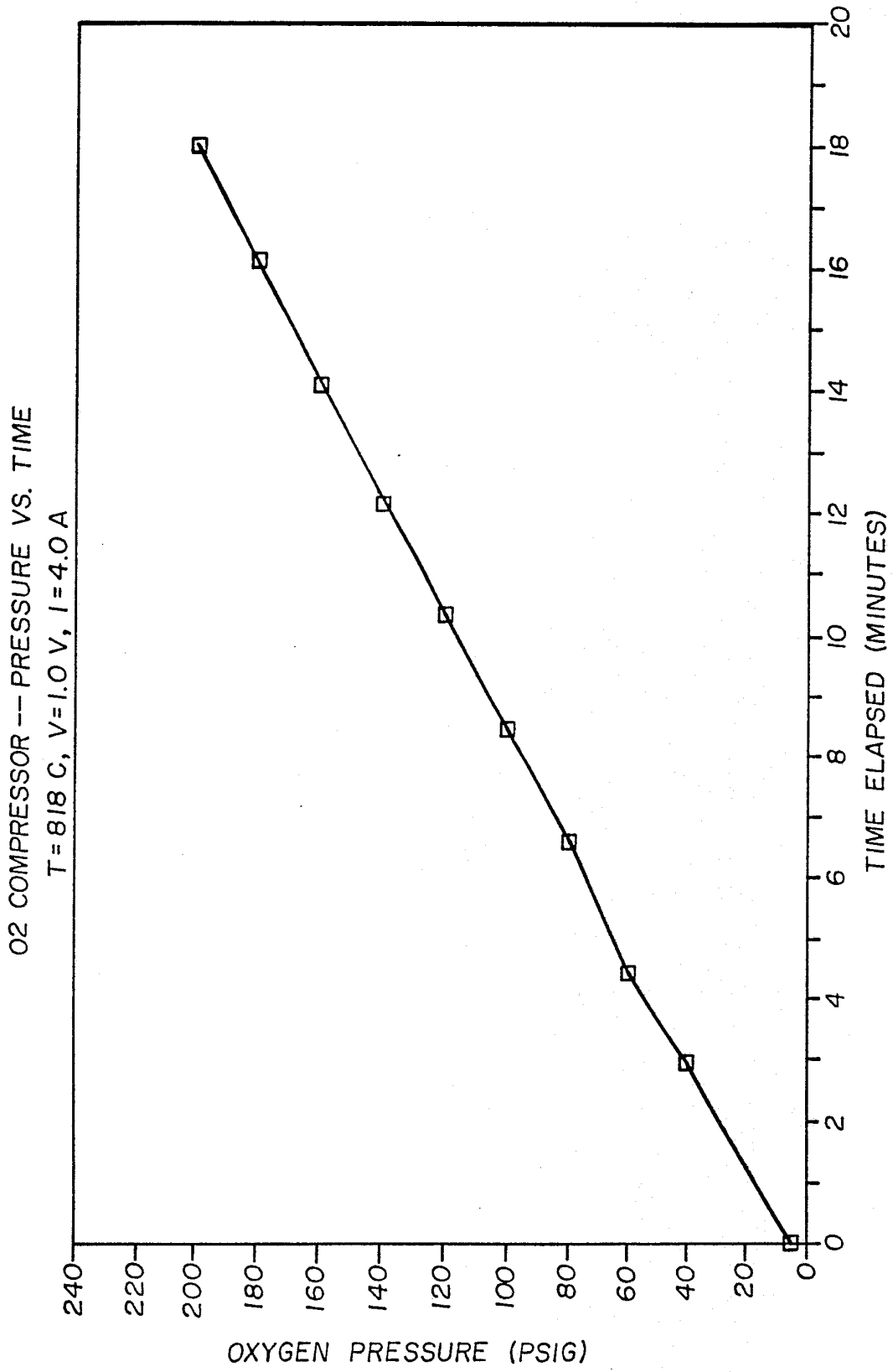
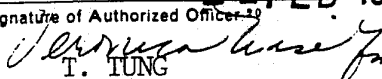


Fig. 7

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# INTERNATIONAL SEARCH REPORT

International Application No PCT/US90/06423

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>3</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC		
INT. CL(5): C25B 1/00		
US CL.: 204/130		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>4</sup>		
Classification System	Classification Symbols	
US	204/130, 153.18, 421, 422, 423, 424, 425, 426.427, 428, 429	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>5</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT</b> <sup>14</sup>		
Category <sup>6</sup>	Citation of Document, <sup>16</sup> with indication, where appropriate, of the relevant passages <sup>17</sup>	Relevant to Claim No. <sup>18</sup>
Y	US, A, Re 28,792 (RUKA et al.) 27 April 1976 See figure 6.	1-10
Y	US, A, 3,669,032 (RAPP) 17 October 1972 See figure 3.	1-10
Y	US, A, 3,838,021 (ARBITER) 24 September 1974 See the figure.	1-10
Y	US, A, 4,007,106 (HONE et al.) 08 February 1977 See column 3, line 58.	1-10
A	US, A, 4,725,346 (JOSHI) 16 February 1988 See figure 1.	1-10
Y	US, A, 4,752,361 (GAUTSCHI) 21 June 1988 See figure 1.	1-10
P,A	US, A, 4,879,016 (JOSHI) 07 November 1989 See figure.	1-10
<p><sup>15</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&amp;" document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search <sup>2</sup>	Date of Mailing of this International Search Report <sup>2</sup>	
18 JANUARY 1991	<b>14 FEB 1991</b>	
International Searching Authority <sup>1</sup>	Signature of Authorized Officer <sup>19</sup>	
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