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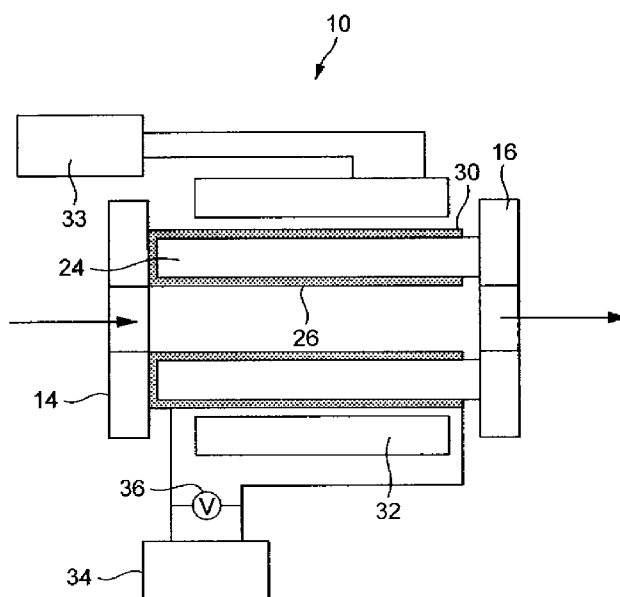
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(54) Title: SOLID-STATE GAS PURIFIER



(57) Abstract: A method is described for removing an impurity from a gas. The method comprises the steps of conveying the gas through a solid-state gas purifier comprising an alkali metal ion conducting membrane having a first electrode located on a first surface thereof exposed to the gas, and a second electrode located on a second surface thereof. A current is generated that flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with an impurity within the gas to form a non-volatile species that is retained on the purifier. The current may be periodically reversed to release the impurity from the purifier in order to re-generate the purifier.

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SOLID-STATE GAS PURIFIER

The present invention relates to a method of purifying gas, and to a solid-state purifier for purifying gas.

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Within a semiconductor fabrication plant, a number of inert gases are used for a variety of purposes, including environment purge and wafer processing. These gases may be supplied to a process chamber from various sources. For example, nitrogen is typically conveyed about a fabrication plant using a
10 gas distribution network, argon is typically supplied to a chamber from gas cylinders, and xenon may be supplied to the chamber from a local recovery and recycling system.

Depending on the nature of the process conducted in the process chamber,
15 the purity of a gas supplied to the chamber may be of considerable importance. In some semiconductor fabrication processes, gases such as nitrogen, helium and argon are often required to not have more than 10 ppb impurity levels to ensure that the impurities do not degrade the quality, and hence the performance of the semiconductor devices. Therefore, it is
20 common for ultra high purity (UHP) gas lines to be fitted with a gas purifier at the point of use to remove impurities from the gas. The key impurities to be removed from the gas stream are typically inorganic in nature, and examples include CO₂, NO_x, SO_x and H₂O.

25 Purifiers are known which use a chemically active getter material to remove impurities from gases by adsorption of the impurities on to the surface of the getter material. Such purifiers have a finite capacity and while the purifiers are sized to provide an acceptable lifetime under normal operation, during fault conditions they may rapidly reach full capacity and cease operation.
30 Such purifiers also have a relatively low capacity to size ratio, so that a large purifier is required to achieve an acceptably high capacity and lifetime.

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In a first aspect, the present invention provides a method of removing an impurity from a gas, the method comprising the steps of conveying the gas through a solid-state gas purifier comprising an alkali metal ion conducting membrane having a first electrode located on a first surface thereof exposed to the gas, and a second electrode located on a second surface thereof; and generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with an impurity within the gas to form a non-volatile species that is retained on the purifier.

Alkali metal ion conducting membranes such as a sodium or potassium beta alumina are able to transport alkali metal ions to an active (first) surface of the membrane through the application of a current between electrodes formed on the surfaces of the membrane. Once an amount of alkali metal has been transported to the active surface of the membrane, the purifier can behave as an electrochemical concentration cell, with the alkali metal ions reacting with impurities such as CO₂, NO_x, SO_x and H₂O to form a salt of the alkali metal which is retained on the active surface of the membrane.

The current may be generated continuously between the membranes during use of the purifier so that there is a steady transport of alkali metal ions towards the active surface of the membrane. Alternatively, the current may be periodically generated between the electrodes to transport fresh alkali metal ions to the first surface as required in order to maintain at least a certain amount of the alkali metal at the first surface. In one embodiment, a current is generated between the electrodes depending on the potential difference between the first electrode and a reference electrode located on the membrane, for example on the second surface. This potential difference will be indicative of the concentration of alkali metal available to react with the impurity, and so, absent the supply of fresh alkali metal ions to the surface, the potential difference will change as the purification takes place. At a pre-defined potential difference, the current may be generated between the

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electrodes to replenish the amount of alkali metal at the active surface. Consequently, the purifier can be self-monitoring and self-activating.

5 The purifier can differentiate between various levels of contamination of the gas. The purity of the gas may be determined from at least one of the frequency at which current is generated between the electrodes, and the potential difference between the first electrode and a reference electrode. For example, if the frequency at which the current was being generated to transport alkali metal ions to the active surface is relatively high, that is, there
10 is only a relatively short interval between each period in which current is generated between the electrodes to replenish the amount of alkali metal ions present at the active surface, this can be indicative of a relatively high contamination of the gas.

15 An alert may be generated depending on at least one of the frequency at which current is generated between the electrodes, and the rate of change of the potential difference between the first electrode and the reference electrode. For example, if there is a rapid change in the potential difference, or if the interval between each period of current generation becomes very
20 short, this may be indicative of a catastrophic event, such as air ingress into the gas entering the purifier. In this case, the generation of the current between the electrodes may be suspended in order to inhibit the formation of the non-volatile species during this event, thereby increasing the lifetime of the purifier. The purifier may be re-activated following rectification of the
25 catastrophic event by transporting fresh alkali metal ions to the active surface.

The reference electrode and/or the second electrode may include a source of alkali metal ions. This source of alkali metal ions may comprise one of a catalytically active electrode material and an inorganic compound of the alkali
30 metal, such as a carbonate or a nitrate. An amalgam of the alkali metal and another metal, such as gold, tin or lead, may provide the source of alkali metal

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ions. The first electrode may be formed from catalytically active material, such as platinum.

