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(71) Demandeur/Applicant:
AVANTIUM INTERNATIONAL B.V., NL

(72) Inventeurs/Inventors:
BOHM, SEBASTIAAN, NL;
VAN DEN BERG, ALBERT, NL;
VAN DER LINDEN, HEIKO JAN, NL

(74) Agent: FETHERSTONHAUGH & CO.

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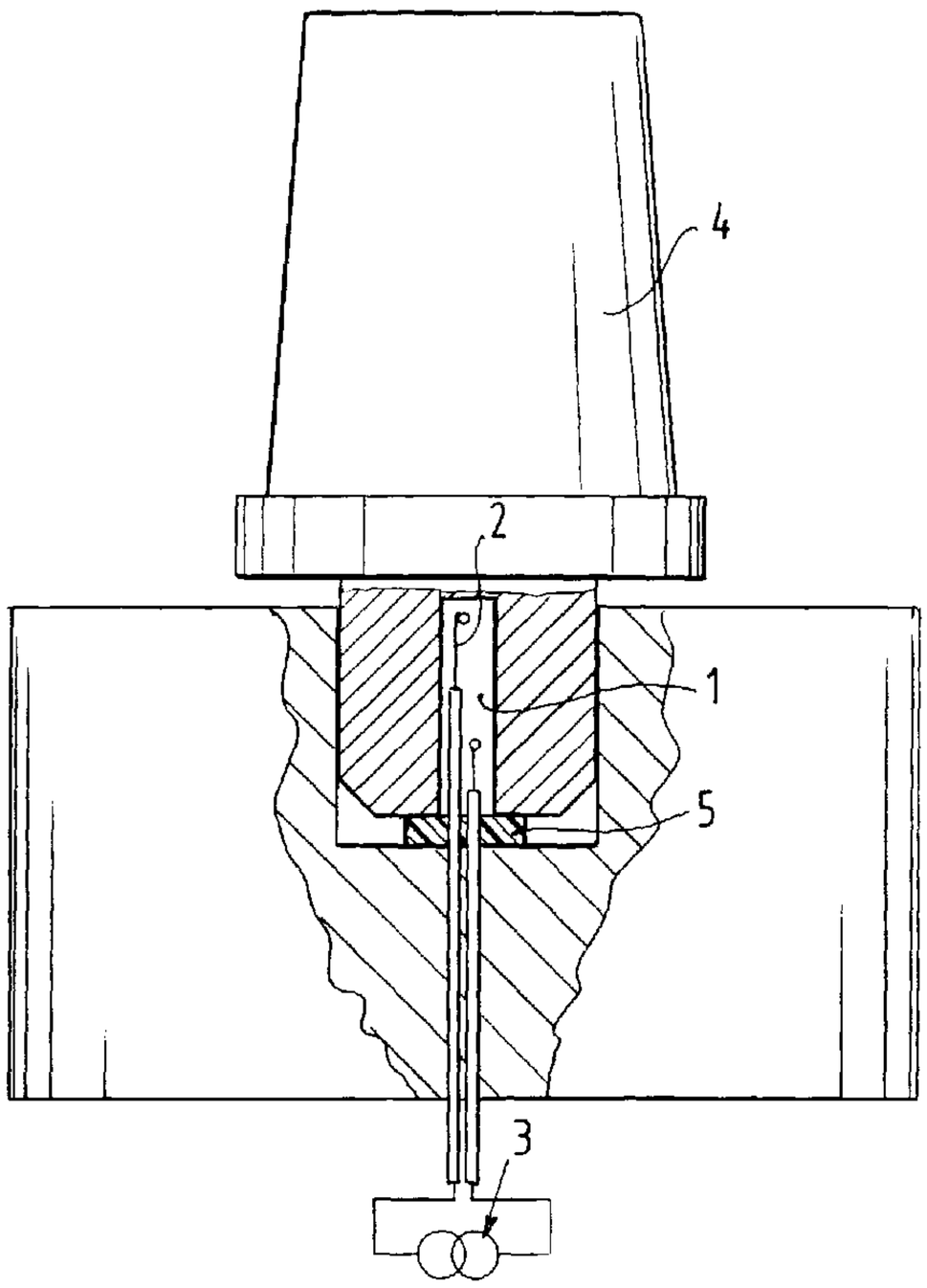
(54) Title: DEVICE AND METHOD FOR ELECTROCHEMICALLY GENERATING ONE OR MORE GASES



(57) Abrégé/Abstract:
The present invention relates to a device for electrochemically generating one or more gases under high pressure, comprising: a container (1, 6) comprising one or more chambers (7, 8) for filling with electrolyte; a first (2, 10) and a second (2, 11) electrode over which a voltage difference can be applied in order to bring about an electrochemical generation in the container (1, 6); wherein the content of a chamber (7, 8) is in the order of magnitude of a few millilitres or less.

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(71) Applicant *(for all designated States except US)*: **UNIVERSITEIT TWENTE MESA RESEARCH INSTITUUT** [NL/NL]; Drienerlolaan 5, NL-7522 NB Enschede (NL).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **BÖHM, Sebastiaan** [NL/NL]; Hanenberglanden 262, NL-7542 EH Enschede (NL). **VAN DEN BERG, Albert** [NL/NL]; Eversbergweg 3, NL-7443 PC Nijverdal (NL). **VAN DER LINDEN, Heiko, Jan** [NL/NL]; De Vluchtestraat 1-104, NL-7523 BE Enschede (NL).

(74) Agent: **VAN SOMEREN, Petronella, Francisca, Hendrika, Maria**; Arnold & Siedsma, Sweelinckplein 1, NL-2517 GK The Hague (NL).

