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(54) Title: FUEL-CELL INTEGRAL MULTIFUNCTION HEATER AND METHODS

(57) Abstract: A fuel cell (10) or battery has an integral multifunction heater element (50) operable as an integral heater, an integral temperature sensor, and/or an integral aid to current collection from the fuel cell. The integral multifunction heater element is embedded within the electrolyte (40), anode (20), or cathode (30), disposed in thermal contact with at least one of the electrolyte, anode, or cathode, and is formed of a thin film of material of suitable resistivity, such as a refractory metal or conductive oxide, patterned in a suitable pattern. Conductive terminals (60) provide electrical contact to the thin film. The integral multifunction heater element (50) may include a thermistor or thermocouple (53). Methods are disclosed for fabricating and using the fuel cell with an integral multifunction heater element.



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FUEL-CELL INTEGRAL MULTIFUNCTION HEATER AND METHODS

10 TECHNICAL FIELD

This invention relates generally to fuel cells, and more particularly to fuel cells having an integral heater element, to electronic devices incorporating such fuel cells, and to methods of fabricating and methods of using such fuel cells.

15 BACKGROUND

Various portable devices, such as laptop computers, personal digital assistants (PDA's), portable digital and video cameras, portable music players, portable electronic games, and cellular phones or other wireless devices, require portable power sources. The weight and inconveniences of single-use batteries and
20 rechargeable batteries have motivated efforts to replace those power sources for portable use. Thus, there is an increasing demand for light-weight, re-usable, efficient, and reliable power sources in such applications and in many other applications as well. In attempts to meet these needs, various portable fuel cells have been developed, such as ceramic-based solid-oxide fuel cells, direct
25 methanol fuel-cell (DMFC) systems, reformed-methanol-to-hydrogen fuel-cell (RMHFC) systems, and other proton-exchange-membrane (PEM) fuel-cell systems. Some fuel-cell designs require heating of the fuel itself or heating of fuel-cell elements for best efficiency, while some fuel-cell designs require cooling. There is a continuing need and a large anticipated market for improved practical
30 compact portable fuel cells with rapid startup times and improved efficiency.

BRIEF DESCRIPTION OF THE DRAWINGS

The features and advantages of the disclosure will readily be appreciated by persons skilled in the art from the following detailed description when read in

5 conjunction with the drawings, wherein:

FIGS. 1A – 1D are cross-sectional side elevation views of various embodiments exemplifying fuel cells made in accordance with the invention.

FIGS. 2A – 2E are top plan views of a particular embodiment exemplifying a fuel cell made in accordance with the invention, at various stages of its fabrication.

10 FIGS. 3A - 3C are top plan views of pattern embodiments for an integral multifunction heater element made in accordance with the invention.

FIG. 4 is a flow chart illustrating an embodiment of a method for fabricating fuel cells in accordance with the invention.

For clarity of the description, the drawings are not drawn to a uniform scale. In

15 particular, vertical and horizontal scales may differ from each other and may vary from one drawing to another.

DETAILED DESCRIPTION OF EMBODIMENTS

Throughout this specification and the appended claims, the term “fuel cell” means
20 a fuel cell in its usual meaning or a battery cell having an anode, a cathode, and an electrolyte.

FIGS. 1A – 1D and FIGS. 2A – 2E show views of various embodiments of a fuel-cell structure **10** having an anode **20**, a cathode **30**, and an electrolyte **40**.

FIGS. 1A – 1D are cross-sectional side elevation views of various embodiments,
25 and FIGS. 2A – 2E are top plan views of a particular embodiment exemplifying a fuel cell made in accordance with the invention, at various stages of its fabrication. Methods of fabrication are described in detail hereinbelow.

An integral heater element **50** is provided in thermal contact with the electrolyte and embedded within the electrolyte **40**, anode **20**, or cathode **30**. Terminals **60** are electrically coupled to integral heater element **50** for providing heating current for heating electrolyte **40** to a desired temperature before operation and
5 (optionally) during operation of the fuel cell. Integral heater element **50** is a multi-function element in that it can be used, not only to heat the electrolyte **40** (herein referred to as "active mode"), but also to perform as a sensor (herein referred to as "passive mode"). When used as a sensor in the passive mode, integral heater element **50** may be used to sense electrolyte temperature, for example.
10 Terminals **60** may be used to electrically couple integral heater element **50** to resistance-measuring circuitry, voltage-measuring circuitry, and/or to a feedback control system for maintaining electrolyte **40** at an optimum temperature.