Depending on the nature of the membrane, the membrane may require
5 heating to transport alkali metal ions to the active surface. For example, sodium and potassium beta aluminas may require heating to a temperature in the range from 150 to 500 °C to become ionically conductive. The membrane may be continuously heated during use of the purifier to maintain the membrane at an elevated temperature, or the temperature of the purifier may
10 be elevated only when alkali metal ions need to be transported through the membrane.

In order to regenerate the purifier, the direction in which the generated current flows between the electrodes may be periodically reversed. Where the non-
15 volatile species form a redox couple with the original gas phase impurities, the alkali metal can be pumped away from the active surface, back into the membrane, by reversing the direction of the flow of current between the electrodes, causing the gas phase impurity to be released from the purifier. Following the release of the impurities from the purifier, the direction of the
20 flow of current between the electrodes can be reversed again to return alkali metal ions to the active surface. Therefore, in a second aspect the present invention provides a method of removing an impurity from a gas, the method comprising the steps of conveying the gas through a solid-state gas purifier comprising an alkali metal ion conducting membrane having a first electrode
25 located on a first surface thereof exposed to the gas, and a second electrode located on a second surface thereof; generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with an impurity within the gas to form a non-volatile species that is retained on the purifier, and periodically reversing the direction
30 in which the generated current flows between the electrodes.

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When the gas is being conveyed to a chamber, the direction of the current flow between the electrodes may be changed at any convenient time, for example during a chamber cleaning process, when samples are being loaded into the chamber or at any other time when a process is not being performed inside the chamber. Alternatively, or additionally, the direction of the current flow between the electrodes may be changed depending on the potential difference between the first electrode and a reference electrode. For example, when the potential difference indicates that the purifier is approaching full capacity, the current may be reversed to release the gas phase impurity from the purifier. The potential difference may be subsequently monitored during the regeneration process to enable the end of the regeneration process to be identified.

Alternatively, or additionally, the direction in which the generated current flows between the electrodes may be changed depending on the composition of the gas. The purifier may form part of a gas recovery and recycling system that removes species from a gas stream to isolate a particular gaseous species for re-use. The gas stream may contain varying relative amounts of gaseous species, for example if the gas stream has been exhausted from a process chamber in which different gases are supplied to the chamber over time, or if the gas stream has been exhausted from a chromatography column in which the composition of the gas stream will vary with time. Depending on the design of the system used to isolate the (first) gaseous species to be recovered, the first gaseous species may be contaminated with a second, unwanted gaseous species. For example, in a chromatography based purification system used to separate xenon from a gas stream, trace amounts of CO₂ may be exhausted from the chromatography column with the xenon. The first gaseous species may be a noble gas, such as xenon or krypton, or an inert gas, such as nitrogen.

30

In this event, when the gas passing through the purifier is rich in the first gaseous species current flows between the electrodes to transport alkali

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metal ions to the first surface of the membrane to react with the second gaseous species to form a non-volatile species that is retained on the purifier, thereby purifying the first gaseous species for subsequent recovery. When the gas is lean in the first gaseous species, the flow of current between the electrodes is reversed to release the second gaseous species from the purifier and thereby regenerate the purifier for a subsequent purification treatment when the gas is again rich in the first gaseous species.

In a third aspect the present invention provides a method of removing an impurity from a gas, the method comprising the steps of conveying the gas through a solid-state gas purifier comprising an alkali metal ion conducting membrane having a first electrode located on a first active surface thereof exposed to the gas, and a second electrode located on a second active surface thereof exposed to the gas; generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with an impurity within the gas to form a non-volatile species that is retained on the first surface of the membrane, and periodically reversing the direction in which the generated current flows between the electrodes to release the impurity from the first surface of the membrane, and to transport alkali metal ions to the second surface of the membrane to react with the impurity.

By exposing also the second surface to the gas, during the reverse flow of current between the electrodes this second surface can become a second active surface of the purifier, and so the purifier can be rapidly regenerated by a single reversal of the direction of the current flow. In this aspect of the invention, both of the electrodes are formed from catalytically active material, such as platinum, with the membrane providing the source of alkali metal ions.

In any of the above aspects, the first electrode is preferably in the form of a mesh or porous layer located on the first surface of the membrane. The

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membrane may be in the form of a plate, the first and second electrodes being located on opposing sides of the plate. The purifier may comprise a plurality of these plates defining a serpentine flow path for a gas stream passing therethrough. Alternatively, the membrane may be in the form of a tube, the first electrode being located on the inner periphery of the tube and the second electrode being located on the outer periphery of the tube. The purifier may comprise a plurality of these tubes each having its inner periphery exposed to the gas stream.

10 In a fourth aspect the present invention provides a solid-state gas purifier comprising a gas inlet; a gas outlet; an alkali metal ion conducting membrane having a first electrode located on a first surface thereof exposed to gas passing from the inlet to the outlet, and a second electrode located on a second surface thereof; and means for generating a current that flows
15 between the electrodes to transport alkali metal ions to the first surface of the membrane.

In a fifth aspect the present invention provides a solid-state gas purifier comprising a gas inlet; a gas outlet; an alkali metal ion conducting membrane
20 having a first electrode located on a first surface thereof exposed to gas passing from the inlet to the outlet, and a second electrode located on a second surface thereof; and means for generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane, and for periodically changing the direction in which the generated
25 current flows between the electrodes.

In a sixth aspect the present invention provides a reversible solid-state gas purifier comprising a housing having a gas inlet and a gas outlet, the housing containing an alkali metal ion conducting membrane having a first electrode
30 located on a first active surface thereof exposed to gas passing from the inlet to the outlet and a second electrode located on a second active surface thereof exposed to gas passing from the inlet to the outlet; and means for

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generating a current that flows between the electrodes to transport alkali metal ions to one of the surfaces of the membrane, and for periodically changing the direction in which the generated current flows between the electrodes to transport alkali metal ions to the other surface of the membrane.

5

Features described above in relation to method aspects of the invention are equally applicable to purifier or apparatus aspects, and vice versa.

