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(54) **Title:** CORRUGATED SOLID OXIDE ELECTROACTIVE SUBSTRATE

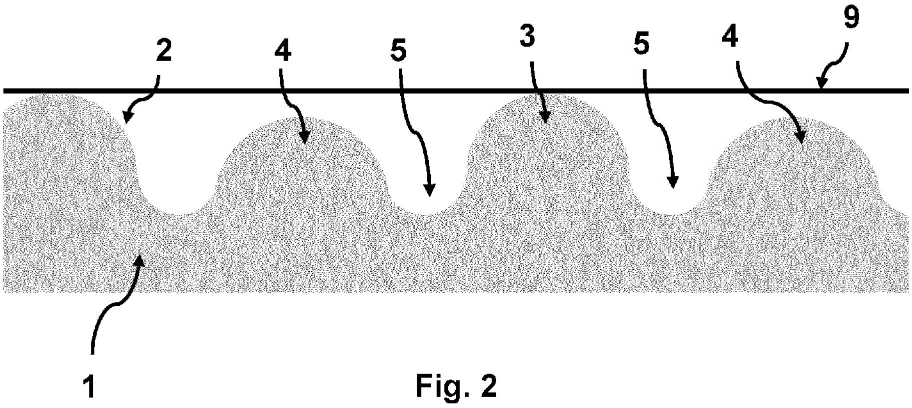

Fig. 2

(57) **Abstract:** Subject of the invention is a plate-shaped solid oxide electroactive substrate (1) having a corrugated surface (2) comprising at least two adjacent ridges (3) each having two ends, wherein the at least two adjacent ridges (3) each have exactly one tapered end section (4) such that each ridge (3) is inclined towards one of its two ends, wherein the tapered end sections (4) are formed at opposing ends of the at least two adjacent ridges (3).

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Corrugated Solid Oxide Electroactive Substrate

Field of the invention

The invention relates to corrugated solid oxide electroactive substrates, to electrochemical cells and cell stacks with such solid oxide electroactive substrates and to uses of such solid oxide electroactive substrates.

Background of the invention

In view of the climate ambitions, many states and companies made commitments to net zero CO_2 emissions. This however brings the need to produce significant amount of so-called green hydrogen to act as an energy vector in the transition from fossil fuels to renewable energy sources, or to help reduce the formed CO_2. Nowadays, the only means of production of green hydrogen is by electrolysis of water when using renewable electricity.

In principle, there are three types of electrolysis – alkaline electrolysis (AEL), proton exchange membrane electrolysis (PEMEL), and solid oxide electrolysis (SOEL). The most noticeable difference between those technologies is the temperature at which the electrolysis proceeds. While AEL and PEMEL are low temperature electrolyses limited to temperatures of <100°C, SOEL regularly uses special ceramic materials which become ion conductive at high temperatures, typically at temperatures of >450°C. For the most developed O^{2-} conductive ceramics, this temperature is even >650°C. These differences seriously impact the theoretical and real cell efficiency of electrolyses performed using such ceramic materials.

On the example of both H_2O and CO_2 electrolyses, it has been found that running the electrolysis at high temperature regularly decreases the electrical energy demands and increases the heat demand in the reaction. This is beneficent for the efficiency of the electrolysis and eventually also lowers operational expenditure (OPEX) of operating an electrolyser for the electrolysis.

In terms of efficiency, SOEL is typically better than AEL or PEMEL. The electrolysis process is namely accompanied by unwanted losses that transfer the delivered energy into excess heat. Thus, the low temperature electrolyses need cooling systems that vent the excess heat from the electrolyser. On the contrary, the SOEL can be designed and run at conditions of thermal equilibrium when the heat generated in the electrolyser compensates the heating requirements of the feed.

Unfortunately, the SOEL electrolysis is not as mature as the other two technologies. This translates especially into high costs needed per kW of installed power that usually exceed 2000 Eur/kW.

Another significant drawback of the SOEL technology is its very small cell size. Whereas AEL and PEMEL can have cells with surface areas of up to several square meters, SOEL is limited in cell size to maximum 30x30 cm or smaller. This is mainly caused by the fragility of the used ceramics which do typically not tolerate high temperature differences in the cell. Increasing the power of the SOEL electrolyser thus adopts rather scale-out strategy.

In an attempt to increase the cell surface area while keeping the cell compact, some researchers suggested to use 3D-printing to introduce modified electrolyte shapes, see Pesce et al., J. Mater. Chem. A 2020, 8, 16926–16932. The 3D-printing of the electrolyte is very demanding as it requires making a dense and very thin ceramic. Additionally, while the design of Pesce et al. increased the electrolyte area by 57%, further increases of the electrolyte area are desirable. Moreover, the electrolyte shape described by Pesce et al. does not form a channel plate, i.e., it does not allow for distributing gasses over an electrolytic cell in which the electrolyte is used.