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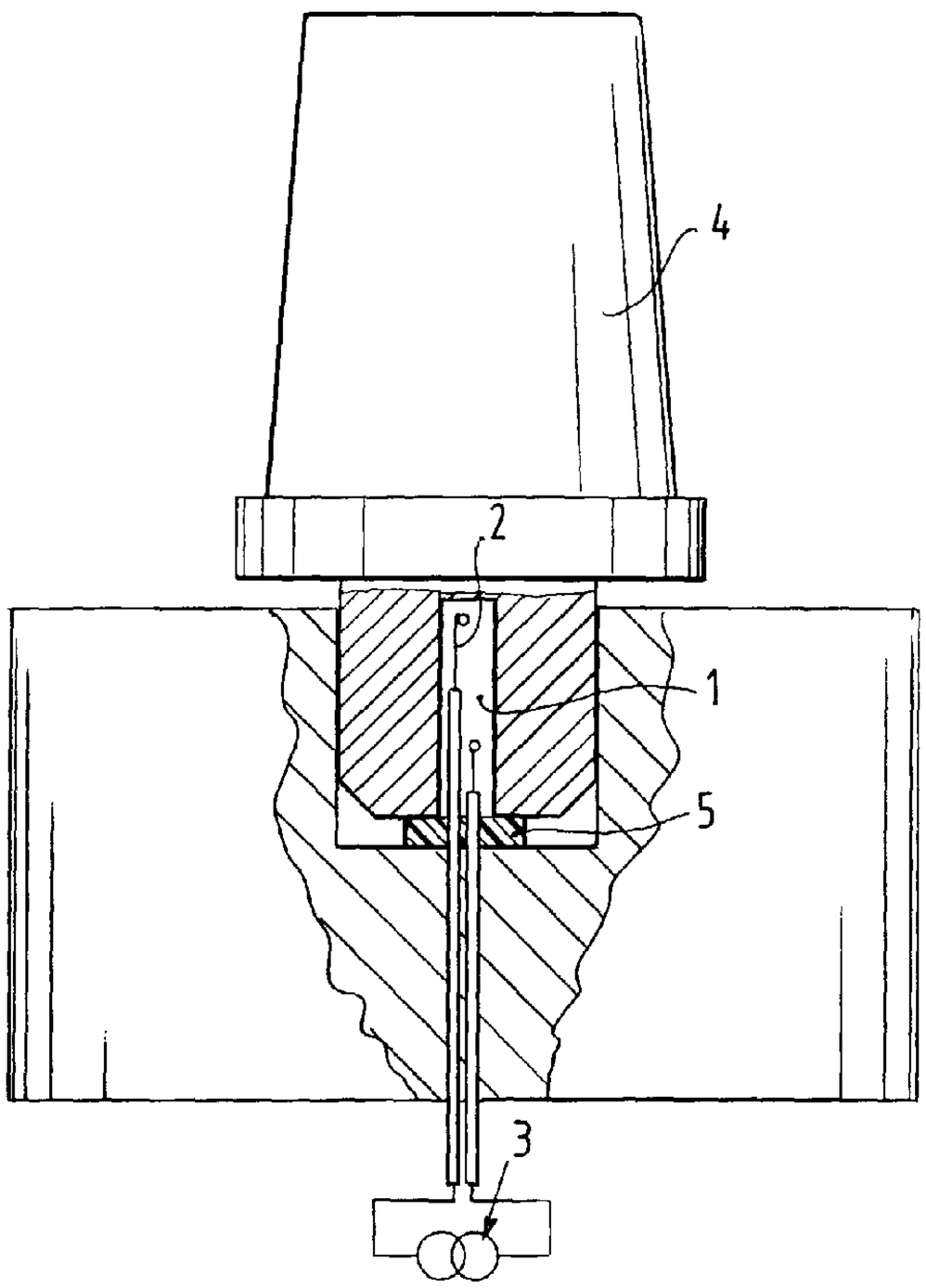
(54) Title: DEVICE AND METHOD FOR ELECTROCHEMICALLY GENERATING ONE OR MORE GASES



(57) Abstract: The present invention relates to a device for electrochemically generating one or more gases under high pressure, comprising: a container (1, 6) comprising one or more chambers (7, 8) for filling with electrolyte; a first (2, 10) and a second (2, 11) electrode over which a voltage difference can be applied in order to bring about an electrochemical generation in the container (1, 6); wherein the content of a chamber (7, 8) is in the order of magnitude of a few millilitres or less.

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DEVICE AND METHOD FOR ELECTROCHEMICALLY GENERATING ONE OR MORE GASES

The present invention relates to a method and device for electrochemically generating one or more gases under high pressure. The invention also relates to an integrated circuit on which a large number of above stated devices are applied and to a method for analysing the action of a catalyst.

For the performing and studying of physical or chemical processes under a high pressure use is made of so-called pressure vessels or "autoclaves". The attainable pressure in usual autoclaves is typically several hundreds to a maximum of a few thousands of atmospheres. A (high) temperature can further be set and controlled in such autoclaves.

A drawback of such an autoclave is the necessity of taking far-reaching and expensive safety precautions in respect of the explosion hazard, for instance in the form of a specially adapted freestanding laboratory with a loose roof. Stringent safety regulations also apply in respect of the construction and use of these autoclaves.

A further drawback is that the usual autoclaves are relatively large and therefore intrinsically slow in respect of variations in pressure, temperature and chemical potential, which makes the autoclaves difficult to control. The required amount of material, for instance catalyst, is moreover relatively large when a chemical reaction is analysed with this catalyst. The above-mentioned drawbacks have the consequence that the use of autoclaves is relatively time-consuming and costly. This has the result that only a limited number of measurements can be performed in practice.

A further drawback of the existing autoclaves is that, since the pressure build-up in the autoclave is effected slowly, the occurrence of particular chemical reactions requiring a rapid pressure build-up is impeded.

It is an object of the present invention to provide a device and a method in which the above stated drawbacks are obviated.

According to a first aspect of the invention a device is provided for electrochemically generating one or more gases under high pressure, comprising:

- a container comprising one or more chambers for filling with electrolyte;
- a first and a second electrode over which a voltage difference can be applied in order to bring about electrochemical generation in the container;

wherein the content of a chamber is in the order of magnitude of a few millilitres or less and the chamber preferably contains a maximum of 150 microlitres of electrolyte. With such a small content of the chamber or chambers the forces on the walls of the container are so low that no special safety provisions have to be made and the device is therefore intrinsically safe. Even in the case of a possible explosion of the gases in the container no appreciable damage will occur to the container and its surroundings. A device is hereby provided with which physical or chemical processes under high pressure can be performed and studied safely and with which a (large) number of experiments can be performed quickly and efficiently in a relatively short period of time using minimal quantities of material, in particular the testing of chemical catalysts under high pressure.

The device provides an intrinsically high speed with short relaxation times for setting and making constant parameters such as pressure, temperature and chemical potential.

Autoclaves are further known of a type wherein the initial pressure is brought about by connecting the reaction chamber of the autoclave to gas bottles or gas lines for feed of gas under relatively high pressure. This has the drawback the device takes a complex and voluminous form. In the device according to the invention this drawback is obviated in that the gases are generated in situ, i.e. in the container itself.

According to a preferred embodiment of the invention the first electrode is positioned in the first chamber of the container for generating therein a gas of a first type, for instance hydrogen, the second electrode is positioned in the second chamber of the container for generating therein a gas of the second type, for instance oxygen, and both chambers are mutually connected by a connecting channel of a length such that the mutual diffusion of the gases is limited. Arranged in the connecting channels is a material which is electrically conductive but which hampers the mutual diffusion of the gases in the separate chambers. In this preferred embodiment the gases created at each electrode can be generated separately, i.e. without mixing, and subsequently used in a chemical or physical process.

According to a further preferred embodiment the device comprises pressure-determining means for determining the pressure of the electrochemically generated gases, for instance in the form of a piezoresistive pressure sensor. This pressure sensor can be arranged directly in (a chamber of) the container, for instance by screwing it fixedly to an outer end of the container. In addition to such pressure sensors other sensors may also be used, for instance sensors which detect determined chemical properties, as well as temperature sensors.

According to a further preferred embodiment the device comprises voltage-controlling means for controlling the voltage between the first and the second electrode and thereby controlling the speed at which the electrochemical generation takes place. In a further preferred embodiment a pressure build-up speed in the container of at least 8 bar per second or even 20 bar per second or more can be achieved. In a further preferred embodiment the voltage-controlling means are connected to control means for substantially real time control of the electrochemical generation speed and, as a consequence thereof, the pressure in (the chambers of) container. In the device according to the invention a rapid pressure build-up occurs inside the container such

that the control means can control the pressure in the container substantially in real time, i.e. within a few minutes or even seconds.

Despite the high pressure in the container, which can rise to several thousand bar, no high structural standards are required for the material from which the container is manufactured. Despite these high pressures, the container can be manufactured from standard available material which easy to process, such as aluminium or plastic. This enables manufacture of the devices in simple manner, on large scale and at low cost.

According to a further aspect of the invention an integrated circuit is provided for simultaneously performing a large number of chemical or physical processes, comprising a substrate, devices of the above stated type arranged on the substrate for performing the processes, in addition to control means provided on the substrate and connected to the devices for controlling the processes performed in the respective containers. Such an integrated circuit is manufactured by per se known micro-manufacturing techniques, wherein a large number of devices manufactured from silicon, glass or similar material are for instance arranged on a silicon substrate.