Preferred features of the present invention will now be described, by way of example only, with reference to the accompanying drawings, in which:

10

Figure 1 illustrates schematically a solid-state purifier for purifying a gas;

15

Figure 2 illustrates a cross-section through a first embodiment of the purifier of Figure 1;

20

Figure 3 illustrates one example of a plate-like purifier element;

Figure 4 illustrates another example of a plate-like purifier element;

Figure 5 illustrates a cross-section through a second embodiment of the purifier of Figure 1; and

25

Figure 6 illustrates a purifier having a plurality of tubular purifier elements; and

With reference first to Figure 1, a solid-state purifier 10 generally comprises a housing 12 having a gas inlet 14 for receiving a gas stream, and a gas outlet 16 for exhausting the gas stream from the housing 12.

30

A first embodiment of the purifier 10 is illustrated in Figure 2. The housing 12 of the purifier 10 houses a number of purifier elements 20. In this embodiment, the housing 12 houses five purifier elements 20 each arranged

substantially orthogonal to the direction in which the gas stream enters the housing to define a serpentine flow path for the gas stream passing through the housing 12. However, the housing 12 may house any number (one or more) of such purifier elements 20 arranged at any angle.

5

A first example of a plate-like purifier element 20 is illustrated in Figure 3. The purifier element 20 comprises a ceramic base 22 upon which is mounted an alkali metal ion conducting membrane 24, for example a sodium or potassium beta alumina. The membrane 24 has a first electrode 26 located on a first,
10 active surface 28 of the membrane 24 that is exposed to the gas stream passing through the housing, and a second electrode 30 located on a second, passive surface of the membrane 24 which faces the base 22 and which is isolated from the gas stream.

15 In this example, the first electrode 26 is formed from catalytically active material, such as platinum, that is able to catalyse the reaction of alkali metal ions with impurities in the gas stream, and is in the form of a mesh or other porous structure for enabling the gas stream to come into contact with the active surface 28 of the membrane 24. The second electrode 30 includes a
20 source of alkali metal ions. Examples include an alloy of the alkali metal, such as an amalgam of the alkali metal and another metal, such as gold, tin or lead, or an inorganic compound of the alkali metal, such as a carbonate or a nitrate coated onto a catalytically active electrode.

25 Depending on the nature of the material used to form the membrane 24, the membrane may require heating to raise its temperature above a critical temperature T_c at which the membrane 20 is able to conduct alkali metal ions. In view of this, the purifier element cell 20 may comprise a heater 32 connected to the base 22 for heating the base 22, and by conduction the
30 membrane 24 to the required temperature for the transfer of alkali metal ions through the membrane 24. Depending again on the material used to form the membrane 24, this temperature may be in the range from 150 to 500°C. A

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heater controller may be provided for controlling the heater 32, for example in response to temperature signals received from a thermocouple located proximate the membrane 24. As an alternative to providing a dedicated heater 32 for each purifier element 20, a single heater 32 may be connected to the housing 12 to heat the purifier 10 to a temperature above T_c for the transfer of alkali metal ions through the membrane 24. The heater 32 may be operated to maintain the temperature of the membrane 24 above T_c during use of the purifier 10, or only when the transport of alkali metal ions through the membrane 24 is required.

A current generating device 34 is provided to generate a current between the first and second electrodes 26, 30 and thus across the membrane 24, to transport alkali metal ions to the active surface 28 of the membrane 24 when the temperature of the membrane is above T_c . A voltage measuring device 36 is also provided to measure the potential difference between the electrodes 26, 30. Gas tight electrical feed-throughs may permit electrical connections from the device 34 to pass to the electrodes 26, 30.

In use, when the temperature of the membrane is above T_c the current generating device 34 is operated to generate a current between the first and second electrodes 26, 30 to transport alkali metal ions to the active surface 28 of the membrane 24. The potential difference measured by the voltage measuring device 36 is indicative of the concentration of alkali metal ions at the active surface 28, and so the current may be generated between the electrodes 26, 30 until a first, relatively high potential difference exists between the electrodes 26, 30. When a gas is conveyed through the purifier 10, the hot alkali metal ions at the active surface 28 react with impurities such as CO_2 , NO_x , SO_x and H_2O in the gas stream to form non-volatile species, such as Na_2CO_3 and NaOH , which are deposited on the active surface 28.

As the impurities are removed from the gas stream, the concentration of alkali metal ions at the active surface 28 will fall. Therefore, when the potential

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difference measured by the voltage measuring device 36 is at or below a second, relatively low value, the current generating device 34 is operated again to generate a current between the first and second electrodes 26, 30 to transport fresh alkali metal ions to the active surface 28 of the membrane 24 to replenish the amount of alkali metal ions available to react with the impurities in the gas stream.

The frequency at which current is generated between the electrodes 26, 30, and/or the variation of the potential difference between the electrodes 26, 30, can provide information regarding the purity of the gas stream passing through the housing. For example, if the frequency at which current is generated between the electrodes is relatively high, that is, there is only a relatively short interval between each period in which current is generated between the electrodes 26, 30 to replenish the amount of alkali metal ions present at the active surface 28, this can be indicative of a relatively low purity of the gas stream. Depending on these values, an alarm may be generated by the current generating device 34 or other control device connected to one or both of the current generating device 34 and the voltage measuring device 36 to alert a user to the high impurity of the gas stream passing through the purifier 10. If this purity is particularly low, this may indicate the occurrence of a catastrophic event, such as air ingress into the gas entering the purifier 10. In this case, the generation of the current between the electrodes 26, 30 may be suspended in order to prevent the active surface 28 from becoming rapidly coated with the non-volatile species, thereby increasing the lifetime of the purifier. Following rectification of the catastrophic event, the purifier 10 may be re-activated to transport fresh alkali metal ions to the active surface 28.

During periods of inactivity, for example when the purifier 10 is not required to purify the gas stream passing therethrough, the purifier 10 may be regenerated by reversing the direction in which the current flows between the electrodes 26, 30. When the non-volatile species include salts such as carbonates, nitrates and sulphates, reversing the current can have the effect

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of reversing the reaction that occurred at the active surface 28 to release the impurity from the active surface 28 and transport the alkali metal ions towards the second electrode 30. Following the release of the impurities from the active surface 28, the direction of the flow of current between the electrodes
5 can be reversed again to return alkali metal ions to the active surface 28 in readiness for re-use of the purifier 10. Periodic regeneration of the purifier element(s) 20 in this manner can substantially increase the lifetime of the purifier 10.

