



Office de la Propriété
Intellectuelle
du Canada

Un organisme
d'Industrie Canada

Canadian
Intellectual Property
Office

An agency of
Industry Canada

CA 2673886 A1 2008/07/17

(21) **2 673 886**

**(12) DEMANDE DE BREVET CANADIEN
CANADIAN PATENT APPLICATION**

(13) A1

(86) Date de dépôt PCT/PCT Filing Date: 2007/12/27
(87) Date publication PCT/PCT Publication Date: 2008/07/17
(85) Entrée phase nationale/National Entry: 2009/06/26
(86) N° demande PCT/PCT Application No.: US 2007/026356
(87) N° publication PCT/PCT Publication No.: 2008/085488
(30) Priorités/Priorities: 2006/12/28 (US60/877,504);
2006/12/28 (US60/877,503)

(51) Cl.Int./Int.Cl. *H01M 8/02* (2006.01)

(71) **Demandeur/Applicant:**
SAINT-GOBAIN CERAMICS & PLASTICS, INC., US

(72) **Inventeur/Inventor:**
NARENDAR, YESHWANTH, US

(74) **Agent:** GOWLING LAFLEUR HENDERSON LLP

(54) Titre : INTERCONNEXIONS EN TITANATE ET EN METAL POUR PILES A COMBUSTIBLE A OXYDE SOLIDE
(54) Title: TITANATE AND METAL INTERCONNECTS FOR SOLID OXIDE FUEL CELLS

(57) Abrégé/Abstract:

A solid oxide fuel cell (SOFC) includes a plurality of sub-cells. Each sub-cell includes a first electrode in fluid communication with a source of oxygen gas, a second electrode in fluid communication with a source of a fuel gas, and a solid electrolyte between the first electrode and the second electrode. The SOFC further includes an interconnect between the sub-cells. In one embodiment, the SOFC has a first surface in contact with the first electrode of each sub-cell and a second surface that is in contact with the second electrode of each sub-cell; and the interconnect consists essentially of a doped M-titanate based perovskite, wherein M is an alkaline earth metal. In another embodiment, the interconnect includes a first layer in contact with the first electrode of each sub-cell, and a second layer in contact with the second electrode of each sub-cell. The first layer includes an electrically conductive material selected from the group consisting of a metal, a metal alloy and a mixture thereof. The second layer includes a doped M-titanate based perovskite, wherein M is an alkaline earth metal. A solid oxide fuel cell described above is formed by connecting each of the sub-cells with an interconnect described above.

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

CORRECTED VERSION

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
17 July 2008 (17.07.2008)

PCT

(10) International Publication Number
WO 2008/085488 A1

(51) International Patent Classification:
H01M 8/02 (2006.01)

(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(21) International Application Number:
PCT/US2007/026356

(22) International Filing Date:
27 December 2007 (27.12.2007)

(25) Filing Language: English

(26) Publication Language: English

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

(30) Priority Data:
60/877,503 28 December 2006 (28.12.2006) US
60/877,504 28 December 2006 (28.12.2006) US

(71) Applicant (for all designated States except US): SAINT-GOBAIN CERAMICS & PLASTICS, INC. [US/US]; One New Bond Street, Worcester, MA 01615 (US).

Published:

— with international search report

(72) Inventor; and
(75) Inventor/Applicant (for US only): NARENDAR, Yeshwanth [IN/US]; 34 Carlisle Road, Westford, MA 01886 (US).

(48) Date of publication of this corrected version:

9 October 2008

(74) Agents: PIERCE, N., Scott et al.; Hamilton, Brook, Smith & Reynolds, P.C., 530 Virginia Road, P.O. Box 9133, Concord, MA 01742-9133 (US).

(15) Information about Correction:
see Notice of 9 October 2008

(54) Title: TITANATE AND METAL INTERCONNECTS FOR SOLID OXIDE FUEL CELLS

(57) Abstract: A solid oxide fuel cell (SOFC) includes a plurality of sub-cells. Each sub-cell includes a first electrode in fluid communication with a source of oxygen gas, a second electrode in fluid communication with a source of a fuel gas, and a solid electrolyte between the first electrode and the second electrode. The SOFC further includes an interconnect between the sub-cells. In one embodiment, the SOFC has a first surface in contact with the first electrode of each sub-cell and a second surface that is in contact with the second electrode of each sub-cell; and the interconnect consists essentially of a doped M-titanate based perovskite, wherein M is an alkaline earth metal. In another embodiment, the interconnect includes a first layer in contact with the first electrode of each sub-cell, and a second layer in contact with the second electrode of each sub-cell. The first layer includes an electrically conductive material selected from the group consisting of an metal, a metal alloy and a mixture thereof. The second layer includes a doped M-titanate based perovskite, wherein M is an alkaline earth metal. A solid oxide fuel cell described above is formed by connecting each of the sub-cells with an interconnect described above.

A1

WO 2008/085488

TITANATE AND METAL INTERCONNECTS FOR SOLID OXIDE FUEL CELLS

RELATED APPLICATIONS

This application claims the benefit of U.S. Provisional Application Nos. 60/877,503 and 60/877,504, both filed December 28, 2006. The entire teachings of these applications are incorporated herein by reference.

5 BACKGROUND

A fuel cell is a device that generates electricity by a chemical reaction. Among various fuel cells, solid oxide fuel cells use a hard, ceramic compound of metal (e.g., calcium or zirconium) oxide as an electrolyte. Typically, in the solid oxide fuel cells, an oxygen gas, such as O₂, is reduced to oxygen ions (O²⁻) at the 10 cathode, and a fuel gas, such as H₂ gas, is oxidized with the oxygen ions to form water at the anode.

Interconnects are one of the critical issues limiting commercialization of solid oxide fuel cells. Currently, most companies and researchers working with planar cells are using coated metal interconnects. For example, ferritic stainless 15 steel based metal interconnects, such as Crofer 22 APU, and powder metallurgy formulations from Plansee AG in Austria, have good CTE (coefficient of thermal expansion) mismatch and good compatibility with solid oxide fuel cells. However, oxidation of metal interconnects during operation, thereby forming metal oxides, such as Cr₂O₃, and subsequent migration of metals of the metal oxides (e.g., 20 chromium migration) to an electrode layer and/or electrode-electrolyte interface is one of the primary mechanisms leading to performance degradation in solid oxide fuel cells.