In research into for instance new catalysts or the improvement of existing catalysts an extensive number of experiments have to be performed with large series of a particular type of catalyst, each experiment having slightly differing material parameters. This approach is known as high throughput experimentation (HTE) and is characterized by a large number of syntheses and analyses. By integrating a large number of the devices in an integrated circuit a large number of analyses and syntheses can be performed according to the invention in parallel and within a very short period of time, and furthermore only a small quantity of catalyst material is required per catalyst.

According to a further aspect of the invention a method is provided for analysing the action of a catalyst, comprising of:

- arranging a first and a second electrode in the container;
- arranging the catalyst in the container;
- filling the container with electrolyte;
- applying a potential difference over the electrodes for electrochemical generation of the electrolyte;
- determining at determined pressure values the compensating current required to substantially hold the pressure in the container at this pressure value for the purpose of analysing the action of the catalyst. The action of the catalyst can hereby be determined in simple and rapid manner at a large number of (high) pressures.

Further advantages, features and details of the present invention will be elucidated in the following description of preferred embodiments thereof. Reference is made in the description to the figures, in which:

- figure 1 shows a cross-section of a micro-pressure cell according to a first embodiment;
- figure 2 shows a cross-section of a micro-pressure cell according to a second embodiment;
- figure 3 shows a graph in which the built-up pressure is shown as a function of time at an amperage of 20 mA;
- figure 4 shows a graph in which the built-up pressure is shown as a function of time at an amperage of 50 mA;
- figure 5 shows a graph of the current required to compensate the reverse reaction (compensating current) as a function of the pressure;
- figure 6 shows a cross-section of a further embodiment of a micro-pressure cell for testing chemical catalysts; and
- figure 7 shows a perspective view of an integrated circuit of micro-pressure cells.

Figure 1 shows an embodiment of a micro-pressure cell with an aluminium container 1 provided with a chamber. Screwed to an outer end of the chamber is a pressure sensor 4, which has a stainless steel housing and an aluminium base plate. Glued into the base plate are two platinum electrodes 2 (127 micrometre platinum wire, glued into a fused silicon dioxide (fused silica) capillary). A standard O-ring 5 is used as high pressure seal. The content of the resulting chamber is in the order of magnitude of a few millilitres or less.

The electrodes 2 are connected to a power source 3 via standard voltage cables fed through container 1. After filling the micro-pressure cell with electrolyte, such as for instance water with added salts, and screwing in the pressure sensor 4, an electric current is generated and a number of gases result from electrolysis in the container, such as for instance hydrogen gas (H_2) and oxygen gas (O_2).

Figure 2 shows an alternative embodiment of the micro-pressure cell. A first chamber 7 and a second chamber 8 are provided in a container 6 which can be manufactured from plastic, in this case plexiglass. Provided between the two chambers 7 and 8 is a connecting channel 9, which provides an open connection between the chambers. An electrode 10 is arranged in chamber 7 and an electrode 11 in chamber 8. Both electrodes are connected via standard electricity wires 14 to a power source 15, which can in turn be controlled with for instance a computer 16 or the like. Chambers 7 and 8 are closed on their top side with respective sealing caps 12 and 13, in which are provided outlet channels 18 and 19 which are provided with valves 20,21. Suppose for instance that electrode 10 functions as anode and electrode 11 functions as cathode, respectively oxygen (O_2) and hydrogen (H_2) are then generated in chamber 7 and chamber 8. Both gases can be discharged separately in this embodiment. The connecting channel 9 has in the centre thereof a branch to a pressure sensor 17 shown in dashed lines, which can measure the pressure prevailing in connecting channel 9.

Figure 3 shows the pressure build-up by the formed gases during electrolysis of a 100 mM KNO_3 aqueous solution in a pressure cell with a content of 150 μl, at an amperage of 20 mA. Application of this inert electrolyte results in per se known electrolysis reactions. Figure 3 shows how the pressure p (in bar) progresses as a function of time t (in seconds) after the power source 3 has been switched on. It can be seen here that the pressure increases in almost linear manner with time due to the constant production of gas. Pressure in the container already rises quite rapidly at this relatively low amperage. After 900 seconds (marking A) the power is switched off and the pressure decreases due to the reverse reaction: the formation of water from hydrogen gas and oxygen gas.

Figure 4 shows the pressure build-up for the same system at an amperage of 50mA (marking C), wherein at each 100 bar pressure rise (markings B_1, B_2,...) the amperage is determined which is required to hold the pressure constant. This "compensating current" is a measure for the speed of the reverse reaction. The figure shows that the pressure increases rapidly, wherein high pressure values of about 1400 bar can be realized.

In figure 5 the "compensating current" is plotted against the pressure. For pressures in excess of 800 bar the compensating current increases exponentially, which is an indication of a drastic increase of the reverse reaction under high pressure.

Figure 6 shows an embodiment of a pressure cell for testing chemical catalysts. In a reaction space 22 closed with caps 28 hydrogen gas under high pressure is generated by platinum electrodes 2 through electrolysis. In the pressure cell there also takes place under the influence of catalyst 27 a catalysed hydrogenating action of a compound which has been introduced into the reaction space 22. From the measured "compensating current" at a given pressure can be derived the reaction speed of the relevant hydrogenating action, and

therewith the action (activity) of the catalyst 27 used. In this way the catalyst can be analysed and tested.

In addition to generating the above mentioned gases such as hydrogen and oxygen, there is also the option of electrochemical in situ generation of other gases such as Cl_2, F_2, D_2, CO_2, I_2 and Br_2.

Owing to the small dimensions and by using microtechniques such as thin or thick film technique, abrasive techniques, etching techniques and lithographic techniques for manufacture, it is possible to manufacture large numbers of pressure cells operating in parallel, whereby large series of experiments can be performed in a relatively short period of time using minimal quantities of material.

In a preferred embodiment of the invention (not shown) a large number of devices (such as micro-pressure cells) as well as the associated electrical connections and the like are arranged on a single substrate in an integrated circuit using for instance abrasive techniques, etching techniques, vacuum technique such as sputtering or vapour deposition, optionally in combination with lithographic techniques. This enables the manufacture of micro-chemical processing units for using in HTE systems, whereby a very large number of analyses can be performed in a short period of time.

Further referred to as possible applications are a chemical micro-reactor, and in particular a micro-fuel cell, or a high pressure micro-pump.

Because a high pressure can be available in this system, determined reactions which are normally performed at a fairly high pressure and at an increased temperature can now be performed at a lower temperature or even at room temperature. An example of such a reaction is the hydrogenation of benzene to cyclohexane. This reaction, which has been performed heretofore with the "normal" means at a raised pressure and an increased temperature, can be performed at room temperature in the cell according to the invention.

In addition, the use of short high pressure pulses can have an influence on the kinetics of the reaction.

This is because of differences in the way the reaction comes about. If the reaction comes about through the collision of two or more molecules, it is the diffusion speed of the molecules which will be of the main importance for the speed at which the reaction proceeds, and not a parameter such as the pressure on the reaction mixture.

If the reaction is unimolecular (as in the case of an isomerization), it is then the amount of energy supplied to the molecule by the system which is of importance for the speed at which the reaction proceeds. This depends on the pressure on the reaction mixture.

If both of the above stated reactions can occur in a reaction mixture, the unimolecular reaction will then be given preference by a pressure pulse. The specificity of the reaction can thus be influenced.