10 Regeneration of the purifier 10 may be triggered by the potential difference recorded by the voltage measuring device 36, for example when the potential difference indicates that there is only a relatively low concentration of the alkali metal ions at the active surface 28. Alternatively, or additionally, when the gas stream passing through the purifier 10 has a variable composition,
15 regeneration may be triggered depending on the composition of the gas stream currently passing through the purifier 10. For example, when the purifier forms part of a recovery and recycling system to recover a particular gas that has been exhaust from a process chamber, the purifier may be regenerated when that gas is not passing through the purifier 10.

20 The example of the purifier element 20' illustrated in Figure 4 operates in much the same way as the purifier element 20 illustrated in Figure 3. In this second example, the indication of the concentration of alkali metal ions at the active surface 28 is provided by monitoring the potential difference between
25 the first electrode 28 and a reference electrode 40 located on the membrane 24, in this example on the second surface of the membrane 24. This reference electrode 40 may include a source of alkali metal ions.

A second embodiment of the purifier 10 is illustrated in Figure 5. In this
30 second embodiment, the membrane 24 is in the form of a cylindrical membrane having a bore 42 through which the gas stream passes from the gas inlet 14 to the gas outlet 16. The first electrode 26 is formed on the inner,

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active surface 28 of the membrane 24 for exposure to the gas stream, and the second electrode 30 is formed on the outer surface of the membrane 24, which also provides an outer surface of the housing 12. In this embodiment, the first electrode 26 is formed from catalytically active material, such as
5 platinum, and the second electrode 30 includes a source of alkali metal ions.

If required, a heater 32 extends about the membrane 24 for heating the membrane 24 to a temperature T_c for the transport of alkali metal ions through the membrane 24. A heater controller 33 may be provided for controlling the
10 heater 32, for example in response to temperature signals received from a thermocouple located proximate the membrane 24. As in the first embodiment, a current generating device 34 is provided to generate a current between the first and second electrodes 26, 30 and thus across the membrane 24, to transport alkali metal ions to the active surface 28 of the
15 membrane 24 when the temperature of the membrane is above T_c . A voltage measuring device 36 is also provided to measure the potential difference between the electrodes 26, 30. As illustrated in Figure 5, the first electrode 26 extends partially over the outer surface of the membrane 24 to facilitate connection of the current generating device 34 thereto.

20

The operation of the second embodiment of the purifier 10 is similar to that of the, or each, of the purifier elements 20 of the first embodiment. When the temperature of the membrane is above T_c the current generating device 34 is operated to generate a current between the first and second electrodes 26, 30
25 to transport alkali metal ions to the active surface 28 of the membrane 24. The potential difference measured by the voltage measuring device 36 is indicative of the concentration of alkali metal ions at the active surface 28, and so the current may be generated between the electrodes 26, 30 until a first, relatively high potential difference exists between the electrodes 26, 30.
30 When a gas is conveyed through the bore 42 of the membrane 24, the hot alkali metal ions at the active surface 28 react with impurities such as CO_2 ,

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NO_x , SO_x and H_2O in the gas stream to form non-volatile species, such as Na_2CO_3 and NaOH , which are deposited on the active surface 28.

As in the first embodiment, the frequency at which current is generated
5 between the electrodes 26, 30, and/or the variation of the potential difference between the electrodes 26, 30, can provide information regarding the purity of the gas stream passing through the bore 42 of the membrane 24. The purifier 10 may also be periodically regenerated by reversing the direction in which current flows between the electrodes 26, 30.

10

In the example illustrated in Figure 6, the purifier comprises a plurality of purifier elements 50, each of which is similar in structure of the membrane 24 of Figure 5 and has a current generating device connected to first and second electrodes thereof. The purifier elements 50 are arranged in parallel within a
15 cylindrical housing 12 of the purifier 10. As illustrated, a single heater 32 may be connected to the housing 12 to heat the housing 12 to elevate the temperature of the purifier elements 50 to a temperature above T_c during the transport of alkali metal ions within each purifier element 50. Each purifier element 50 has first and second electrodes located thereon in an
20 arrangement similar to that illustrated in Figure 5. A current generating device and voltage measuring device are connected to the electrodes of each purifier element in a manner similar to that shown in Figure 5.

In one example, the electrodes may have a similar composition to the
25 electrodes located on the membrane 24 of Figure 5, that is with the first electrode located on the bore of purifier element being formed from catalytically active material, and the second electrode located on the outer surface of the purifier element including a source of alkali metal ions. In this case, a manifold (not shown) may be located between the inlet of the purifier
30 10 and the purifier elements 50 to direct the gas stream entering the purifier into the bores of the purifier elements 50. A similar manifold may be located

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between the purifier elements 50 and the outlet 16 to direct the gas stream from the bores of the purifier elements 50 to the outlet 16.

In another example, each of the electrodes are formed from catalytically active material such as platinum, with the membrane itself providing the source of alkali metal ions for the purifier element 50. In this case, the inner and the outer surface of each cylindrical membrane can each provide, in turn, an active surface for the reaction of alkali metal ions with impurities contained in the gas stream passing through the purifier 10 depending on the direction in which the current flows between the electrodes. For example, initially current may be generated that flows between the electrodes to transport alkali metal ions to the bore of each membrane to react with an impurity within the gas to form a non-volatile species that is retained on the bore of the membrane. When the purifier elements are to be regenerated, for example depending on the composition of the gas stream passing through the purifier and/or on the potential difference between the electrodes, the direction in which the generated current flows between the electrodes is reversed to release the impurity from the bore of each membrane, and to transport alkali metal ions to the outer surface of the membrane to react with the impurity within the gas stream. This can provide a reversible solid-state purifier, with the active surface switching between the inner and the outer surface of each purifier element at the end of each regeneration period.

CLAIMS

1. A method of removing an impurity from a gas, the method comprising
5 the steps of conveying the gas through a solid-state gas purifier
comprising an alkali metal ion conducting membrane having a first
electrode located on a first surface thereof exposed to the gas, and a
second electrode located on a second surface thereof; and generating
a current that flows between the electrodes to transport alkali metal
10 ions to the first surface of the membrane to react with an impurity within
the gas to form a non-volatile species that is retained on the purifier.
2. A method according to Claim 1, wherein a current is periodically
15 generated between the electrodes to transport fresh alkali metal ions to
the first surface.
3. A method according to Claim 1 or Claim 2, wherein a current is
20 generated between the electrodes depending on the potential
difference between the first electrode and a reference electrode.
4. A method according to any preceding claim, wherein the purity of the
gas is determined from at least one of the frequency at which current is
25 generated between the electrodes, and the potential difference
between the first electrode and a reference electrode.
5. A method according to any preceding claim, wherein an alert is
30 generated depending on at least one of the frequency at which current
is generated between the electrodes, and the rate of change of the
potential difference between the first electrode and a reference
electrode.