While metal interconnects are relatively easy to fabricate and process, they generally suffer from high power degradation rates (e.g. 10%/1,000 h) partly due to 25 formation of metal oxide, such as Cr₂O₃, at an interconnect-anode/cathode interface during operation. Ceramic interconnects based on lanthanum chromites (LaCrO₃)

have lower degradation rates than metal interconnects partly due to relatively high thermodynamic stability and low Cr vapor pressure of LaCrO_3 compared to Cr_2O_3 formed on interfaces of the metal interconnects and electrode. However, doped LaCrO_3 generally suffers from dimensional changes, such as warping or some other 5 forms of distortion, and consequent seal failures in reducing conditions. Another issue related to LaCrO_3 is its relatively low sinterability.

Therefore, there is a need for development of new interconnects for solid oxide fuel cells, addressing one or more of the aforementioned problems.

10 SUMMARY OF THE INVENTION

The invention is directed to a solid oxide fuel cell (SOFC) that includes a plurality of sub-cells and to a method of preparing the SOFC. Each sub-cell includes a first electrode in fluid communication with a source of oxygen gas, a second electrode in fluid communication with a source of a fuel gas, and a solid 15 electrolyte between the first electrode and the second electrode. The SOFC further includes an interconnect between the sub-cells.

In one embodiment, the interconnect has a first surface in contact with the first electrode of each sub-cell, and a second surface that is in contact with the second electrode of each sub-cell. The interconnect consists essentially of a doped 20 M-titanate based perovskite, wherein M is an alkaline earth metal.

In another embodiment, the interconnect includes a first layer in contact with the first electrode of each sub-cell, and a second layer in contact with the second electrode of each sub-cell. The first layer includes an electrically conductive material selected from the group consisting of a metal, a metal alloy and a mixture 25 thereof. The second layer includes a doped M-titanate based perovskite, wherein M is an alkaline earth metal.

The invention also includes a method of forming a solid oxide fuel cell described above. The method includes connecting each of the sub-cells with an interconnect described above.

30 It is believed that the doped M-titanates (e.g., MTiO_3) in the invention, particularly, n-doped M-titanates, such as n-doped SrTiO_3 or CaTiO_3 , exhibit less oxygen vacancy formation during operation of SOFCs, as compared to conventional

p-doped LaCrO_3 , thereby limiting or eliminating lattice expansion problems associated with conventional p-doped LaCrO_3 . In addition, the doped M-titanates can function as an effective oxidation barrier to thereby extend operation life of SOFCs.

5 BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic cross-sectional view of one embodiment of the invention.

FIG. 2 is a schematic cross-sectional view of another embodiment of the invention.

10 FIG. 3 is a schematic diagram of an embodiment of a fuel cell of the invention, having a planar, stacked design.

FIG. 4 is a schematic diagram of still another embodiment of a fuel cell of the invention, having a tubular design.

DETAILED DESCRIPTION OF THE INVENTION

15 The foregoing will be apparent from the following more particular description of example embodiments of the invention, as illustrated in the accompanying drawings in which like reference characters refer to the same parts throughout the different views. The drawing is not necessarily to scale, emphasis instead being placed upon illustrating embodiments of the present invention.

20 FIG. 1 shows fuel cell 10 of the invention. Fuel cell 10 includes a plurality of sub-cells 12. Each sub-cell 12 includes first electrode 14 and second electrode 16. Typically, first and second electrodes 14 and 16 are porous. In fuel cell 10, first electrode 14 at least in part defines a plurality of first gas channels 18 in fluid communication with a source of oxygen gas, such as air. Second electrode 16 at 25 least in part defines a plurality of second gas channels 20 in fluid communication with a fuel gas source, such as H_2 gas or a natural gas which can be converted into H_2 *in situ* at second electrode 16.

30 Although, in FIG. 1, first electrodes 14 and second electrodes 16 define a plurality of gas channels 18 and 20, other types of gas channels, such as a microstructured channel (e.g., grooved channel) at each of the electrodes or as a separate layer in fluid communication with the electrode, can also be used in the

invention. For example, referring to FIG. 2, first gas channel 18 is defined at least in part by first electrode 14 and by at least in part by interconnect 24, and second gas channel 20 is defined at least in part by second electrode 16 and by at least in part by interconnect 24.

5 Any suitable cathode materials known in the art can be used for first electrode 14, for example, in "High Temperature Solid Oxide Fuel Cells: Fundamentals, Design and Applications," pp. 119-143, Dinghal, *et al.* Ed., Elsevier Ltd. (2003), the entire teachings of which are incorporated herein by reference. In one embodiment, first electrode 14 includes a La-manganate (e.g., $\text{La}_{1-a}\text{MnO}_3$, where 10 a is equal to or greater than zero, and equal to or less than 0.1) or La-ferrite based material. Typically, the La-manganate or La-ferrite based material is doped with one or more suitable dopants, such as Sr, Ca, Ba, Mg, Ni, Co or Fe. Examples of doped La-manganate based materials include LaSr-manganates (LSM) (e.g., $\text{La}_{1-k}\text{Sr}_k\text{MnO}_3$, where k is equal to or greater than 0.1, and equal to or less than 0.3, 15 $(\text{La} + \text{Sr})/\text{Mn}$ is in a range of between about 1.0 and about 0.95 (molar ratio)) and LaCa-manganates (e.g., $\text{La}_{1-k}\text{Ca}_k\text{MnO}_3$, k is equal to or greater than 0.1, and equal to or less than 0.3, $(\text{La} + \text{Ca})/\text{Mn}$ is in a range of between about 1.0 and about 0.95 (molar ratio)). Examples of doped La-ferrite based materials include LaSrCo-ferrite 20 (LSCF) (e.g. $\text{La}_{1-q}\text{Sr}_q\text{Co}_{1-j}\text{Fe}_j\text{O}_3$, where each of q and j independently is equal to or greater than 0.1, and equal to or less than 0.4, $(\text{La}+\text{Sr})/(\text{Fe}+\text{Co})$ is in a range of between about 1.0 and about 0.95 (molar ratio)). In one specific embodiment, first electrode 14 includes at least one of a LaSr-manganate (LSM) (e.g., $\text{La}_{1-k}\text{Sr}_k\text{MnO}_3$) and a LaSrCo-ferrite (LSCF). Common examples include $(\text{La}_{0.8}\text{Sr}_{0.2})_{0.98}\text{MnO}_{3\pm\delta}$ (δ is equal to or greater than zero, and equal to or less than 0.3) and 25 $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$.