In order to enable performing of more reactions, a number of extra "means" can be applied which have an effect on the reaction conditions. These means have the purpose of giving the molecules just that extra energy they need to react. The molecules can also be influenced with these methods such that this reaction progresses more specifically. A reaction wherein for instance two stereoisomers are formed then proceeds such that more of the one stereoisomer is formed than of the other. The formation of a so-called enantiomeric excess therefore occurs in the reaction.

These means can be:

- heating means for increasing the temperature in the container, preferably by integrating into the cell a heating element in the form of an electrical resistor. This resistor can be embodied simply in thin film technology by applying a metal film. By passing a current through this metal film it becomes warm and thus heats its environment. The heating means can be arranged along the walls of the chamber so that the content of the chamber is heated from the outside, but also along an electrode or catalyst so that the content of the chamber is heated from this side.

- means for applying an electrical field over the reaction mixture. This can be done by arranging two metal plates parallel to the chamber in which the reaction takes place, which plates function as capacitor plates over which a voltage can be applied. By applying a voltage (some tens to thousands of volts) an electrical field is created over the reaction mixture, which causes a change in the molecules in the reaction mixture. This can bring about a change in the orientation of the atoms in the molecules whereby particular reactions will progress better than other reactions. It is necessary here to envisage stereochemical reactions wherein more of the one stereoisomer will be formed than the other. The electrical field can be both continuous (caused by a DC voltage) and alternating (caused by an alternating voltage over the capacitor plates).

- means for applying a magnetic field over the reaction mixture. This can be done by making a coil round the reaction mixture over which a direct voltage or an alternating voltage is applied. Both continuous fields and alternating fields may also be used in this case.

- means for irradiating the reaction mixture with optical or electromagnetic radiation, in particular radiation from the deep infrared spectrum to the gamma range. Another purpose of the radiation can be to record a photo-spectrum of the reaction mixture in order to see which reaction products are being formed.

- means for supplying acoustic energy, for instance ultrasonic energy, to the content of the container.

In addition, catalysts may be added to the reaction mixture in order to see which reaction products are formed or to cause a determined reaction to proceed "catalytically".

The application of means for introducing high-energy electrons into the reaction chamber can influence the reaction of components of the reaction mixture as desired.

An example of a reaction which has significant advantages if performed at high pressure is the Diels-Alder reaction of butadiene with di-(R)-methyl fumarate. This reaction gives two possible stereoisomers. If the reaction is performed at high pressure, 10 times as much S-isomer results as when the reaction is performed at one atmosphere. Owing to the rapid pressure build-up in the present pressure cell there will moreover occur preferred reactions, wherein other less preferred reactions are reduced or do not even take place.

Further shown in figure 7 is a schematic perspective view of an integrated circuit on which a number of micro-pressure sensors are arranged. The circuit comprises a substrate 29 on which fifteen micro-pressure cells are arranged. Each micro-pressure cell comprises a first chamber 30, a second chamber 31 and a connecting channel 32. Using a control unit 33 liquid and electricity lines 34 are controlled which are connected to each of the pressure cells (only shown schematically in the figure). The voltage values required to operate the micro-pressure cells can be set in the order of magnitude of the voltage values for operating electronic circuits (not shown) arranged on the substrate, such as transistors, diodes, resistors, etc. A hybrid chip has hereby been created in which an integration of electronic circuits and micro-pressure cells on a substrate is realized.

The present invention is not limited to the described preferred embodiments thereof; the rights sought are defined by the claims, within the scope of which many modifications can be envisaged.

International application PCT/NL01/00364
Enclosure to letter dated 22/07/2002

AMENDED CLAIMS

1. Integrated circuit for simultaneously performing a large number of chemical or physical processes, comprising:

- a number of devices for electrochemically generating one or more gases under high pressure;
- a substrate on which the devices are arranged;
- control means provided on the substrate and connected to the devices for controlling the processes performed in the respective containers;

wherein a device comprises:

- a container comprising a first chamber and a second chamber for filling with electrolites, each of the chambers having a content in the order of magnitude of a few millilitres or less;
- a first and a second electrode over which a voltage difference can be applied in order to bring about the electrochemical generation in the container, the first electrode being positioned in the first chamber of the container for generating therein a gas of a first type, the second electrode being positioned in the second chamber of the container for generating therein a gas of a second type, and both chambers being mutually connected by a connecting channel.

2. Integrated circuit according to claim 1, wherein the content is a maximum of 150 microlitres of electrolyte.

3. Integrated circuit according to claim 1 or 2, wherein the length of the connecting channel is such that diffusion of the gases is limited.

4. Integrated circuit according to any of the claims 1-3, wherein each of the chambers has a content in the order of magnitude of 150 microlitres or less.

5. Integrated circuit or device according to any of claims 3-4, wherein an electrically conductive material is arranged in the connecting channel so as to further limit the mutual diffusion of the gasses.

6. Integrated circuit or device as claimed in any of claims 1-5, wherein the control means comprise pressure-determining means for determining the pressure of the electrochemically generated gases.

7. Integrated circuit or device as claimed in claim 6, wherein the pressure-determining means comprise a piezoresistive pressure sensor.

8. Integrated circuit or device as claimed in any of the foregoing claims, wherein the control means comprise voltage-controlling means for controlling the voltage between the first and second electrodes of the devices.

9. Integrated circuit or device as claimed in claim 8, wherein the voltage controlling means control the voltage between the first and second electrode of a device so as to achieve a pressure build-up speed in the container of said device of 8 bar per second or more, or even 20 bar or more.

10. Integrated circuit or device as claimed in claim 8 or 9, wherein the voltage controlling means are operable so as to control substantially in real time the pressure in the container of a device.

11. Integrated circuit or device as claimed in any of the foregoing claims, comprising heating means for heating the content of one of more of the chambers of said devices.

12. Integrated circuit or device as claimed in claim 11, wherein the heating means comprise a metal film arranged in the container and connected to an electrical power supply.

13. Integrated circuit or device as claimed in any of the foregoing claims, provided with means for applying an electrical field over the container.

14. Integrated circuit or device as claimed in any of the foregoing claims, provided with means for applying a magnetic field in the container.

15. Integrated circuit or device as claimed in any of the foregoing claims, wherein the container is substantially optically transparent.

16. Integrated circuit or device as claimed in claim 13, comprising optical radiating means for irradiating the content of the container.

17. Integrated circuit or device as claimed in any of the foregoing claims, provided with means for supplying acoustic energy to the content of the container.

18. Integrated circuit or device as claimed in any of the foregoing claims, comprising catalyst fixing means for fixing a catalyst in the container.

19. Integrated circuit or device as claimed in any of the foregoing claims, comprising means for providing high-energy electrons in the container.

20. Integrated circuit or device as claimed in any of the foregoing claims, wherein the container is manufactured from standard technical material such as aluminium or plastic, and in particular plexiglass.

21. Integrated circuit or device as claimed in any of the foregoing claims, wherein the electrochemically generated gas is H_2, O_2, Cl_2, F_2, D_2, CO_2, I_2 and/or Br_2.

22. Method for electrochemically generating one or more gases in the integrated circuit according to any of the preceding claims, comprising of:

- arranging electrolyte in a container of the device;

- arranging a first and a second electrode in the container of said device;

- applying a potential difference over the electrodes so as to bring about the electrochemical generation in the container.

23. Method according to claim 22, comprising of:

- arranging a catalyst in the container;

- arranging the content of the container when a potential difference over the electrodes is applied for electrochemical generation of gas; by determining the compensation current required to substantially hold the pressure in the container constant.