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- 5
6. A method according to any preceding claim, wherein the generation of the current between the electrodes is suspended depending on the rate of change of the potential difference between the first electrode and a reference electrode.
7. A method according to any of Claims 3 to 6, wherein the reference electrode includes a source of alkali metal ions.
- 10
8. A method according to any preceding claim, wherein the second electrode includes a source of alkali metal ions.
- 15
9. A method according to Claim 7 or Claim 8, wherein the source of alkali metal ions comprises one of a catalytically active electrode coated with an inorganic compound of the alkali metal, such as a carbonate or a nitrate.
- 20
10. A method according to any of Claims 7 to 9, wherein the source of alkali metal ions comprises an amalgam of the alkali metal and another metal, such as gold, tin or lead.
11. A method according to any preceding claim, wherein the first electrode is formed from catalytically active material, such as platinum.
- 25
12. A method according to any preceding claim, wherein the temperature of the membrane is elevated to transport alkali metal ions to the first surface.
- 30
13. A method according to Claim 12, wherein the temperature of the membrane is elevated to a temperature in the range from 150 to 500°C.

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14. A method according to any preceding claim, wherein the impurity comprises at least one of CO₂, NO_x, SO_x and H₂O.
- 5 15. A method according to any preceding claim, wherein the direction in which the generated current flows between the electrodes is periodically reversed.
- 10 16. A method according to Claim 15, wherein the direction in which the generated current flows between the electrodes is changed depending on the composition of the gas.
- 15 17. A method of removing an impurity from a gas, the method comprising the steps of conveying the gas through a solid-state gas purifier comprising an alkali metal ion conducting membrane having a first electrode located on a first surface thereof exposed to the gas, and a second electrode located on a second surface thereof; generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with an impurity within the gas to form a non-volatile species that is retained on the purifier, and
20 periodically reversing the direction in which the generated current flows between the electrodes.
- 25 18. A method according to Claim 17, wherein the direction in which the generated current flows between the electrodes is changed depending on the composition of the gas.
- 30 19. A method according to Claim 18, wherein the gas contains varying amounts of first and second gaseous species, and when the gas is rich in the first gaseous species, current flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with the second gaseous species to form a non-volatile species that is retained on the purifier, and when the gas is lean in the first gaseous

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species, the flow of current between the electrodes is reversed to release the second gaseous species from the purifier.

20. A method according to Claim 19, wherein the second gaseous species
5 comprises at least one of CO₂, NO_x, and SO_x.
21. A method according to Claim 19 or Claim 20, wherein the first gaseous species comprises a noble gas.
- 10 22. A method according to any of Claims 17 to 21, wherein the direction in which the generated current flows between the electrodes is changed depending on the potential difference between the first electrode and a reference electrode.
- 15 23. A method according to Claim 22, wherein the reference electrode includes a source of alkali metal ions.
24. A method according to any of Claims 17 to 23, wherein the second electrode includes a source of alkali metal ions.
- 20 25. A method according to Claim 23 or Claim 24, wherein the source of alkali metal ions comprises one of a catalytically active electrode coated with an inorganic compound of the alkali metal, such as a carbonate or a nitrate.
- 25 26. A method according to any of Claims 23 to 25, wherein the source of alkali metal ions comprises an amalgam of the alkali metal and another metal, such as gold, tin or lead.
- 30 27. A method according to any of Claims 17 to 26, wherein the first electrode is formed from catalytically active material, such as platinum.

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28. A method according to any of Claims 17 to 27, wherein the temperature of the membrane is elevated to transport alkali metal ions through the membrane.
- 5 29. A method according to Claim 28, wherein the temperature of the membrane is elevated to a temperature in the range from 150 to 500°C.
- 10 30. A method of removing an impurity from a gas, the method comprising the steps of conveying the gas through a solid-state gas purifier comprising an alkali metal ion conducting membrane having a first electrode located on a first active surface thereof exposed to the gas, and a second electrode located on a second active surface thereof exposed to the gas; generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with an impurity within the gas to form a non-volatile species that is retained on the first surface of the membrane, and periodically reversing the direction in which the generated current flows between the electrodes to release the impurity from the first surface of the membrane, and to transport alkali metal ions to the second surface of the membrane to react with the impurity.
- 15 31. A method according to Claim 30, wherein the direction in which the generated current flows between the electrodes is changed depending on the composition of the gas.
- 25 32. A method according to Claim 31, wherein the gas contains varying amounts of first and second gaseous species, and when the gas is rich in the first gaseous species, current flows between the electrodes to transport alkali metal ions to the first surface of the membrane to react with the second gaseous species to form the non-volatile species, and,
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when the gas is lean in the first gaseous species, the flow of current between the electrodes is reversed..

- 5 33. A method according to Claim 32, wherein the second gaseous species comprises at least one of CO₂, NO_x, and SO_x.
34. A method according to Claim 32 or Claim 33, wherein the first gaseous species comprises a noble gas, such as xenon or krypton, or a relatively inert gas, such as nitrogen.
- 10 35. A method according to any of Claims 30 to 34, wherein the electrodes are formed from catalytically active material, such as platinum.
- 15 36. A method according to any of Claims 30 to 35, wherein the temperature of the membrane is elevated to transport alkali metal ions through the membrane.
- 20 37. A method according to Claim 36, wherein the temperature of the membrane is elevated to a temperature in the range from 150 to 500°C.
38. A method according to any preceding claim, wherein the first electrode is in the form of a mesh located on the first surface of the membrane.
- 25 39. A method according to any preceding claim, wherein the alkali metal comprises one of sodium and potassium.
40. A method according to any preceding claim, wherein the membrane comprises a beta alumina.