In some embodiments of the invention (cf. FIGS. 1A and 1B), integral heater element **50** may be disposed between anode **20** and cathode **30**, e. g., when
15 electrolyte **40** is between anode **20** and cathode **30**. Expressed in terms of the thickness of electrolyte **40**, integral heater element **50** may be placed entirely within that thickness, and it may be substantially centered between anode **20** and cathode **30**. Integral heater element **50** is spaced apart from both anode **20** and cathode **30** to prevent short circuits. However, in some embodiments of the
20 invention, integral heater element **50** may be disposed adjacent to anode **20** or adjacent to cathode **30**. Integral heater element **50** may extend over a minor or major portion of the projected area of electrolyte **40** (while leaving as much open area as possible for ionic conduction in the electrolyte, e.g., by including suitable interstices). It may be substantially centered with respect to the projected area of
25 electrolyte **40**. If necessary, to insulate integral heater element **50** from the anode and/or cathode, an insulator **55** may be employed, as shown in FIGS 1C and 1D.

In the embodiments shown in drawing FIGS. 1A – 1D and FIGS. 2A – 2E, integral heater element **50** comprises a thin film of a material having suitable resistivity,
30 e.g., a refractory metal such as nickel, palladium, platinum, chromium, molybdenum, tungsten, vanadium, niobium, tantalum, rhenium, ruthenium, osmium, rhodium, iridium, or alloys, combinations, or mixtures of such metals.

Such materials are useful in fuel cells such as solid-oxide fuel cells for which operating temperatures can exceed 800 °C.

Alternatively, integral heater element **50** may be formed of a conductive oxide.

By way of illustrative example, ceramics such as $\text{La}(\text{Sr})\text{Cr}(\text{Mn})\text{O}_3$ and

- 5 $\text{La}(\text{Sr})\text{Cr}(\text{Ti})\text{O}_3$, where the elements in parentheses are dopants, can be used for the heater. These are suitable "interconnect materials" having high electrical conductivity but low ionic conductivity, and can also be used in the anode, cathode or electrolyte. In situations where one wants to maximize the ionic conductivity through the fuel cell, materials such as $\text{La}(\text{Sr})\text{Cr}(\text{Mn})\text{O}_3$,
10 $\text{La}(\text{Sr})\text{Cr}(\text{Ti})\text{O}_3$, $\text{Sm}(\text{Sr})\text{CoO}_3$, or $\text{La}(\text{Sr})\text{Co}(\text{Fe})\text{O}_3$, [where (Sr), (Mn), (Ti), and (Fe) respectively denote dopants], can be used in the cathode and electrolyte, where these materials have both ionic and electrical conductivity.

The thin film of integral heater element **50** is patterned to form, for example, an elongated strip as shown in FIG. 3A arranged in a serpentine pattern, such as a

- 15 boustrophedonic pattern (like a bidirectional raster pattern) as shown in FIG. 3B. The pattern can be chosen to provide a desired resistance or reactance value and/or to provide coverage of a desired portion of electrolyte **40**, e.g., a geometric portion near the center of electrolyte **40**, and to allow suitable electrical conduction through electrolyte **40** during operation of fuel cell **10**. Thus, if integral
20 heater element **50** is disposed between anode **20** and cathode **30**, for example, it may be configured with openings or interstices suitable for allowing ionic current to flow between anode **20** and cathode **30**, as shown in the example configurations illustrated in FIGS. 3A - 3C.

- Fuel cell **10** may include an integral temperature sensor, which may comprise
25 integral heater element **50** itself, or be a distinct element made like integral heater element **50**. When used in that manner, the integral temperature sensor may be formed of the same material as integral heater element **50**. For example, when integral heater element **50** is not being used to heat electrolyte **40** or other parts of the fuel cell, its resistance may be used as a measure of the fuel cell

temperature by virtue of its temperature coefficient of resistance, i.e., it may be used as an integral temperature sensor.

Alternatively, integral heater element **50** may be formed by combining two dissimilar metals in a thermocouple configuration, with which a temperature
5 within the fuel cell may be measured by using the thermocouple contact potential (EMF). That is, integral heater element is formed by two dissimilar metallic portions **51** and **52** joined at a single junction **53**. Such a configuration is illustrated by FIG. 3C, in which portions **51** and **52** of integral heater element **50** are made of different metals, and their junction **53** forms the thermocouple
10 junction. Thus, in such an embodiment, integral heater element **50** may be used as an integral temperature sensor, by sensing the thermocouple contact potential in a conventional manner. Similarly, integral heater element **50** may include a thermistor as a temperature-sensing element.

Integral heater element **50** may include a current collector, which may be the
15 integral heater element itself or a distinct element made in the same manner. To provide the function of assisting in collection of current, suitable portions of integral heater element **50** may be made of thicker films and/or be formed of higher conductivity materials, such as silver or gold. Electrical terminals **60** may also be made of thicker films and/or be formed of higher conductivity materials,
20 such as copper, silver, or gold.