24. Method according to claim 23, wherein the step of analyzing comprises determining at determined pressure values the compensating current required to substantially hold the pressure in the container at this pressure value.

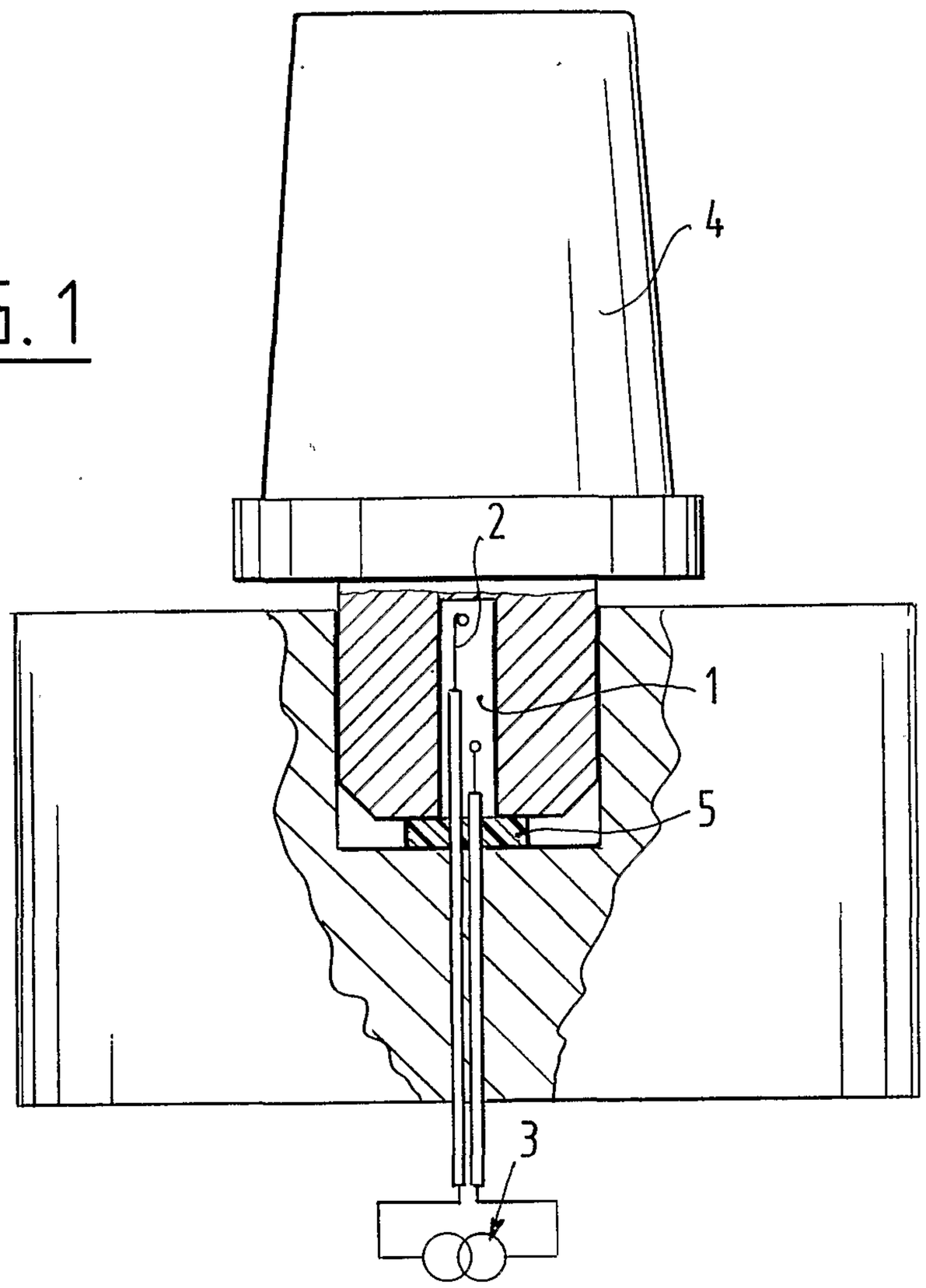


FIG. 1

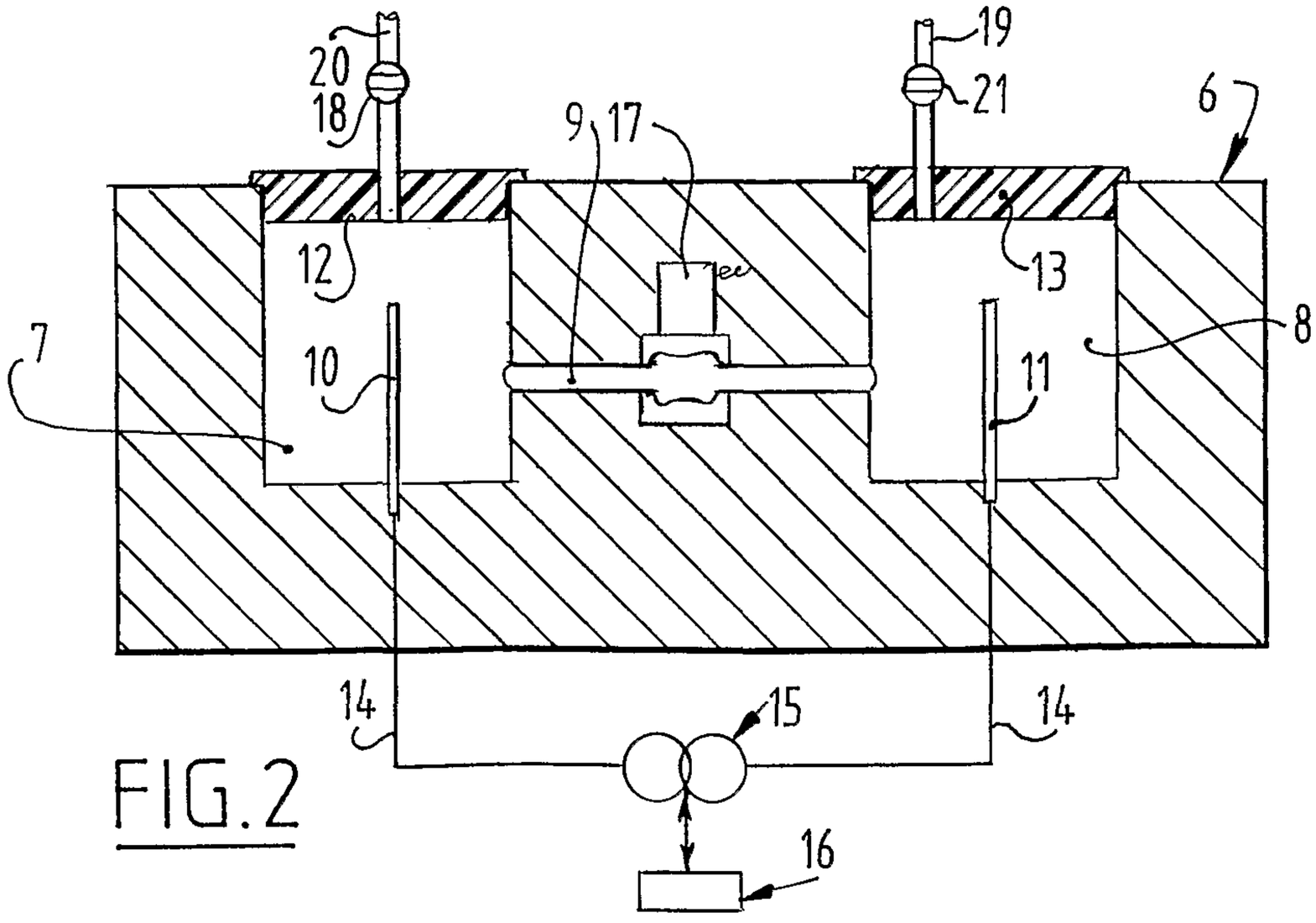
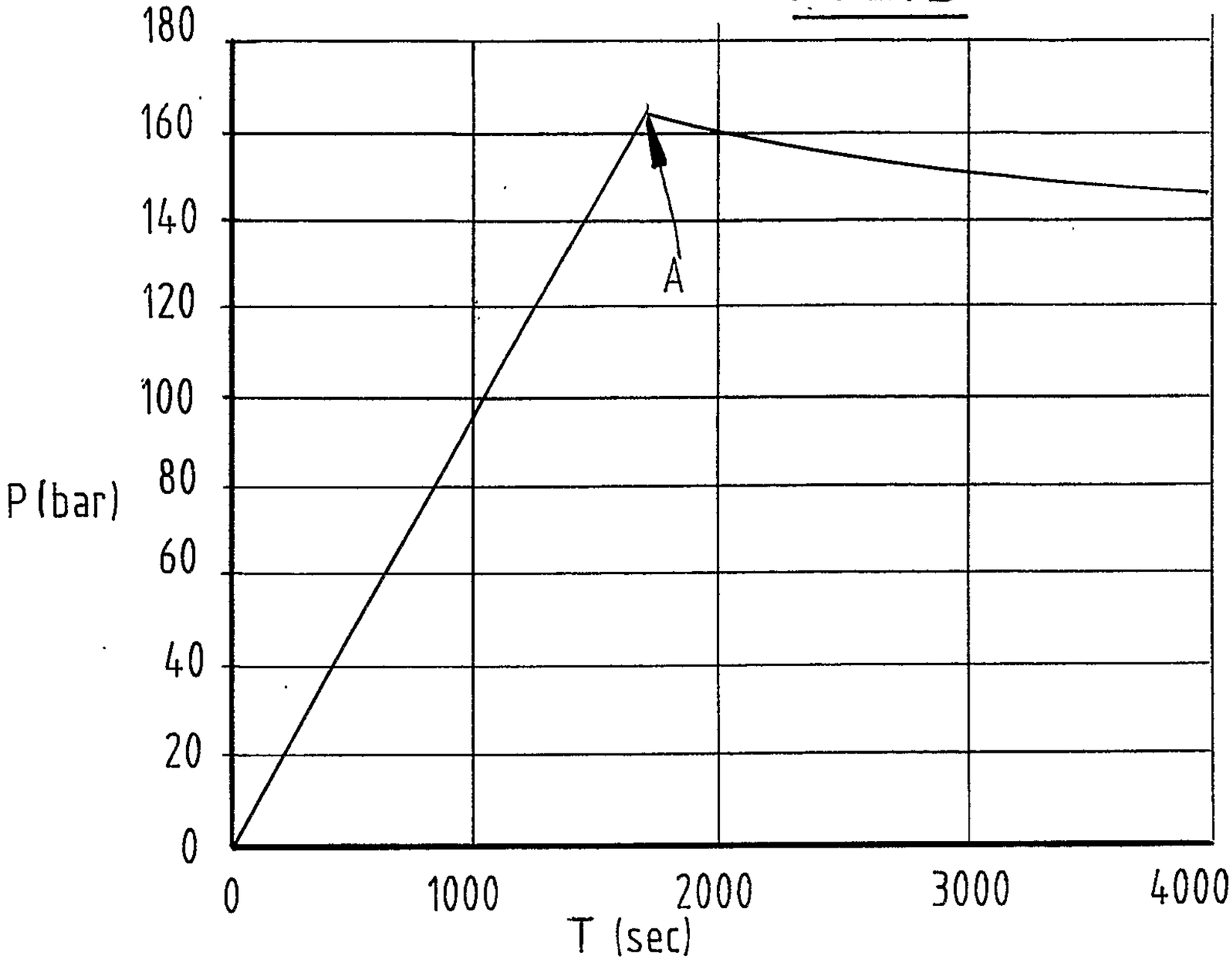