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41. A method according to any preceding claim, wherein the membrane is in the form of a plate, the first and second electrodes being located on opposing sides of the plate.
- 5 42. A method according to Claim 41, wherein the purifier comprises a plurality of said plates defining a serpentine flow path for a gas stream passing therethrough.
- 10 43. A method according to any of Claims 1 to 40, wherein the membrane is in the form of a tube, the first electrode being located on the inner periphery of the tube and the second electrode being located on the outer periphery of the tube.
- 15 44. A method according to Claim 43, wherein the purifier comprises a plurality of said tubes each having its inner periphery exposed to the gas stream.
- 20 45. A solid-state gas purifier comprising a gas inlet; a gas outlet; an alkali metal ion conducting membrane having a first electrode located on a first surface thereof exposed to gas passing from the inlet to the outlet, and a second electrode located on a second surface thereof; and means for generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane.
- 25 46. A purifier according to Claim 45, wherein the current generating means is configured to periodically generate a current between the electrodes.
- 30 47. A purifier according to Claim 45 or Claim 46, comprising a reference electrode located on the membrane, and means for measuring a potential difference between the first and reference electrodes, the current generating means being configured to generate a current

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between the first and second electrodes depending on the measured potential difference.

- 5 48. A purifier according to Claim 47, comprising means for determining the purity of the gas stream depending on at least one of the frequency at which current is generated between the electrodes, and the measured potential difference.
- 10 49. A purifier according to Claim 47 or Claim 48, comprising means for generating an alert depending on at least one of the rate of change of the measured potential difference and the frequency at which current is generated between the electrodes.
- 15 50. A purifier according to any of Claims 47 to 49, wherein the current generating means is configured to suspend the generation of the current between the electrodes depending on the rate of change of the measured potential difference.
- 20 51. A purifier according to any of Claims 47 to 50, wherein the reference electrode includes a source of alkali metal ions.
52. A purifier according to any of Claims 45 to 51, wherein the second electrode includes a source of alkali metal ions.
- 25 53. A purifier according to Claim 51 or Claim 52, wherein the source of alkali metal ions comprises one of a catalytically active electrode coated with an inorganic compound of the alkali metal, such as a carbonate or a nitrate.

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54. A purifier according to any of Claims 51 to 53, wherein the source of alkali metal ions comprises an amalgam of the alkali metal and another metal, such as gold, tin or lead.
- 5 55. A purifier according to any of Claims 45 to 54, wherein the first electrode is formed from catalytically active material, such as platinum.
- 10 56. A purifier according to any of Claims 45 to 55, wherein the current generating means is configured to periodically reverse the direction in which the generated current flows between the electrodes.
- 15 57. A purifier according to Claim 56, wherein the current generating means is configured to change the direction in which the generated current flows between the electrodes depending on the composition of a gas passing from the gas inlet to the gas outlet.
- 20 58. A solid-state gas purifier comprising a gas inlet; a gas outlet; an alkali metal ion conducting membrane having a first electrode located on a first surface thereof exposed to gas passing from the inlet to the outlet, and a second electrode located on a second surface thereof; and means for generating a current that flows between the electrodes to transport alkali metal ions to the first surface of the membrane, and for periodically changing the direction in which the generated current flows
25 between the electrodes.
- 30 59. A purifier according to Claim 58, wherein the current generating means is configured to change the direction in which the generated current flows between the electrodes depending on the composition of a gas passing from the gas inlet to the gas outlet.

60. A purifier according to Claim 58 or Claim 59, comprising a reference electrode located on the membrane, and wherein the current generating means is configured to change the direction in which the generated current flows between the electrodes depending on the potential difference between the first electrode and a reference electrode.
61. A purifier according to Claim 60, wherein the reference electrode includes a source of alkali metal ions.
62. A purifier according to any of Claims 58 to 61, wherein the second electrode includes a source of alkali metal ions.
63. A purifier according to Claim 61 or Claim 62, wherein the source of alkali metal ions comprises one of a catalytically active electrode coated with an inorganic compound of the alkali metal, such as a carbonate or a nitrate.
64. A purifier according to any of Claims 61 to 63, wherein the source of alkali metal ions comprises an amalgam of the alkali metal and another metal, such as gold, tin or lead.
65. A purifier according to any of Claims 58 to 64, wherein the first electrode is formed from catalytically active material, such as platinum.
66. A reversible solid-state gas purifier comprising a housing having a gas inlet and a gas outlet, the housing containing an alkali metal ion conducting membrane having a first electrode located on a first active surface thereof exposed to gas passing from the inlet to the outlet and a second electrode located on a second active surface thereof exposed to gas passing from the inlet to the outlet; and means for generating a current that flows between the electrodes to transport alkali metal ions

to one of the surfaces of the membrane, and for periodically changing the direction in which the generated current flows between the electrodes to transport alkali metal ions to the other surface of the membrane.

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67. A purifier according to Claim 66, wherein the current generating means is configured to change the direction in which the generated current flows between the electrodes depending on the composition of a gas passing from the gas inlet to the gas outlet.

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68. A purifier according to Claim 66 or Claim 67, wherein the electrodes are formed from catalytically active material, such as platinum.

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69. A purifier according to any of Claims 45 to 68, comprising means for controlling the temperature of the membrane.

70. A purifier according to Claim 69, wherein the temperature control means is arranged to elevate the temperature of the membrane during transport of alkali metals.

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71. A purifier according to any of Claims 45 to 70, wherein the first electrode is in the form of a mesh located on the first surface of the membrane.

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72. A purifier according to any of Claims 45 to 71, wherein the alkali metal comprises one of sodium and potassium.

73. A purifier according to any of Claims 45 to 72, wherein the membrane is formed from a beta alumina.

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74. A purifier according to any of Claims 45 to 73, wherein the membrane is in the form of a plate, the first and second electrodes being located on opposing sides of the plate.
- 5 75. A purifier according to Claim 74, comprises a plurality of membranes defining a serpentine flow path for a gas stream passing from the gas inlet to the gas outlet.
- 10 76. A purifier according to any of Claims 45 to 73, wherein the membrane is in the form of a tube, the first electrode being located on the inner periphery of the tube and the second electrode being located on the outer periphery of the tube.
- 15 77. A purifier according to Claim 76, comprising a plurality of said tubes each having its outer periphery exposed to a gas stream passing from the gas inlet to the gas outlet.
- 20 78. A gas recovery system comprising a purifier according to any of Claims 45 to 77.