Thus, integral heater element **50** is a multi-function element. In an active mode, as an integral heater, it may be used to heat electrolyte **40** and/or other parts of the fuel cell. In a passive mode of operation, it may assist in collection of current from the fuel cell. As a sensor in a passive mode of operation, it may be used to
25 sense electrolyte temperature, for example. Thus, as described above, terminals **60** may be used to electrically couple integral heater element **50** to conventional resistance-measuring circuitry, and/or to a conventional feedback control system for maintaining electrolyte **40** at an optimum temperature. When integral heater element **50** is used in such a temperature control configuration, temperature may
30 be increased by switching integral heater element **50** to active mode heating, and temperature may be decreased by controlling flow rate of fuel, for example.

Thus, each of the embodiments described has an anode, a cathode, an electrolyte, and integral means for heating at least one of those elements (anode, cathode, and electrolyte). The integral means for heating those elements is disposed within the fuel cell and in thermal contact with at least one of the
5 elements (anode, cathode, and electrolyte). Each of the embodiments also includes means for providing heating current, for heating before operation, and optionally for heating during operation of the fuel cell.

The structures of fuel cell embodiments disclosed herein are specially adapted for fabrication by specific methods described in the following detailed description
10 and by the appended claims and their equivalents.

FABRICATION METHODS

FIG. 4 is a flow chart in which steps are denoted by reference numerals **S10**, ..., **S70**, illustrating an embodiment of an overall method for fabricating fuel cells in
15 accordance with the invention. Arrows indicate various alternate paths through the flow chart. One skilled in the art will recognize that embodiments of the invention may be made by following various paths through the flow chart, depending on the specific structure embodiment desired. The following description describes a sequence for an exemplary embodiment.

20 A suitable substrate **70** is provided (**S10**) for formation of a fuel cell, and a layer of electrolyte **40** is deposited (**S20**) upon substrate **70**. Substrate **70** may be a silicon wafer, an oxidized silicon wafer, a smooth, flat ceramic plate or other dielectric substrate, or a metallic plate, for example. A top plan view of electrolyte **40** on substrate **70** is shown in FIG. 2A. Electrolyte **40** may be deposited
25 through a mask to define its extent on substrate **70**. Electrolyte **40** is any suitable electrolyte. One skilled in the art will recognize that a variety of electrode and electrolyte materials may be exploited for use with the invention. For example, cubic fluorites such as Sm- or Gd-doped CeO₂ and yttria-stabilized zirconia (YSZ), e.g., 8 mole% yttria, doped perovskite oxides such as

La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O₃, proton-conducting perovskites such as BaZrO₃, SrCeO₃, and BaCeO₃, other proton-exchange ceramics, or ion- conductive polymers such as a perfluorosulfonic acid resin membrane (e.g., NafionTM, available from DuPont Chemicals, Inc., Wilmington, DE) may be used as electrolytes in fuel
5 cells made in accordance with the invention.

A thin heater film is deposited and patterned (**S30**) to form integral heater element **50**. As described above, integral heater element **50** can comprise a film of platinum or another refractory metal such as nickel, palladium, chromium, molybdenum, tungsten, or an alloy, combination, or mixture of such metals, or
10 conducting oxides. FIG. 2B shows integral heater **50** after deposition and patterning. Integral heater element **50** is embedded within electrolyte **40** in the embodiments of FIGS. 1B and 2E. Optionally, another layer of electrolyte **40** may be deposited (**S40**) over the heater film so that integral heater element **50** is disposed entirely within the thickness of a thicker electrolyte. If the two layers of
15 electrolyte are of about equal thicknesses, integral heater element **50** will be disposed in about the center of the resultant electrolyte thickness.

In step **S50**, an anode film is deposited and patterned to form anode **20**, as shown in FIG. 2D. Metallic cathodes and anodes, e.g., silver and nickel, may be employed. Cermets such as Ni-YSZ, and Cu-YSZ (catalyst-electrolyte), or Ni or
20 Cu modified doped ceria (e.g., Ce_{0.8}Sm_{0.2}O_{1.9}, Ce_{0.9}Gd_{0.1}O_{1.9}) may be employed as anode materials. Exemplary cathodes include doped perovskites, such as Sm_{0.5}Sr_{0.5}CoO₃, Ba_{0.8}La_{0.2}CoO₃, and Gd_{0.5}Sr_{0.5}CoO₃. Alternatively, the B sites of these perovskites may be doped, for example, with Fe or Mn. The electrolyte, cathode, and anode may be porous or dense. As the term "dense" is used
25 herein, a dense material has at least 80% of its theoretical density.