FIG. 2

FIG. 3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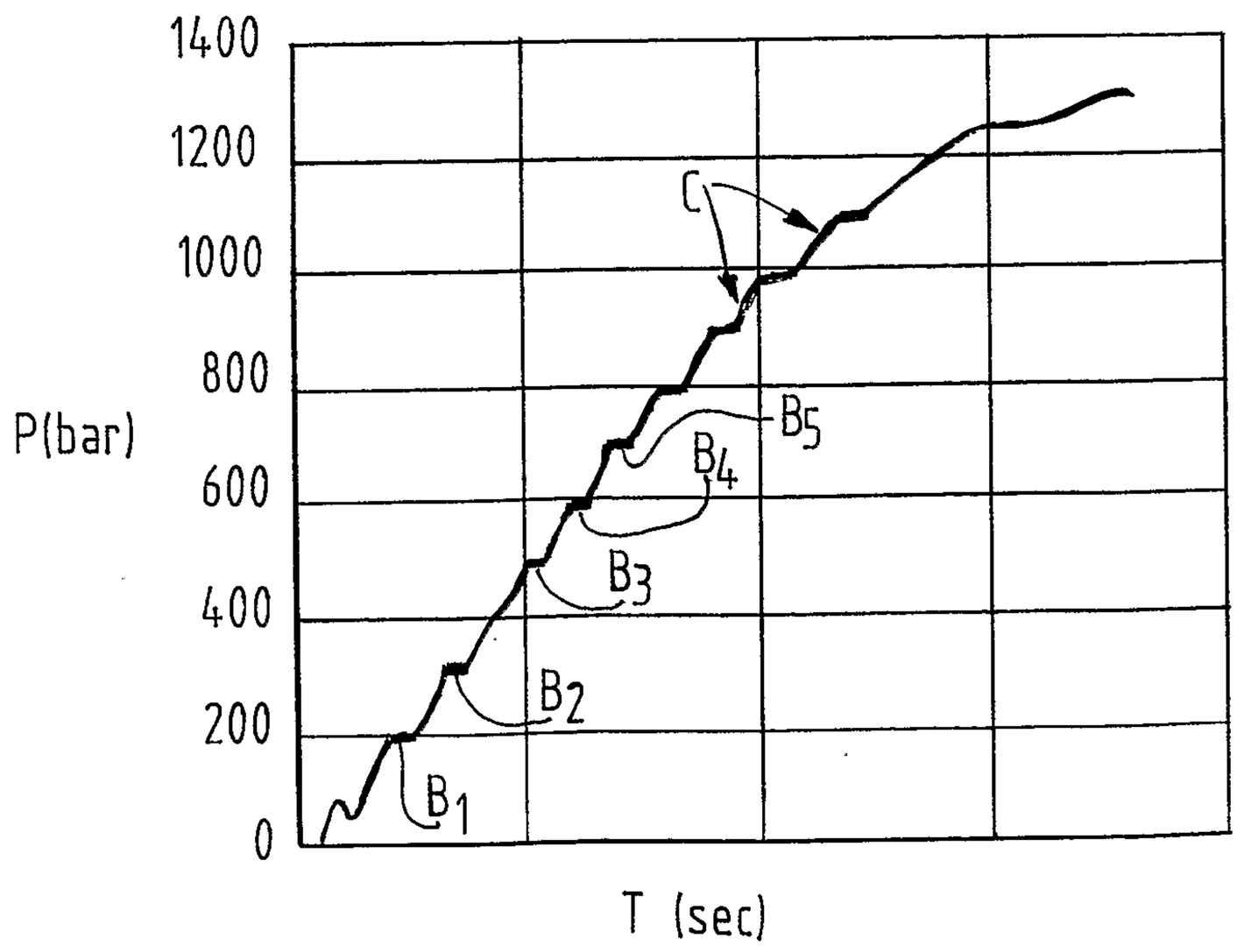
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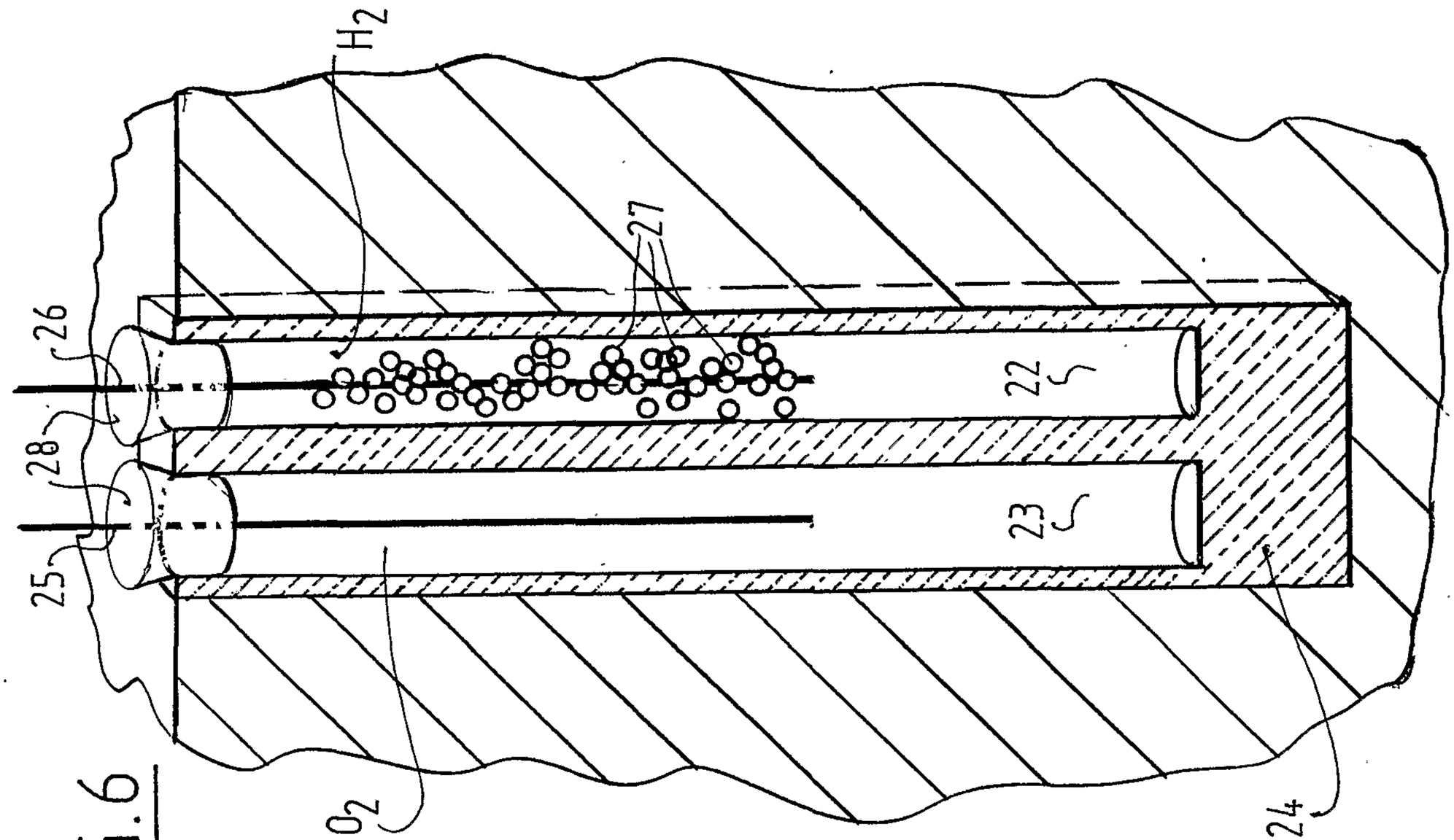
FIG. 4


H2
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O2
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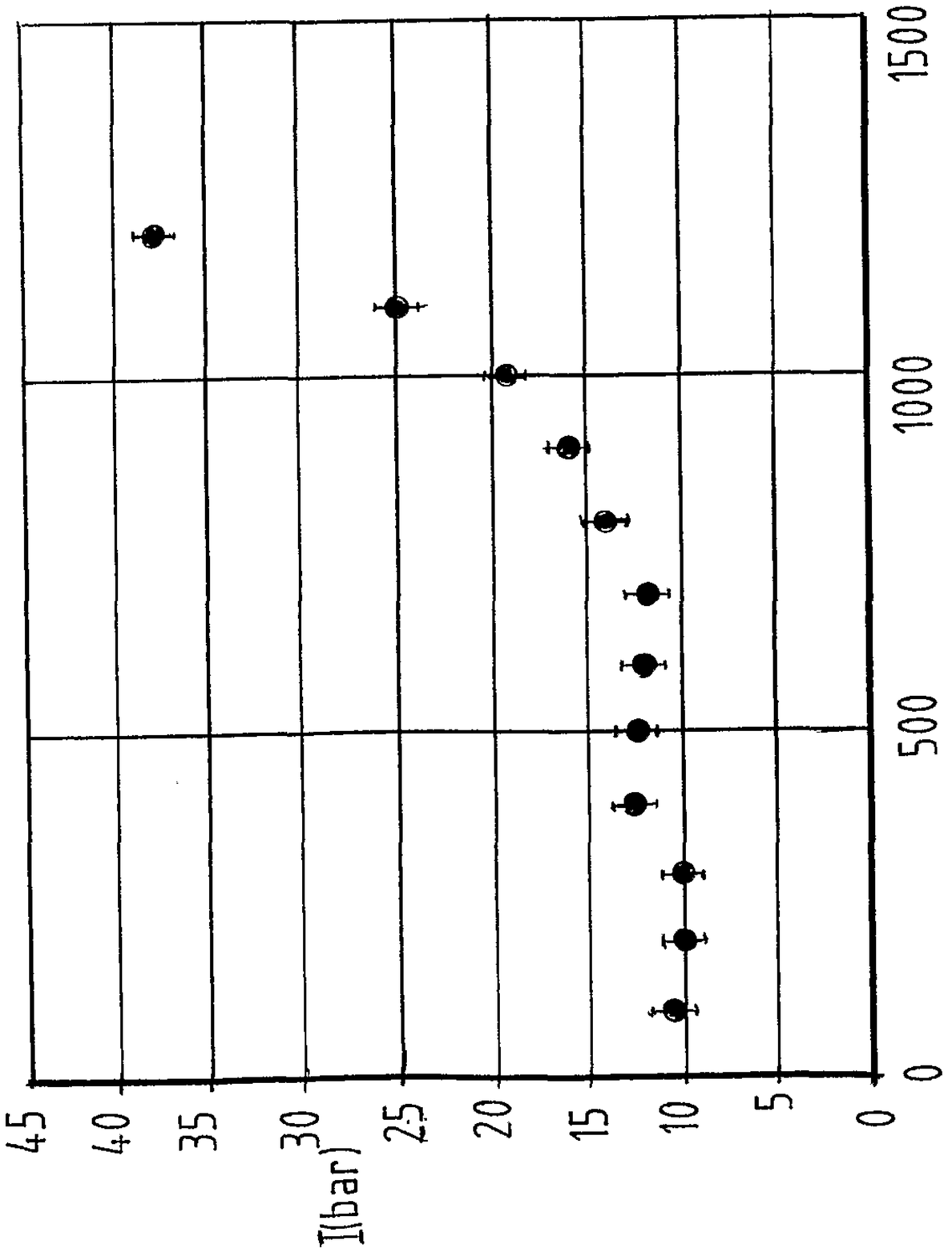
FIG.6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I(bar)