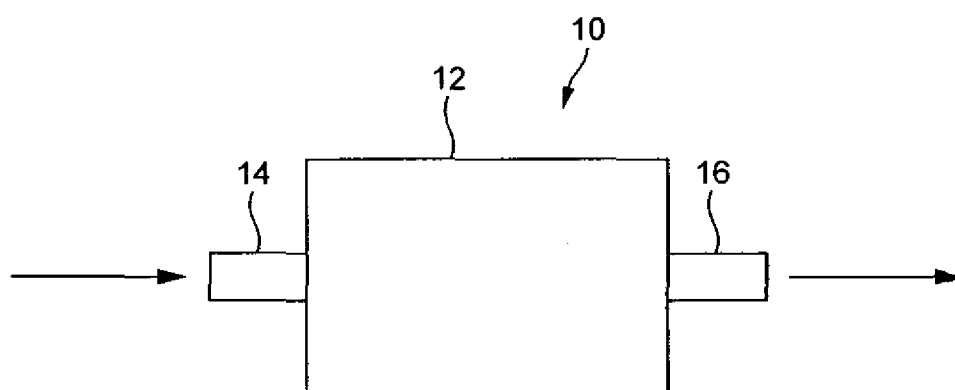


FIG. 1

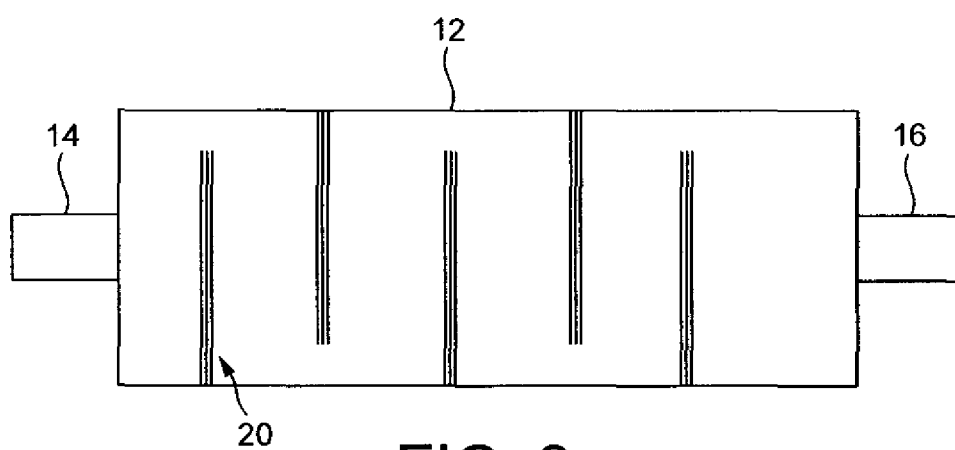


FIG. 2

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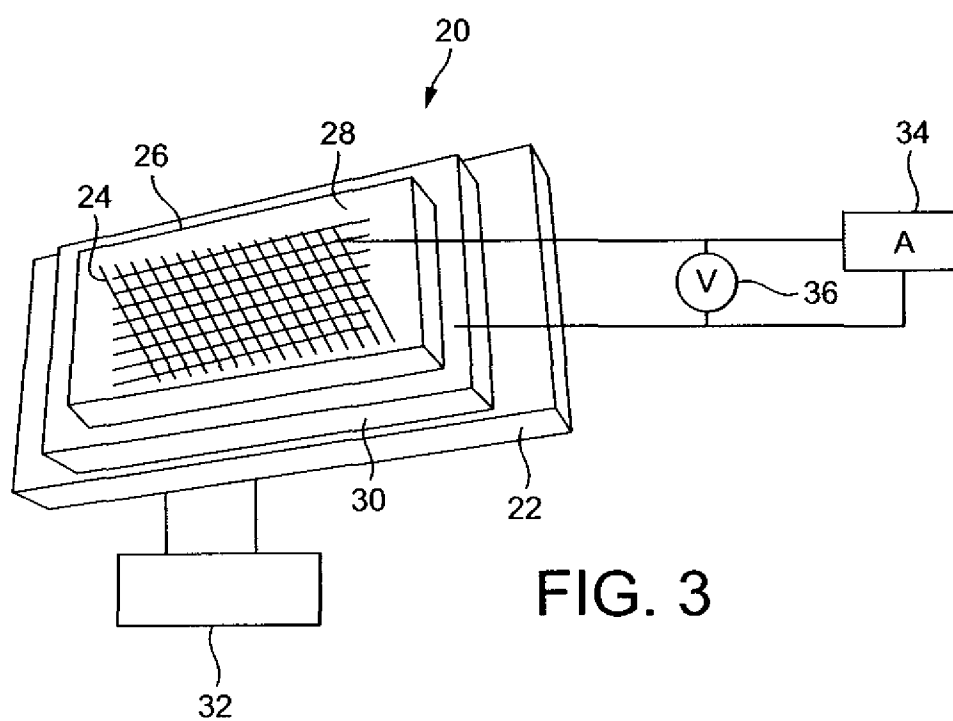