In step **S60**, a cathode film is deposited and patterned to form cathode **30**, as shown in FIG. 2E. Thus, a fuel-cell structure **10** has been formed on substrate **70** by performing the sub-steps **S10** – **S60**. Such a structure is shown in cross-section in FIG. 1D.

In step **S70**, electrical connections are provided by depositing (and, if necessary, patterning) conductive terminal electrodes or pads **60** in electrical contact with the thin film of integral heater element **50**. Terminal electrodes **60** may be formed of relatively thicker films of highly conductive materials, such as copper, silver, or
5 gold, with appropriate conventional interfacial metallurgy if required.

For some embodiments, an additional step may be performed of etching away at least a portion of substrate **70** under the fuel-cell structure to leave the fuel-cell structure supported in membrane form. Such embodiments are shown in FIGS. 1A, 1B, and 1C. To facilitate this method, the substrate may include a sacrificial
10 layer (not shown) that is removed (e.g., by etching) to leave the fuel-cell structure supported in membrane form. If the etching is performed while leaving the fuel-cell structure supported by only one edge, then the resultant fuel-cell structure is cantilevered as shown in FIGS. 1A – 1C.

Materials such as $\text{La}(\text{Sr})\text{Cr}(\text{Mn})\text{O}_3$, $\text{La}(\text{Sr})\text{Cr}(\text{Ti})\text{O}_3$, $\text{Sm}(\text{Sr})\text{CoO}_3$,
15 $\text{La}(\text{Sr})\text{Co}(\text{Fe})\text{O}_3$, where the elements in parentheses are dopants, can be used in the cathode and electrolyte. These materials have both ionic and electrical conductivity.

It will be recognized that a number of fuel cell structures made in accordance with the invention may be combined, e.g., by stacking them electrically coupled in
20 series, in parallel, or in series-parallel combinations to provide a desired voltage, current-delivery capability, or energy. An electronic device may incorporate one fuel cell structure, a number of independent fuel cell structures, or a combination consisting of a number of fuel cell structures electrically coupled. Such a device may be made with a single fuel supply shared by the various component fuel
25 cells.

Such electronic devices can include any of the portable devices mentioned hereinabove, other portable devices, and also non-portable devices, such as uninterruptible power supplies.

The embodiments disclosed also demonstrate new and useful methods of
30 operating and using a fuel cell. A fuel cell having an anode, a cathode, and an

electrolyte, is provided with an integral heater element embedded in and disposed in thermal contact with at least one of the elements (electrolyte, anode, and cathode). The integral heater element has electrical terminals for providing current for heating at least a portion of the fuel cell to a desired temperature
5 before operation, and optionally, during operation of the fuel cell. In an active mode of operation, the fuel cell is heated by feeding current to the integral heater element. In some embodiments, this heating may be limited to an initial startup interval. Thus, a method of operating a fuel cell in accordance with the invention (including introducing fuel into the fuel cell) can include providing heating current
10 for an initial time interval. The initial time interval can begin about the time fuel is first introduced into the fuel cell or just before fuel flow starts.

In a passive mode the same integral heater element or a distinct integral element of similar construction is used as a sensor, sensing fuel cell internal temperature. The internal temperature measured is localized at least within the interior of the
15 fuel cell, or even more locally within the electrolyte portion, the anode, or the cathode of the fuel cell. The sensor signal (e.g., resistance or thermocouple EMF) is used by a conventional feedback control system to control heating current supplied to the integral heater element in its active mode, and optionally to control other fuel-cell operating parameters such as fuel supply rate. Thus a
20 fuel cell's internal temperature, using a localized temperature measurement, can be controlled with a fast response time, to achieve and maintain a desired temperature. The temperature set-point can be chosen for maximum fuel-cell efficiency, minimum startup time, or any suitable desired criterion.

25 INDUSTRIAL APPLICABILITY

Fuel cell integral multifunction heater elements made in accordance with the invention are useful in fuel cells of various types, for improved fuel cell efficiency, for accurately localized temperature sensing within fuel cells, for current collection, and for rapid automatic temperature control of fuel cells. Fuel cells
30 made in accordance with the invention can include, for example, solid oxide fuel cells and other fuel cells in which operating temperatures can exceed 800 °C.

Specially adapted methods performed in accordance with the invention are useful in efficient batch fabrication of fuel cells and in use of such fuel cells, which, conversely, have a structure specially adapted for such methods.

Although the foregoing has been a description and illustration
5 of specific embodiments of the invention, various modifications and changes thereto can be made by persons skilled in the art without departing from the scope and spirit of the invention as defined by the appended claims.