Any suitable anode materials known in the art can be used for second electrode 16, for example, in "High Temperature Solid Oxide Fuel Cells: Fundamentals, Design and Applications," pp. 149-169, Dinghal, *et al.* Ed., Elsevier Ltd. (2003), the entire teachings of which are incorporated herein by reference. In 30 one embodiment, second electrode 16 includes a nickel (Ni) cermet. As used herein, the phrase "Ni cermet" means a ceramic metal composite that includes Ni, such as about 20wt% - 70wt% of Ni. Examples of Ni cermets are materials that include Ni

and yttria-stabilized zirconia (YSZ), such as ZrO_2 containing about 15 wt% of Y_2O_3 , and materials that include Ni and Y-zirconia or Sc-zirconia. An additional example of an anode material is Cu-cerium oxide. A specific example of an Ni cermet includes 67 wt%Ni and 33wt%YSZ.

5 Typically, the thickness of each of first and second electrodes 14 and 16 is, independently, in a range of between about 0.5 mm and about 2 mm. Specifically, the thickness of each of first and second electrodes 14 and 16 is, independently, in a range of between about 1 mm and about 2 mm.

Solid electrolyte 22 is between first electrode 14 and second electrode 16.

10 Any suitable solid electrolytes known in the art can be used in the invention, such as those described, in "High Temperature Solid Oxide Fuel Cells: Fundamentals, Design and Applications," pp. 83-112, Dinghal, *et al.* Ed., Elsevier Ltd. (2003), the entire teachings of which are incorporated herein by reference. Examples include ZrO_2 based materials, such as Sc_2O_3 -doped ZrO_2 , Y_2O_3 -doped ZrO_2 , and Yb_2O_3 -doped ZrO_2 ; CeO_2 based materials, such as Sm_2O_3 -doped CeO_2 , Gd_2O_3 -doped CeO_2 , Y_2O_3 -doped CeO_2 and CaO -doped CeO_2 ; Ln -gallate based materials ($\text{Ln} = \text{a lanthanide}$, such as La, Pr, Nd or Sm), such as LaGaO_3 doped with Ca, Sr, Ba, Mg, Co, Ni, Fe or a mixture thereof (e.g., $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_3$, $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.15}\text{Co}_{0.05}\text{O}_3$, $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_3$, LaSrGaO_4 , $\text{LaSrGa}_3\text{O}_7$ or $20 \text{La}_{0.9}\text{A}_{0.1}\text{Ga}_3$ where $\text{A} = \text{Sr, Ca or Ba}$); and mixtures thereof. Other examples include doped yttrium-zirconate (e.g., YZr_2O_7), doped gadolinium-titanate (e.g., $\text{Gd}_2\text{Ti}_2\text{O}_7$) and brownmillerites (e.g., $\text{Ba}_2\text{In}_2\text{O}_6$ or $\text{Ba}_2\text{In}_2\text{O}_5$). In a specific embodiment, electrolyte 22 includes ZrO_2 doped with 8 mol% Y_2O_3 (i.e., 8 mol% Y_2O_3 -doped ZrO_2 .)

25 Typically, the thickness of solid electrolyte 22 is in a range of between about 5 μm and about 20 μm , such as between about 5 μm and about 10 μm . Alternatively, the thickness of solid electrolyte 22 is thicker than about 100 μm (e.g., between about 100 μm and about 500 100 μm). In this embodiment employing solid electrolyte 22 having a thickness greater than about 100 μm , solid electrolyte 22 can provide structural support for fuel cell 10.

Fuel cell 10 further includes interconnect 24 between cells 12. Referring to FIG. 1, in one embodiment, interconnect 24 has first surface 26 in contact with first

electrode 14, and second surface 28 in contact with second electrode 16.

Interconnect 24 consists essentially of a doped M-titanate based perovskite, wherein M is an alkaline earth metal. As used herein, the phrase "consists essentially of" means that interconnect 24 does not include any other metal oxides that may

5 substantially affect the property of interconnect 24. Examples of suitable dopant materials include La, Y, Nb, Mn, V, Cr, W, Mo, Si, Sr, Ca, Mg, Ni, Co, V and Ti.

In a specific embodiment, the M-titanate based perovskite is Sr-titanate (e.g., SrTiO₃), Ca-titanate (e.g., CaTiO₃), Ba-titanate (e.g., BaTiO₃), or Mg-titanate (e.g., MgTiO₃). In another specific embodiment, the M-titanate based perovskite is n-

10 doped, such as n-doped Sr-titanate or n-doped Ca-titanate. In yet another specific embodiment, the M-titanate based perovskite is a Sr-titanate doped with at least one dopant selected from the group consisting of La, Y, Nb, Mn, V, Cr, W, Mo and Si.

In another embodiment, as shown in FIG. 2, interconnect 24 includes first layer 36 in contact with first electrode 14, and second layer 38 in contact with 15 second electrode 16. First layer 36 includes an electrically conductive material selected from a metal, a metal alloy and a mixture thereof. Examples of the electrically conductive materials suitable for first layer 36 include Cr-based alloys, ferritic steels, Ni-based super alloys and Ni-Fe-Y₂O₃ alloys. Second layer 38 includes a doped M-titanate based perovskite, wherein M is an alkaline earth metal. 20 Preferably, the material included in second layer 38 is n-doped. Examples of suitable dopant materials include La, Y, Nb, Mn, V, Cr, W, Mo, Si, Sr, Ca, Mg, Ni, Co, V and Ti. Suitable n-dopants include La, Y, Nb, Mn, V, Cr, W, Mo and Si. In a specific embodiment, the M-titanate based perovskite is Sr-titanate (e.g., SrTiO₃), Ca- 25 titanate (e.g., CaTiO₃), Ba-titanate (e.g., BaTiO₃), or Mg-titanate (e.g., MgTiO₃). In another specific embodiment, the M-titanate based perovskite is n-doped, such as n-doped Sr-titanate or n-doped Ca-titanate. In yet another specific embodiment, the M-titanate based perovskite is a Sr-titanate doped with at least one dopant selected from the group consisting of La, Y, Nb, Mn, V, Cr, W, Mo and Si.