FIG.5

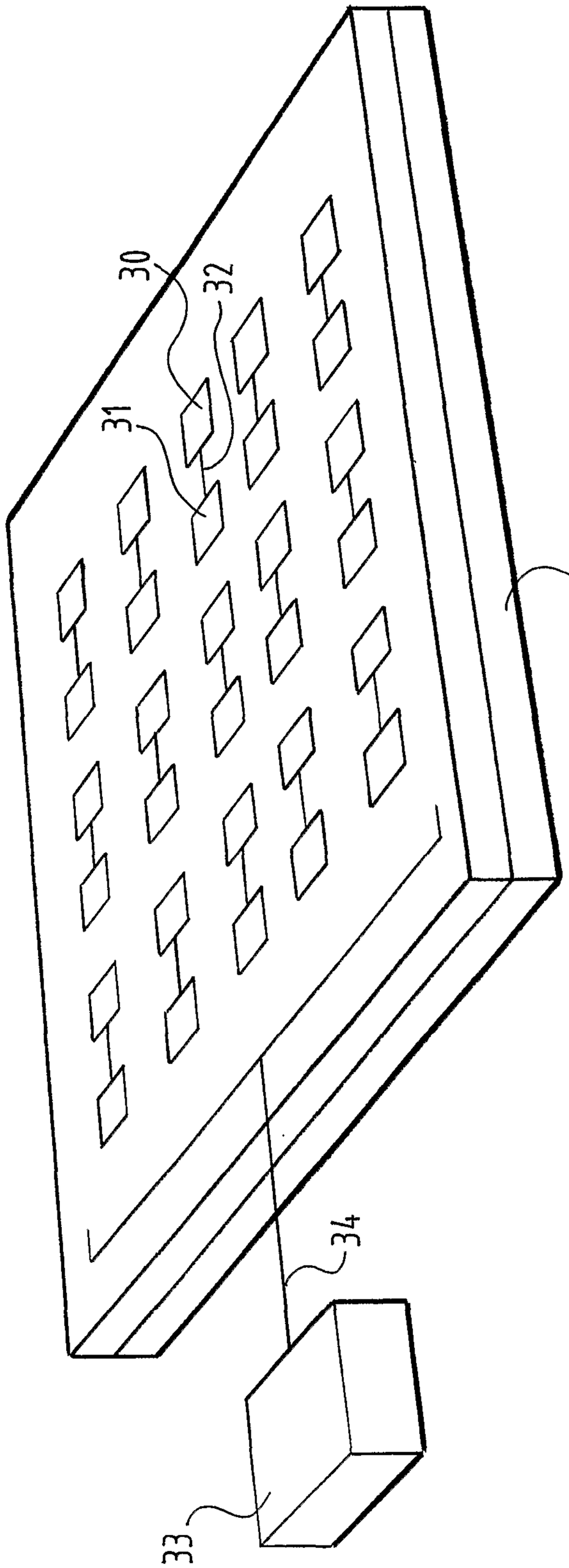
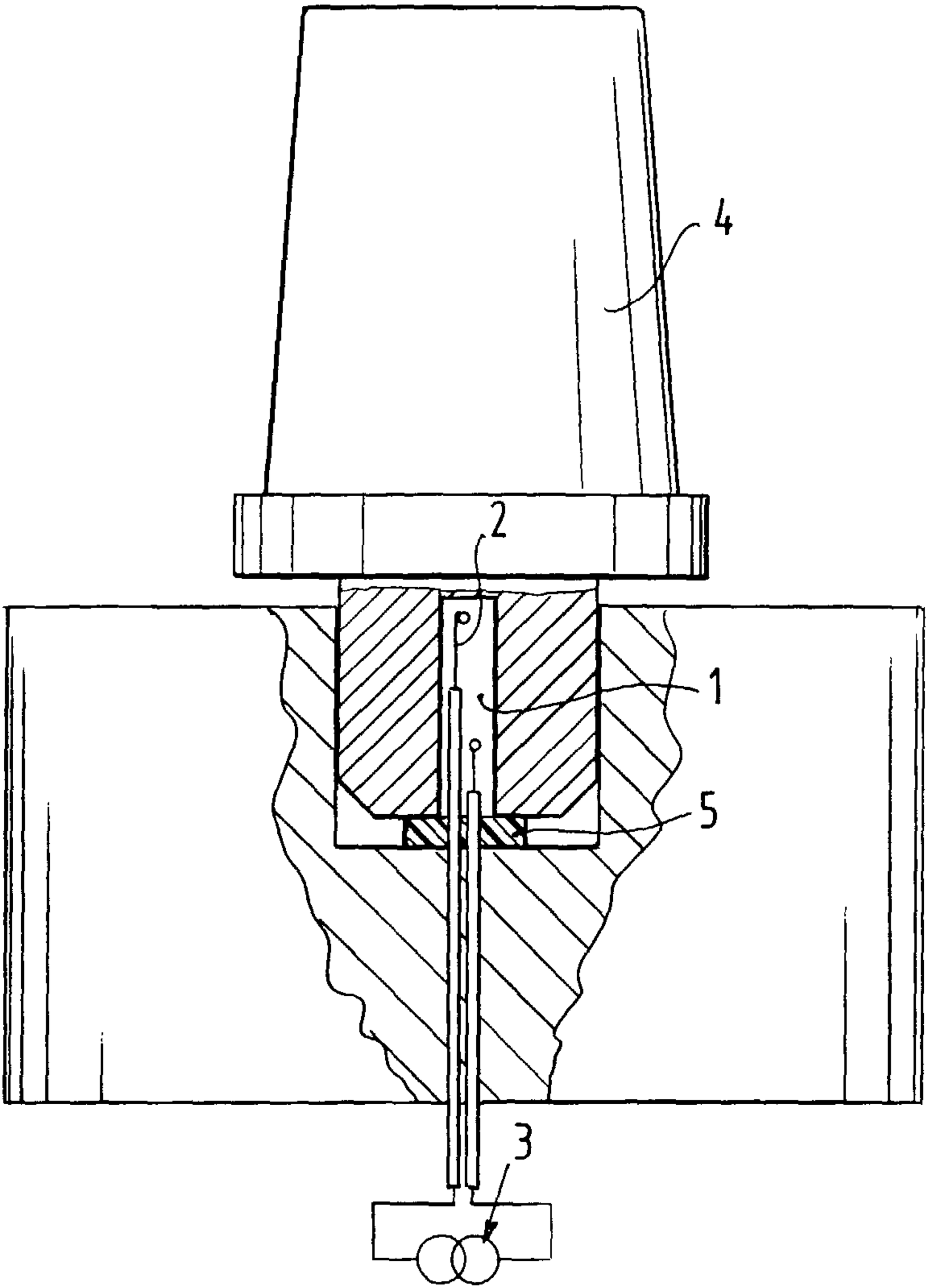


FIG. 7


4
2
1
5
3