CLAIMS

What is claimed is:

1. A fuel cell of the type having an anode, a cathode, and an electrolyte, said
5 fuel cell comprising:
an integral heater element, and terminals electrically coupled to said integral
heater element for providing heating current for heating said electrolyte to a
desired temperature before operation and during operation of said fuel cell,
said fuel cell being characterized in that:
10 said integral heating element is embedded within at least one of said electrolyte,
anode, and cathode, and disposed in thermal contact with at least one of said
electrolyte, anode, and cathode.
2. A fuel cell of the type having an anode, a cathode, and an electrolyte, said
15 fuel cell comprising:
an integral heater element embedded within at least one of said electrolyte,
anode, and cathode, and disposed in thermal contact with at least one of said
electrolyte, anode, and cathode, and
terminals electrically coupled to said integral heater element for providing heating
20 current for heating at least one of said electrolyte, anode, and cathode to a
desired temperature before operation and during operation of said fuel cell.
3. The fuel cell of claim 1 or 2, wherein said integral heater element comprises a
thin film of a material having suitable resistivity, and said heating current is
25 conducted through said thin film.
4. The fuel cell of any of claims 1 - 3, further comprising a temperature sensor
and a feedback control system responsive to said temperature sensor to control

said integral heater element for achieving and maintaining said desired temperature.

5. A fuel cell comprising:

5 an anode, a cathode, an electrolyte,

integral means for heating at least one of said anode, cathode, and electrolyte, said integral means for heating being disposed within said fuel cell and embedded in thermal contact with at least one of said anode, cathode, and electrolyte, and

10 means for providing heating current to said integral means for heating before operation and during operation of said fuel cell.

6. The fuel cell of claim 5, further comprising means for sensing a temperature within said fuel cell.

15

7. The fuel cell of claim 6, further comprising means for controlling said temperature to a desired temperature, said means for controlling being responsive to said temperature within said fuel cell.

20 8. The fuel cell of claim 5, further comprising means for controlling said integral means for heating, for achieving and maintaining a desired temperature.

9. The fuel cell of claim 5, wherein said integral means for heating further comprises means for collecting current generated by said fuel cell.

25

10. A method for fabricating a fuel cell, said method comprising the steps of:

a) providing a substrate; and

b) forming a fuel-cell structure on said substrate by performing the substeps of:

i) depositing an electrolyte upon said substrate;

ii) depositing and patterning a thin film to form an integral heater element;

iii) optionally depositing an electrolyte over said integral heater element;

5 iv) depositing and patterning an anode upon at least said electrolyte; and

v) depositing and patterning a cathode upon at least said electrolyte, said cathode being spaced from said anode.

11. A fuel cell made by the method of claim 10.

10

12. A method of using a fuel cell having an anode, a cathode, and an electrolyte, said method comprising the steps of:

providing an integral heater element having electrical terminals, said integral heater element being embedded within at least one of the electrolyte, anode, and

15 cathode; and

providing heating current to said electrical terminals for heating at least one of said electrolyte, anode, and cathode of the fuel cell to a desired temperature before operation, and optionally, during operation of said fuel cell.

20 13. The method of claim 12, wherein said integral heater element is disposed in said electrolyte and said heating current is provided to heat said electrolyte.

14. The method of claim 12, further comprising the step of introducing fuel into said fuel cell, and wherein said heating current is provided for an initial time

25 interval, said initial time interval beginning before introducing said fuel into said fuel cell.

15. The method of claim 12, further comprising the step of sensing a fuel-cell internal temperature.

16. The method of claim 15, wherein said fuel-cell internal temperature is
5 localized at least within the interior of said fuel cell.

17. The method of claim 15, wherein said fuel-cell internal temperature is localized within one of said electrolyte, anode, or cathode.

10 18. The method of claim 15, wherein said fuel-cell internal temperature is sensed by determining electrical resistance of said integral heater element.

19. The method of claim 15, further comprising the steps of feeding back a signal responsive to said fuel-cell internal temperature and of controlling said fuel-cell
15 internal temperature to a desired temperature.

20. A method of using a fuel cell having an anode, a cathode, and an electrolyte, said method comprising the steps of:

providing an integral heater element having electrical terminals, said integral
20 heater element being adapted for use as a temperature sensor, and said integral heater element being embedded within at least one of the electrolyte, anode, and cathode;

providing heating current to said electrical terminals for heating said at least one of the electrolyte, anode, and cathode of the fuel cell to a desired temperature
25 before operation, and optionally, during operation of said fuel cell; and
using said integral heater element as a temperature sensor.