M-titanate based perovskite has the perovskite structure known in the art, for 30 example, in "High Temperature Solid Oxide Fuel Cells: Fundamentals, Design and Applications," pp. 120-123, Dinghal, et al. Ed., Elsevier Ltd. (2003), the entire teachings of which are incorporated herein by reference. The perovskite structure is

adopted by many oxides that have the chemical formula ABO_3 . The general crystal structure is a primitive cube with the A-cation in the center of a unit cell, the B-cation at the corners of the unit cell, and the anion (i.e., O^{2-}) at the centers of each edge of the unit cell. The idealized structure is a primitive cube, but differences in 5 ratio between the A and B cations can cause a number of different so-called distortions, of which tilting is the most common one. As used herein, the phrase "M-titanate based perovskite" also includes such distortions. In the "M-titanate based perovskites," M atoms generally occupy the A-cation sites, while Ti atoms generally occupy the B-cation sites.

10 Interconnect 24 can be in any shape, such as a planar shape (*see* FIGs. 1 and 2) or a microstructured (e.g., grooved) shape (*see* FIG. 3). In one specific embodiment, at least one interconnect 24 of fuel cell 10 is substantially planar.

In one embodiment, the thickness of interconnect 24 is in a range of between 15 about 10 μm and about 1,000 μm . Alternatively, the thickness of interconnect 24 is in a range of between about 0.005 mm and about 2.0 mm. In one specific embodiment, the thickness of interconnect 24 is in a range of 10 μm and about 500 μm . In another embodiment, the thickness of interconnect 24 is in a range of 10 μm and about 200 μm . In yet another embodiment, the thickness of interconnect 24 is between about 10 μm and about 100 μm . In yet another embodiment, the thickness of interconnect 24 is 20 between about 10 μm and about 75 μm . In yet another embodiment, the thickness of interconnect 24 is between about 15 μm and about 65 μm .

In one specific embodiment, first electrode 14 and/or second electrode 16 has 25 a thickness of between about 0.5 mm and about 2 mm thick, more specifically between about 1 mm and about 2 mm thick; and interconnect 24 has a thickness of between about 10 μm and about 200 μm , specifically between about 10 μm and about 200 μm , and more specifically between about 10 μm and about 100 μm .

In another specific embodiment, second layer 28 includes a SrLa-titanate 30 based perovskite described above; and interconnect 24 has a thickness of between about 10 μm and about 100 μm , specifically between about 10 μm and about 75 μm , and more specifically between about 15 μm and about 65 μm .

In yet another specific embodiment, at least one cell 12 includes porous first and second electrodes 14 and 16, each of which is between about 0.5 mm and about 2 mm thick, more specifically between about 1 mm and about 2 mm thick; solid electrolyte 22 has a thickness of between about 5 μm and about 20 μm ; and 5 interconnect 24 is substantially planar and has a thickness of between about 10 μm and about 200 μm .

In yet another specific embodiment, interconnect 24 is substantially planar; each of first and second electrodes 14 and 16 is porous; and first electrode 14 includes a La-manganate or La- ferrite based material (e.g., $\text{La}_{1-k}\text{Sr}_k\text{MnO}_3$ or $\text{La}_{1-q}\text{Sr}_q\text{Co}_j\text{Fe}_{1-j}\text{O}_3$, wherein values of each of k, 1 and j independently are as described above), and second electrode 16 includes a Ni cermet (e.g., 67 wt% Ni and 33wt% YSZ). In one aspect of this specific embodiment, electrolyte 22 includes 8 mol% Y_2O_3 -doped ZrO_2 .

In yet another specific embodiment, first electrode 14 includes 15 $(\text{La}_{0.8}\text{Sr}_{0.2})_{0.98}\text{MnO}_{3\pm\delta}$ or $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$; and second electrode 16 includes 67 wt% Ni and 33wt% YSZ. In this embodiment, specifically, electrolyte 22 includes 8 mol% Y_2O_3 -doped ZrO_2 .

Fuel cell 10 of the invention can include any suitable number of a plurality of sub-cells 12. In one embodiment, fuel cell 10 of the invention includes at least 20 30-50 sub-cells 12. Sub-cells 12 of fuel cell 10 can be connected in series or in parallel.

A fuel cell of the invention can be a planar stacked fuel cell, as shown in FIG. 3. Alternatively, as shown in FIG. 4, a fuel cell of the invention can be a tubular fuel cell. Fuel cells shown in FIGs. 3 and 4 independently have the characteristics, including specific variables, as described for fuel cell 10 shown in 25 FIGs. 1 and 2 (for clarity, details of cell components are not depicted in FIGs. 3 and 4). Typically, in the planar design, as shown in FIG. 3, the components are assembled in flat stacks, with air and fuel flowing through channels built into the interconnect. Typically, in the tubular design, as shown in FIG. 4, the components are assembled in the form of a hollow tube, with the cell constructed in layers 30 around a tubular cathode; air flows through the inside of the tube and fuel flows around the exterior.

The invention also includes a method of forming fuel cells as described above. The method includes forming a plurality of sub-cells 12 as described above, and connecting each sub-cell 12 with interconnect 24. Fabrication of sub-cells 12 and interconnect 24 can employ any suitable techniques known in the art, for 5 example, in "High Temperature Solid Oxide Fuel Cells: Fundamentals, Design and Applications," pp. 83-225, Dinghal, *et al.* Ed., Elsevier Ltd. (2003), the entire teachings of which are incorporated herein by reference. For example, planar stacked fuel cells of the invention can be fabricated by particulate processes or deposition processes. Tubular fuel cells of the invention can be fabricated by having 10 the cell components in the form of thin layers on a porous cylindrical tube, such as calcia-stabilized zirconia.