FIG. 3

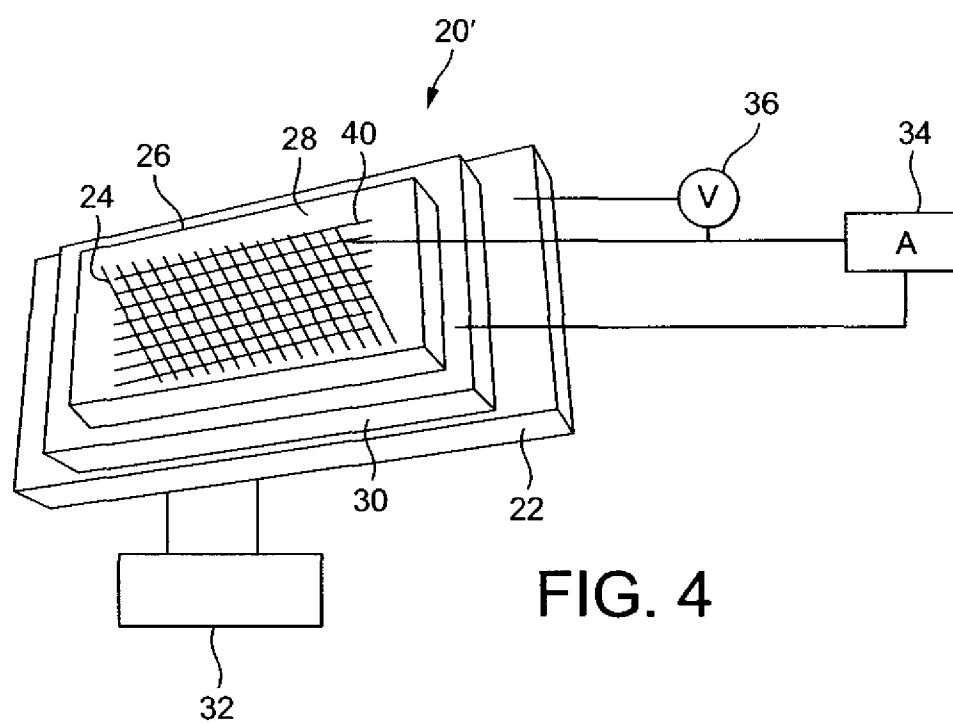


FIG. 4

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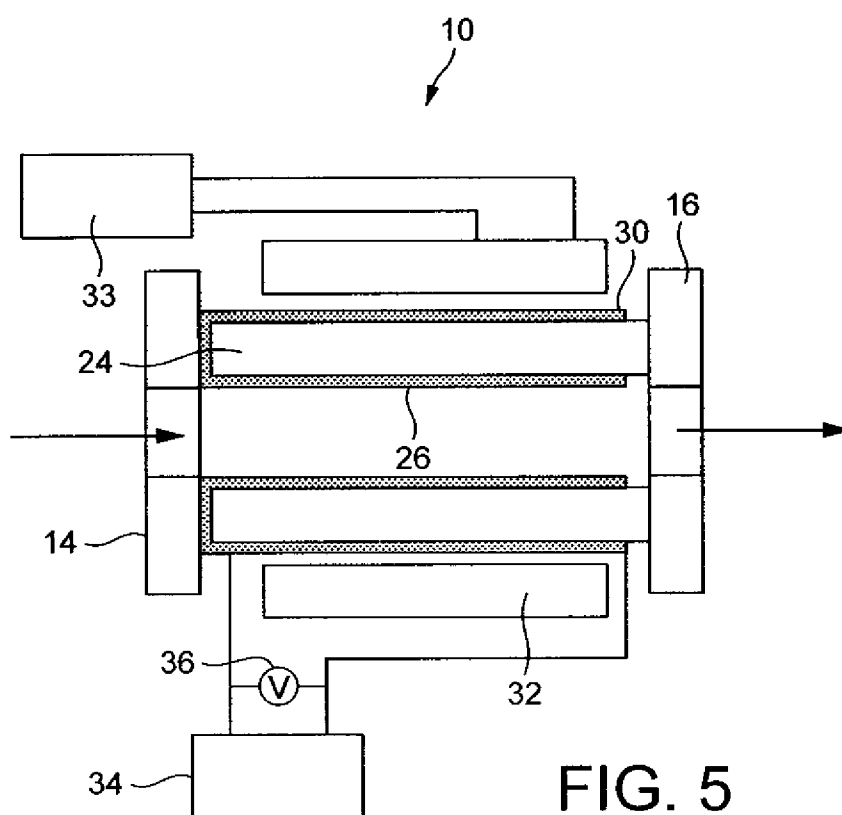


FIG. 5

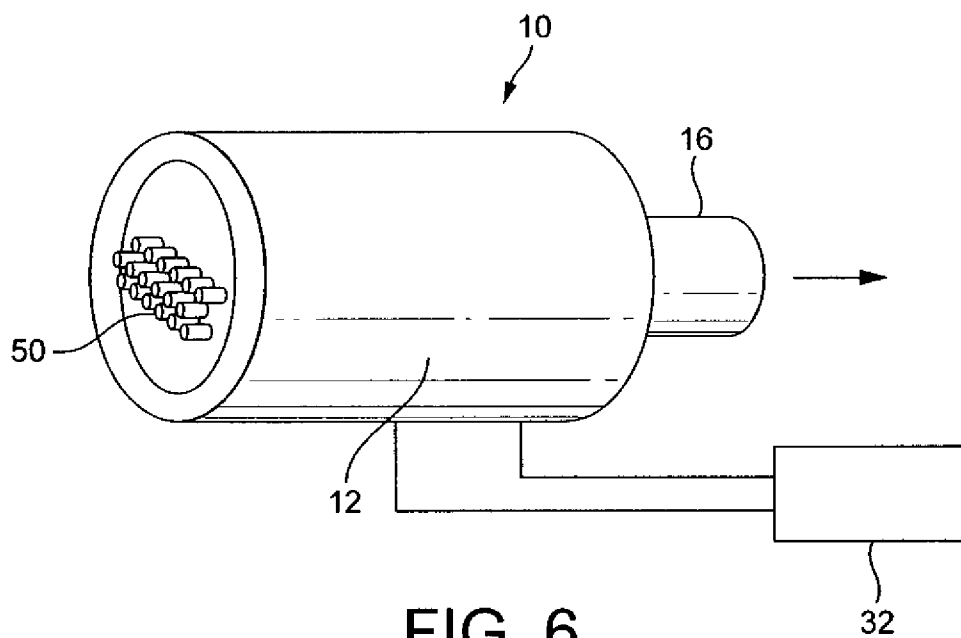


FIG. 6

INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2007/050119

A. CLASSIFICATION OF SUBJECT MATTER

INV. B01D53/32

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

B01D F01N C25B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 296 110 A (TABATABAIE-RAISSI ALI [US]) 22 March 1994 (1994-03-22) column 3, line 59 - column 4, line 57; figure 2	45,78
X	US 4 772 366 A (WINNICK JACK [US]) 20 September 1988 (1988-09-20) column 2, line 9 - column 3, line 27; claims; figure 1	1,45,78
A	EP 0 566 071 A1 (TOYODA CHUO KENKYUSHO KK [JP]) 20 October 1993 (1993-10-20) figure 1; example 1	1-78
A	US 5 399 246 A (JOSHI ASHOK V [US]) 21 March 1995 (1995-03-21) column 9, line 32 - column 11, line 23; claims 15-17; figure 9	1-78

☐ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents:

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O document referring to an oral disclosure, use, exhibition or other means

P document published prior to the international filing date but later than the priority date claimed

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

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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.

Z document member of the same patent family

Date of the actual completion of the international search

12 June 2007

Date of mailing of the international search report

27/06/2007

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/GB2007/050119

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