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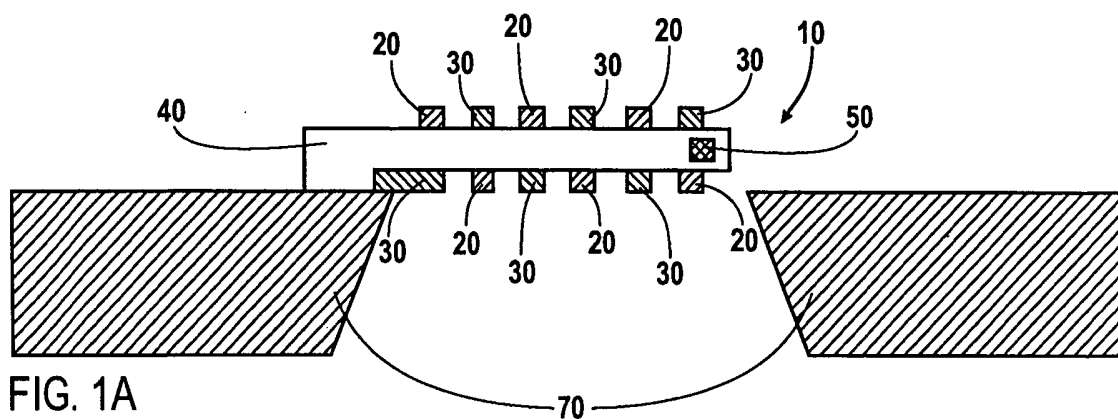