Typically, a suitable particulate process, such as tape casting or tape calendering, involves compaction of powders, such as ceramic powders, into fuel cell components (e.g., electrodes, electrolytes and interconnects) and densification at 15 elevated temperatures. For example, suitable powder materials for electrolytes, electrodes or interconnects of the invention, are made by solid state reaction of constituent oxides. Suitable high surface area powders can be precipitated from nitrate and other solutions as a gel product, which are dried, calcined and comminuted to give crystalline particles. The deposition processes can involve 20 formation of cell components on a support by a suitable chemical or physical process. Examples of the deposition include chemical vapor deposition, plasma spraying and spray pyrolysis.

In one specific embodiment, interconnect 24 consists essentially of a doped M-titanate based perovskite, and is prepared by disposing a material of a doped M-titanate based perovskite over either electrode 14 or 16, and sintering the material to 25 form interconnect 24. Generally, the material of a doped M-titanate based perovskite is disposed at a temperature in a range of between about 50 °C and about 80 °C with a loading of between about 5 and about 50 tons. Generally, the material of a doped M-titanate based perovskite is sintered to form interconnect 24 having a 30 high theoretical density (e.g., greater than about 90% theoretical density, or greater than about 95% theoretical density).

In another specific embodiment, interconnect 24 includes first layer 36 and second layer 38, and is formed by depositing an electrically conductive material over electrode 14 using any suitable deposition method known in the art, such as chemical vapor deposition to from first layer 36, and disposing a material of a doped 5 M-titanate based perovskite over first layer 36. Generally, the material of a doped M-titanate based perovskite is disposed at a temperature in a range of between about 50 °C and about 80 °C with a loading of between about 5 and about 50 tons. Generally, the material of a doped M-titanate based perovskite is sintered to form 10 interconnect 24 having a high theoretical density (e.g., greater than about 90% theoretical density, or greater than about 95% theoretical density).

The SOFCs of the invention can be portable. Also, the SOFCs of the invention, can be employed as a source of electricity in homes, for example, to generate hot water.

15 EQUIVALENT

While this invention has been particularly shown and described with references to example embodiments thereof, it will be understood by those skilled in the art that various changes in form and details may be made therein without departing from the scope of the invention encompassed by the appended claims.

CLAIMS

What is claimed is:

1. A solid oxide fuel cell, comprising:
 - a) a plurality of sub-cells, each sub-cell including:
 - i) a first electrode in fluid communication with a source of oxygen gas;
 - ii) a second electrode in fluid communication with a source of a fuel gas; and
 - iii) a solid electrolyte between the first electrode and the second electrode; and
 - b) an interconnect between the sub-cells, the interconnect having a first surface in contact with the first electrode of each sub-cell and a second surface that is in contact with the second electrode of each sub-cell, the interconnect consisting essentially of a doped M-titanate based perovskite, wherein M is an alkaline earth metal.
- 15 2. The solid oxide fuel cell of Claim 1, wherein each sub-cell further includes a first gas channel in fluid communication with the oxygen gas source and with the first electrode, and a second gas channel in fluid communication with the fuel gas source and with the second electrode.
- 20 3. The solid oxide fuel cell of Claim 2, wherein the first electrode at least in part defines the first gas channel, and the second electrode at least in part defines the second gas channel.
- 25 4. The solid oxide fuel cell of Claim 1, wherein each of the first and second electrodes is porous.

5. The solid oxide fuel cell of Claim 4, wherein the interconnect is substantially planar.
6. The solid oxide fuel cell of Claim 1, wherein the M-titanate based perovskite is selected from the group consisting of Sr-titanate, Ca-titanate, Ba-titanate and Mg-titanate.
- 10 7. The solid oxide fuel cell of Claim 6, wherein the interconnect includes an n-doped Sr-titanate or n-doped Ca-titanate.
8. The solid oxide fuel cell of Claim 7, wherein the interconnect includes a Sr-titanate doped with at least one dopant selected from the group consisting of La, Y, Nb, Mn, V, Cr, W, Mo and Si.
- 15 9. The solid oxide fuel cell of Claim 1, wherein the solid electrolyte includes at least one material selected from the group consisting of ZrO₂ based material, CeO₂ based material and lanthanide-gallate based material.
- 20 10. The solid oxide fuel cell of Claim 1, wherein the first electrode includes a La-manganate based material.
11. The solid oxide fuel cell of Claim 1, wherein the second electrode includes a nickel cermet.
- 25 12. The solid oxide fuel cell of Claim 1, wherein the thickness of each of the first and second electrodes of at least one of the cells is in a range of between about 1 mm and about 2 mm.
- 30 13. The solid oxide fuel cell of Claim 12, wherein the thickness of the interconnect is in a range of between about 10 μ m and about 1,000 μ m.

14. The solid oxide fuel cell of Claim 13, wherein the thickness of the interconnect is in a range of between about 10 μm and about 200 μm .

5 15. The solid oxide fuel cell of Claim 14, wherein the thickness of the interconnect is in a range of between about 50 μm and about 200 μm .

10 16. The solid oxide fuel cell of Claim 1, wherein the cells are connected with each other in series.

15 17. A method of forming a solid oxide fuel cell cell that includes a plurality of sub-cells, comprising the step of connecting each of the sub-cells with an interconnect, wherein each sub-cell includes:

15 i) a first electrode in fluid communication with a source of oxygen gas,

ii) a second electrode in fluid communication with a source of a fuel gas, and

iii) a solid electrolyte between the first electrode and the second electrode, and

20 wherein the interconnect consists essentially of a doped M-titanate based perovskite, wherein M is an alkaline earth metal, the interconnect having a first surface in contact with the first electrode of each cell and a second surface that is in contact with the second electrode of each sub-cell.

25 18. A solid oxide fuel cell, comprising:

30 a) a plurality of sub-cells, each sub-cell including:

i) a first electrode in fluid communication with a source of oxygen gas;

ii) a second electrode in fluid communication with a source of a fuel gas; and

- iii) a solid electrolyte between the first electrode and the second electrode; and
- b) an interconnect between the sub-cells, the interconnect including:
 - 5 i) a first layer that includes an electrically conductive material selected from the group consisting of a metal, a metal alloy and a mixture of metals, wherein the first layer is in contact with the first electrode of each sub-cell; and
 - 10 ii) a second layer that includes a doped M-titanate based perovskite, wherein M is an alkaline earth metal, and wherein the second layer is in contact with the second electrode of each sub-cell.

15 19. The solid oxide fuel cell of Claim 18, wherein each sub-cell further includes a first gas channel in fluid communication with the oxygen gas source, and a second gas channel in fluid communication with the fuel gas source.

20 20. The solid oxide fuel cell of Claim 19, wherein the first electrode at least in part defines the first gas channel, and the second electrode at least in part defines the second gas channel.

25 21. The solid oxide fuel cell of Claim 18, wherein each of the first and second electrodes is porous.

22. The solid oxide fuel cell of Claim 21, wherein the interconnect is substantially planar.

30 23. The solid oxide fuel cell of Claim 18, wherein the doped M-titanate based perovskite is selected from the group consisting of a doped Sr-

titanate, a doped Ca-titanate, a doped Ba-titanate and a doped Mg-titanate.

24. The solid oxide fuel cell of Claim 23, wherein the doped M-titanate
5 based perovskite includes an n-doped Sr-titanate or n-doped Ca-titanate.
25. The solid oxide fuel cell of Claim 24, wherein the doped M-titanate
10 based perovskite includes a Sr-titanate doped with at least one dopant
selected from the group consisting of La, Y, Nb, Mn, V, Cr, W, Mo
and Si.
26. The solid oxide fuel cell of Claim 18, wherein the solid electrolyte
includes at least one material selected from the group consisting of
15 ZrO_2 based material, CeO_2 based material and lanthanide-gallate
based material.
27. The solid oxide fuel cell of Claim 18, wherein the first electrode
includes a La-manganate based material.
20
28. The solid oxide fuel cell of Claim 18, wherein the second electrode
includes a Ni cermet.
29. The solid oxide fuel cell of Claim 18, wherein the first layer of the
25 interconnect includes at least one material selected from the group
consisting of Cr-based alloys, ferritic steels, Ni-based super alloys
and Ni-Fe- Y_2O_3 alloy.
30. The solid oxide fuel cell of Claim 18, wherein the thickness of each
30 of the first and second electrodes of at least one of the cells is in a
range of between about 1 mm and about 2 mm.

31. The solid oxide fuel cell of Claim 30, wherein the thickness of the interconnect is in a range of between about 10 μm and about 1,000 μm .
- 5 32. The solid oxide fuel cell of Claim 31, wherein the thickness of the interconnect is in a range of between about 10 μm and about 200 μm .
33. The solid oxide fuel cell of Claim 32, wherein the thickness of the interconnect is in a range of between about 50 μm and about 200 μm .
- 10 34. A method of forming a solid oxide fuel cell that includes a plurality of sub-cells, comprising the step of connecting each of the sub-cells with an interconnect, wherein each sub-cell includes:
 - i) a first electrode in fluid communication with a source of oxygen gas,
 - 15 ii) a second electrode in fluid communication with a source of a fuel gas, and
 - iii) a solid electrolyte between the first electrode and the second electrode, and
- 20 wherein the interconnect includes:
 - i) a first layer of an electrically conductive metal, a metal alloy or a mixture of metals, wherein the first layer is in contact with the first electrode of each cell; and
 - ii) a second layer that includes a doped M-titanate based perovskite, wherein M is an alkaline earth metal, and wherein the second layer is in contact with the second electrode of each sub-cell.
- 25