FIG. 1A

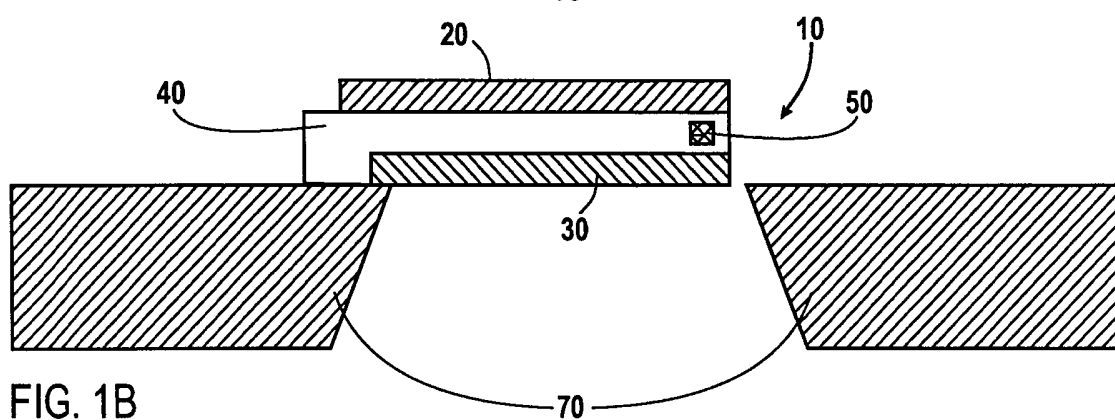


FIG. 1B

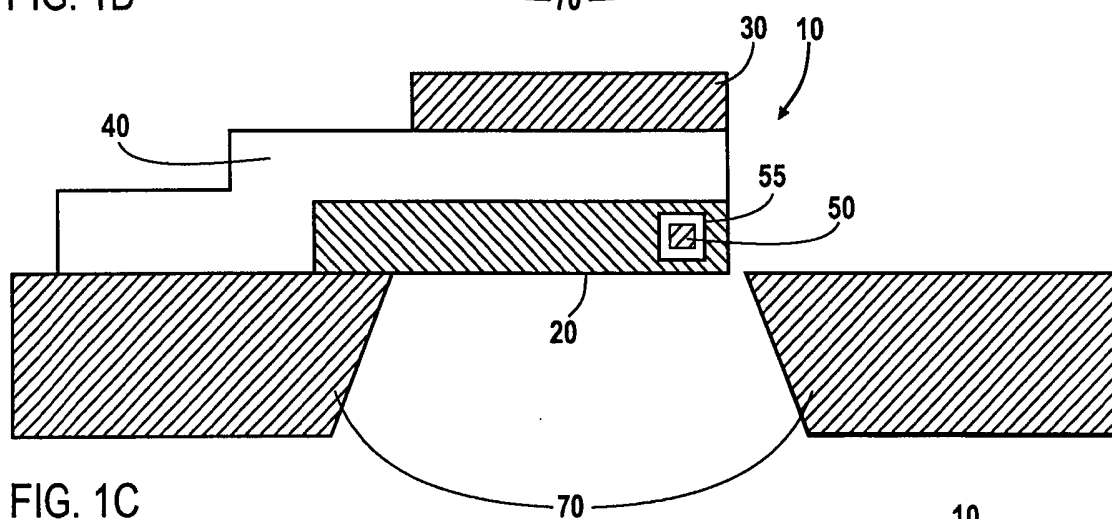


FIG. 1C

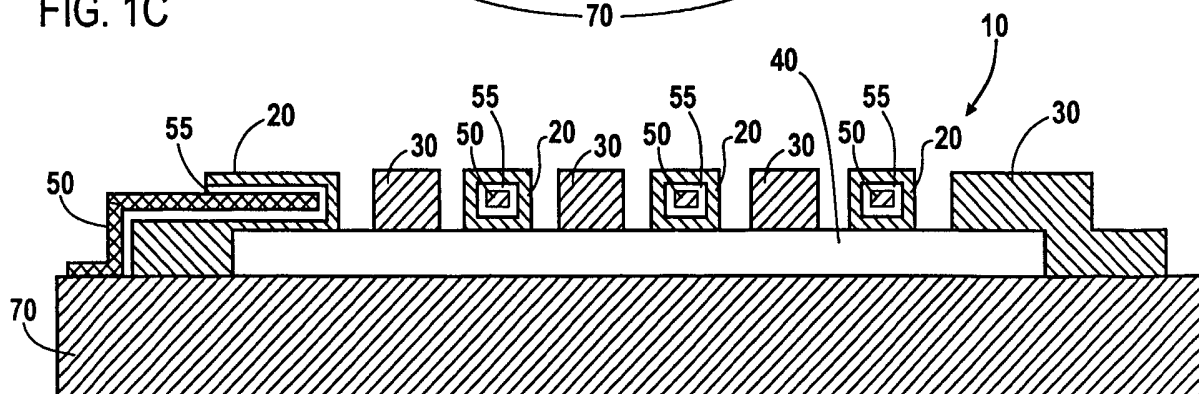


FIG. 1D

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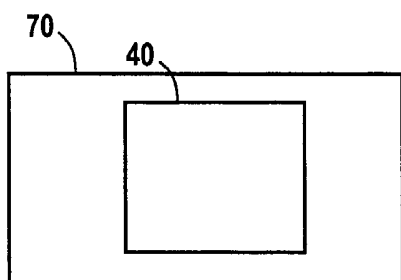


FIG. 2A

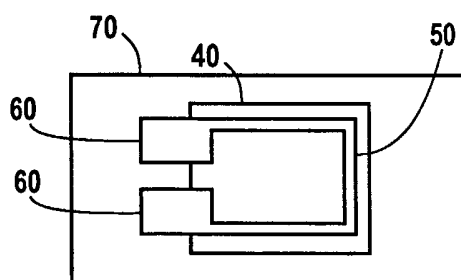


FIG. 2B

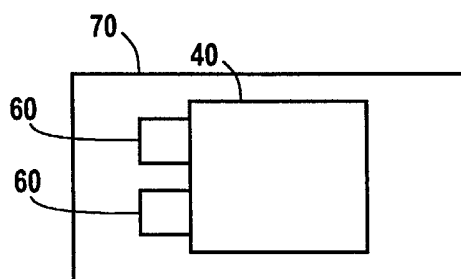


FIG. 2C

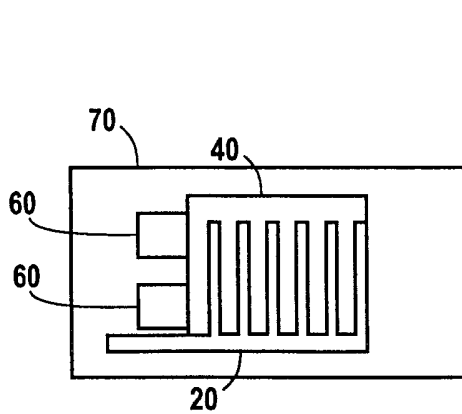


FIG. 2D

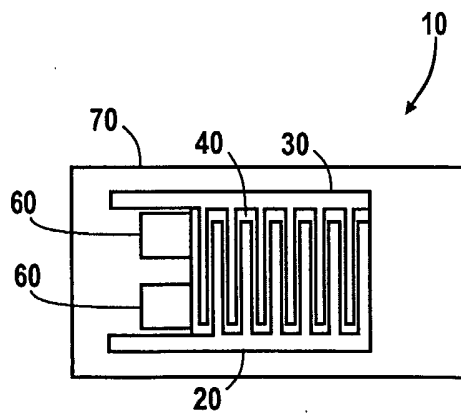


FIG. 2E

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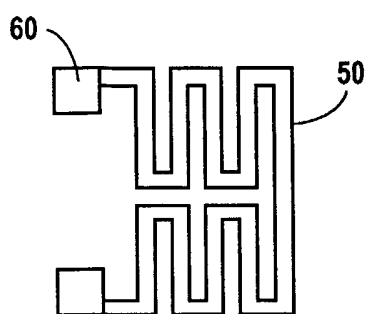