1 / 3

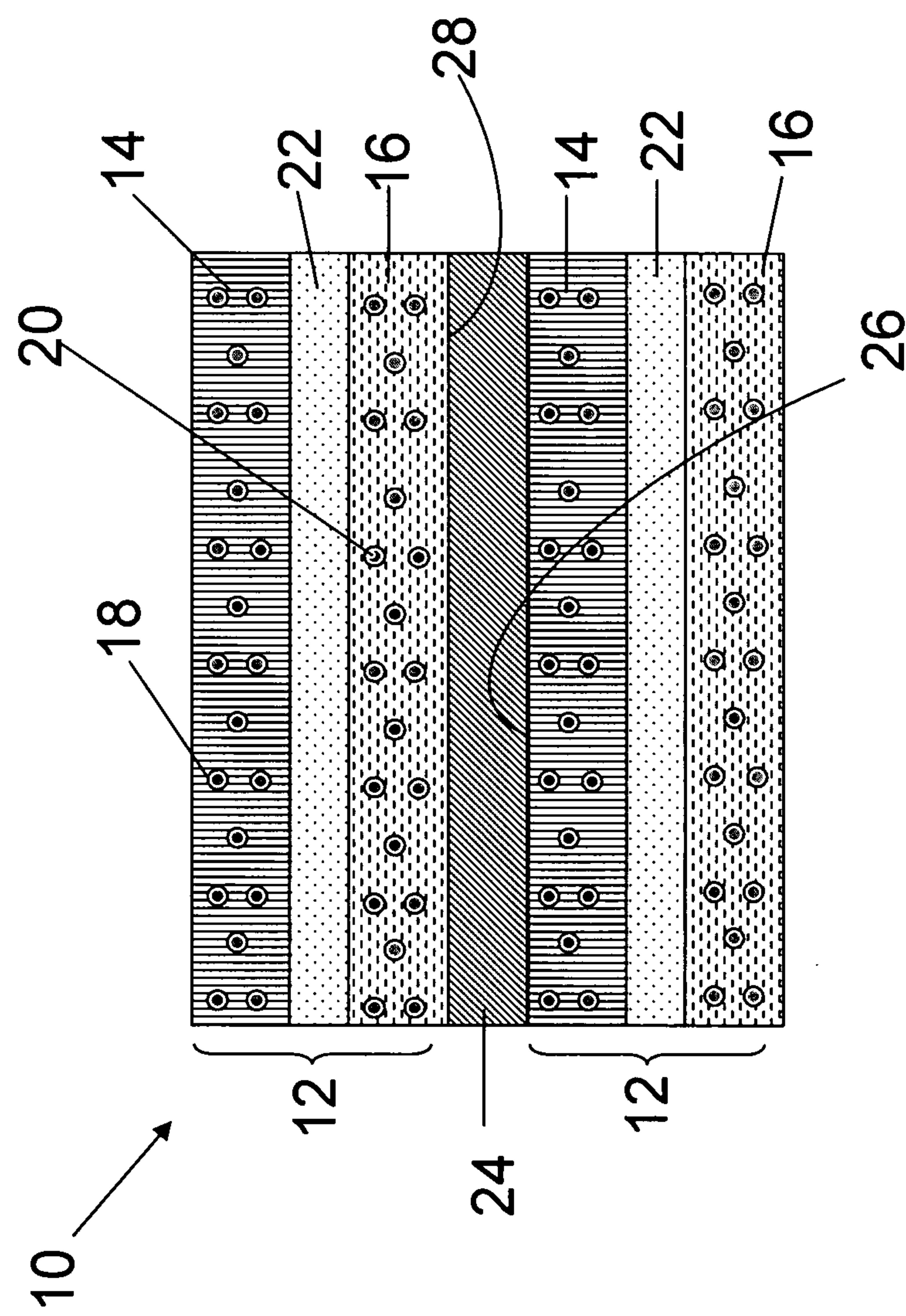


FIG. 1

2 / 3

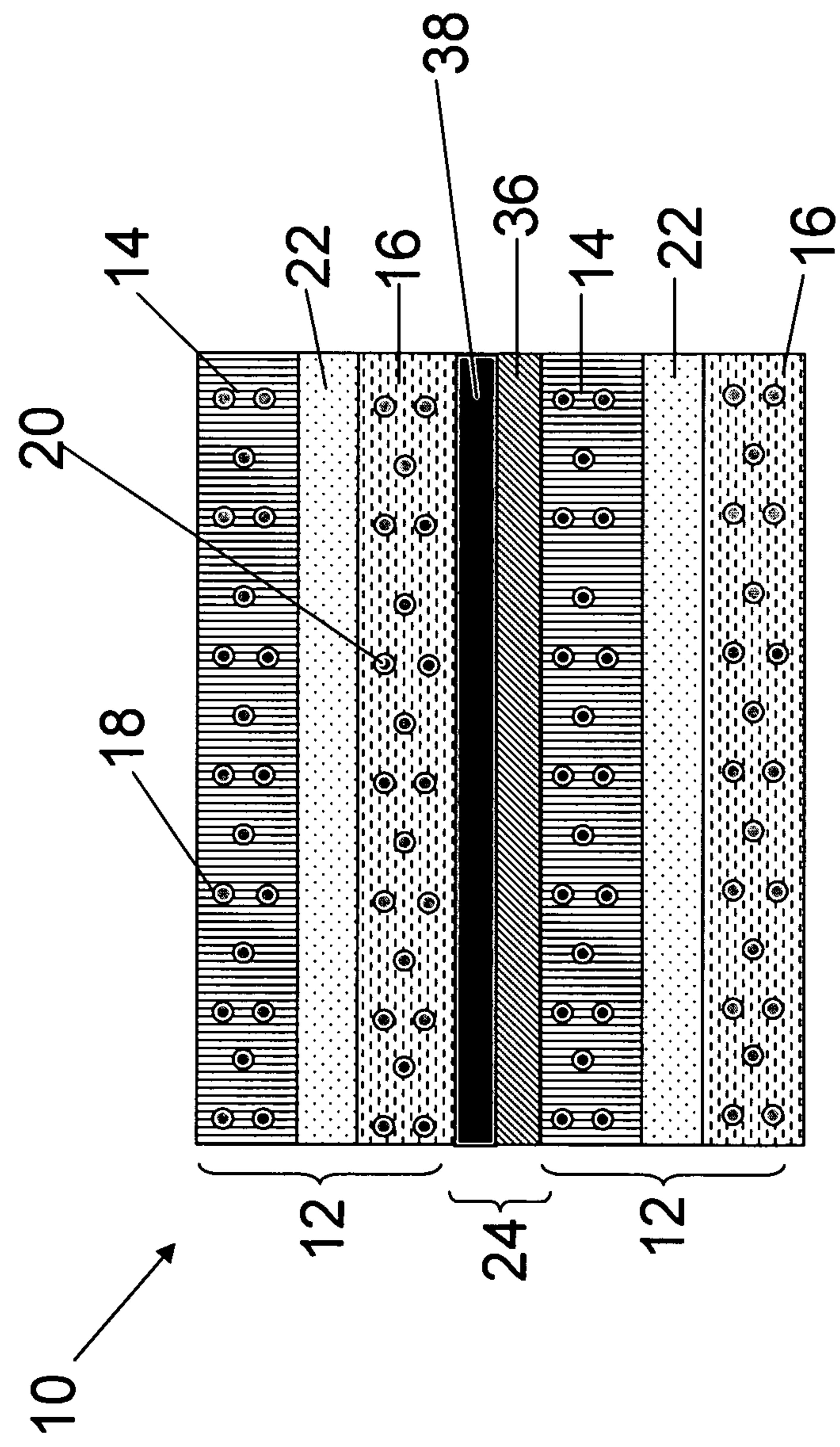


FIG. 2

3 / 3

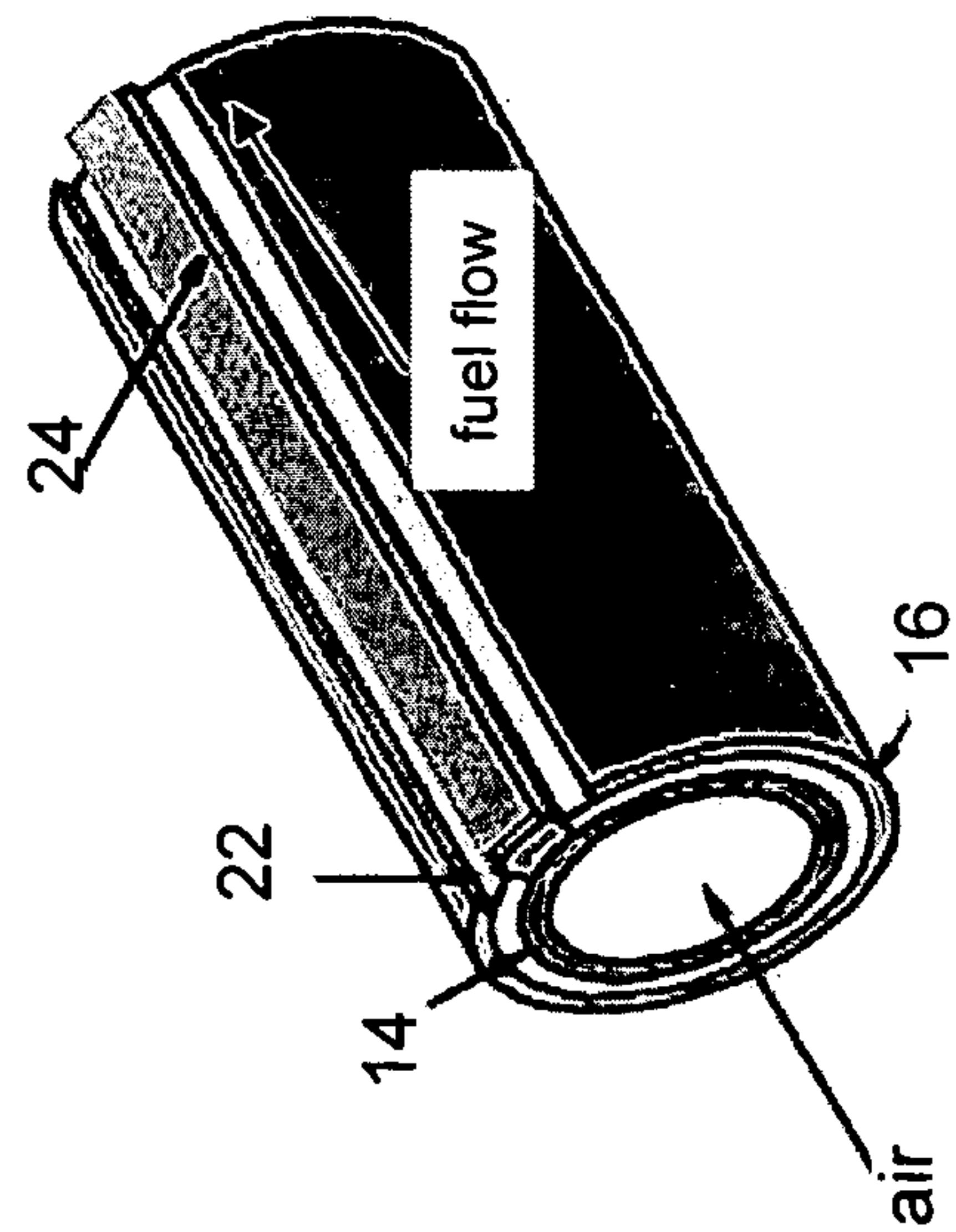


FIG. 4

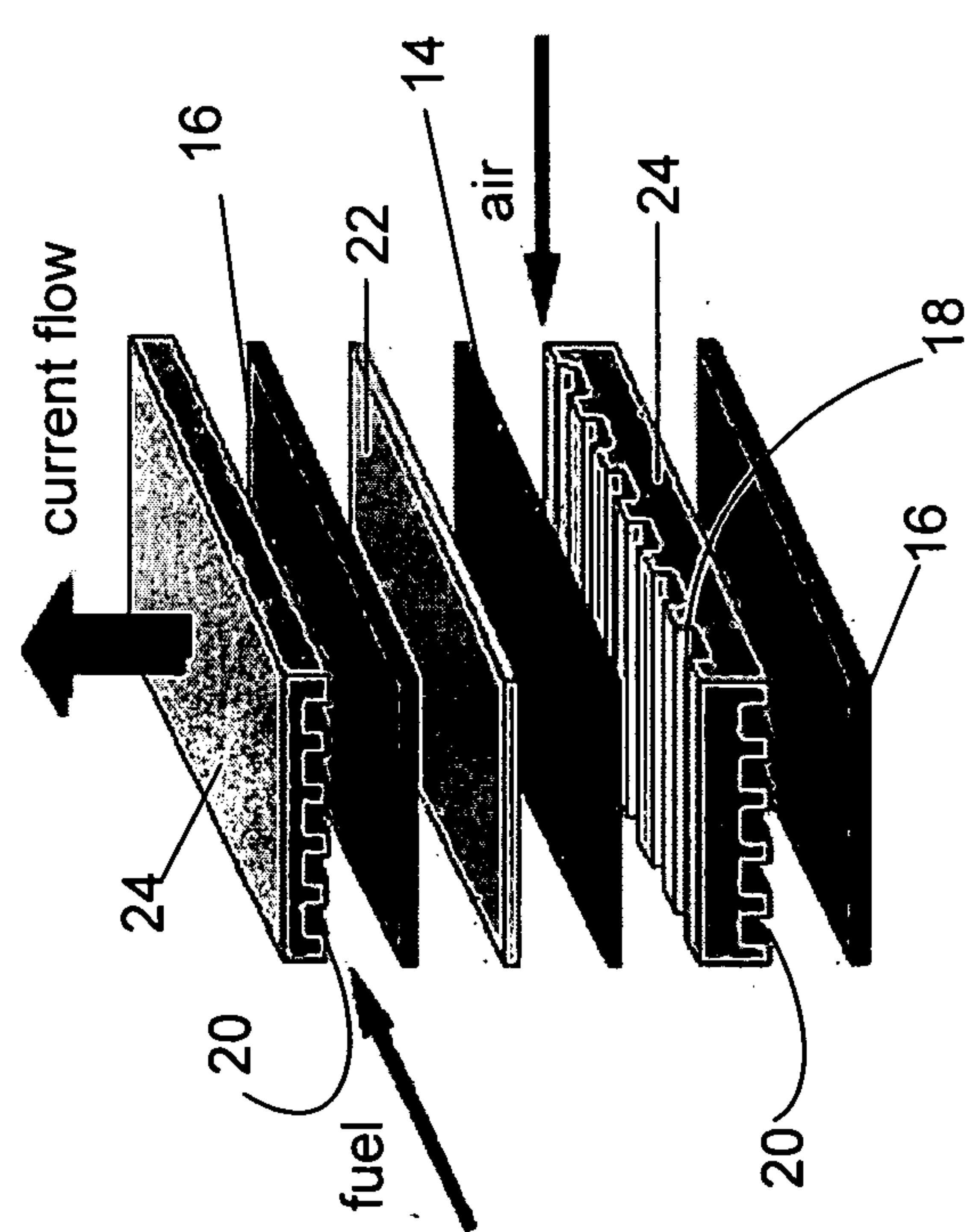


FIG. 3