FIG. 3A

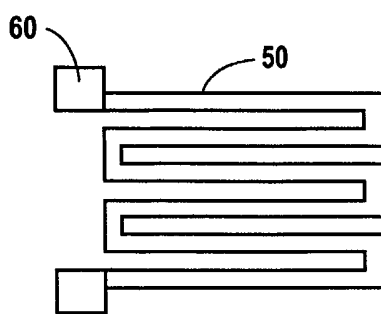


FIG. 3B

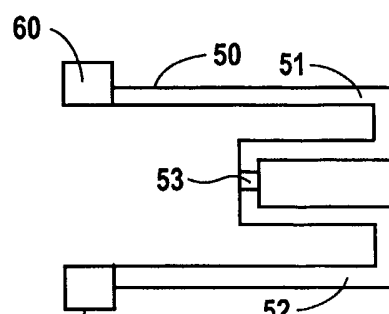


FIG. 3C

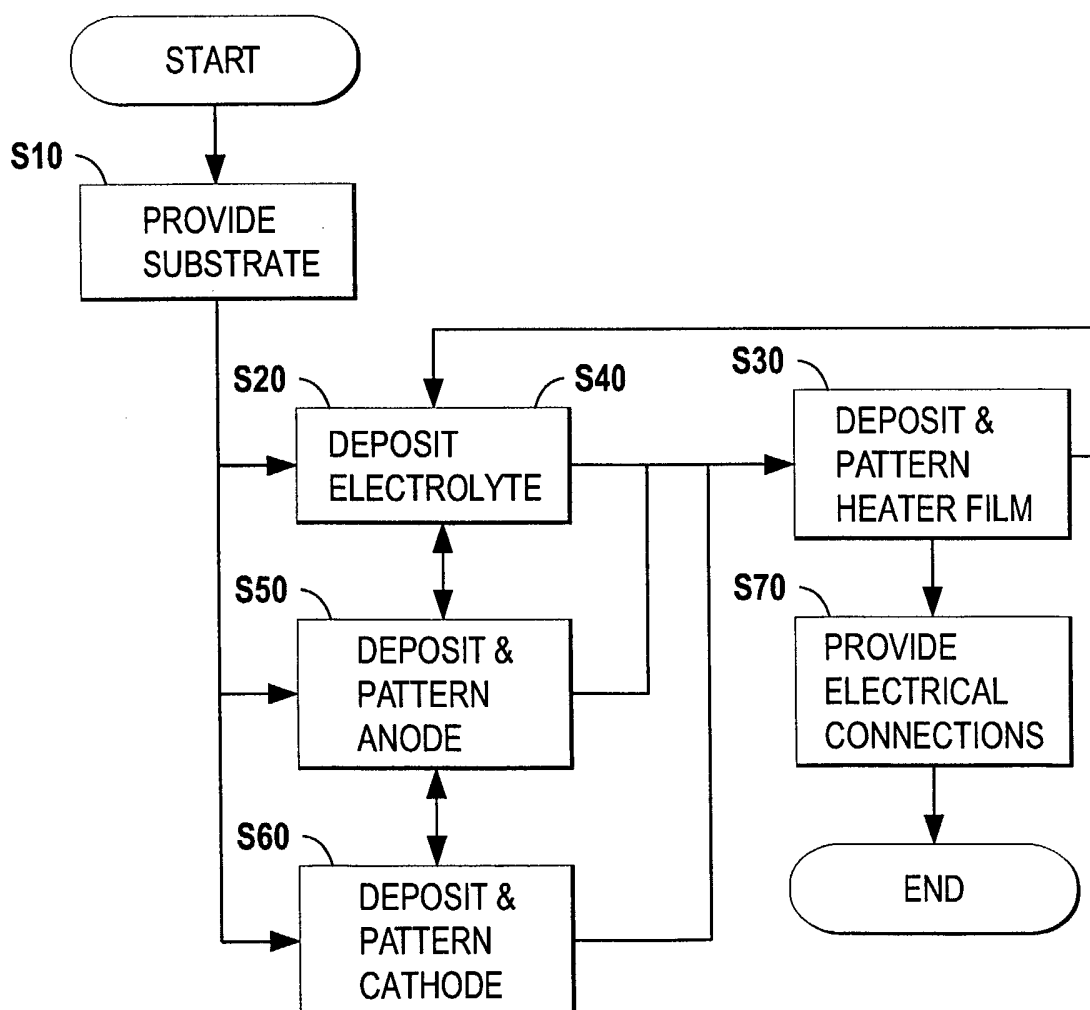


FIG. 4