Overall, there remains a general desire for an improved solid oxide electroactive substrate.

Problem underlying the invention

It is an object of the present invention to provide a solid oxide electroactive substrate which at least partially overcomes the drawbacks encountered in the art.

It is in particular an object of the present invention to provide a solid oxide electroactive substrate which allows for distributing gasses over an electrochemical cell in which the solid oxide electroactive substrate may be used.

It is furthermore an object of the present invention to provide a solid oxide electroactive substrate which has an increased surface area.

It is moreover an object of the present invention to provide a solid oxide electroactive substrate which allows for downsizing electrochemical cells without loss of performance.

It is additionally an object of the present invention to provide a solid oxide electroactive substrate which can lower the costs when used in an SOEL.

It is also an object of the present invention to provide an electrochemical cell, a cell stack and a use, respectively, which at least partially overcome the drawbacks encountered in the art.

Disclosure of the invention

Surprisingly, it has been found that the problem underlying the invention is overcome by a solid oxide electroactive substrate, an electrochemical cell, a cell stack and a use of a solid oxide electroactive substrate according to the claims. Further embodiments of the invention are outlined throughout the description.

Subject of the invention is a plate-shaped solid oxide electroactive substrate having a corrugated surface comprising at least two adjacent ridges each having two ends, wherein the at least two adjacent ridges each have exactly one tapered end section such that each ridge is inclined towards one of its two ends, wherein the tapered end sections are formed at opposing ends of the at least two adjacent ridges.

A solid oxide electroactive substrate according to the present invention provides interconnected channels in its plate-shaped structure. Accordingly, the inventive solid oxide electroactive substrate can allow for distributing gasses when it is used in an electrochemical cell. Additionally, the inventive solid oxide electroactive substrate can have an increased surface area, so that the size of the electrochemical cell may be reduced without loss of performance of the cell. Further, the costs of an SOEL can be advantageously lowered when the inventive solid oxide electroactive substrate is used in such an SOEL.

As used herein, plate-shaped means that the solid oxide electroactive substrate is a three-dimensional object, i.e., it has three dimensions. Two dimensions (length direction and width direction, respectively) are larger than the third, smallest dimension (or thickness). The two larger dimensions may be identical or different, and are preferably identical. When looked at from the top, that is in the direction of its thickness, the plate-shaped solid oxide electroactive substrate can for example have a squared shape, a rectangular shape, a circular shape or an oval shape.

As used herein, solid oxide means that the solid oxide electroactive substrate comprises at least one metal oxide and is in the solid state under standard conditions (temperature of 273.15 K; pressure of 0.1 MPa). The solid oxide electroactive substrate according to the present invention may in particular be configured to work as an electrode or as an electrolyte.

As used herein, corrugated means that a surface of the solid oxide electroactive substrate has a curvy or wavy cross-section, for example a sine curve cross-section or a sawtooth curve cross-section. The curvy or wavy cross-section also includes an angled cross-section, like a zig-zag cross-section. Corrugated preferably means that the corrugated surface of the solid oxide electroactive substrate has a periodic cross-section. Exemplary periodic cross-sections having a repeated basic section formed by one ridge and one groove are shown in Figs. 1 to 6.

As used herein, at least two adjacent ridges means that two maxima of the cross-section of the corrugated surface are adjacent to each other, i.e., there is no maximum between them. The two maxima extend in either the length direction or in the width direction of the solid oxide electroactive substrate, thereby forming a ridge. The ridge thus also extends in either the length direction or the width direction. Preferably, the two maxima are identical, i.e., the at least two adjacent ridges have the same height. Preferably, adjacent means substantially parallel so that the at least two adjacent ridges are at least two substantially parallel ridges. The solid oxide electroactive substrate according to the present invention may comprise more than two adjacent ridges, for example three or more, four or more, five or more, six or more, seven or more, eight or more, nine or more, or ten or more adjacent ridges.

As used herein, tapered end section means that the maximum of the cross-section of the ridge having the tapered end section is substantially identical over the entire length of the ridge except for one end section. In this end section, the maximum drops so that the ridge is inclined towards one of its two ends. The end section is preferably a section extending from one end of the ridge for at most 50%, more preferably for at most 40%, still more preferably for at most 30%, even more preferably for at most 20% and particularly preferably for at most 10% of the entire length of the ridge (extension of the tapered end section in percent of the entire extension of the ridge). Preferably, the extension of the tapered end section in percent is identical for the tapered end sections of both (or all) adjacent ridges.

As used herein, one tapered end section means that the ridge having the tapered end section has exactly one tapered end section. Each ridge has two end sections. Accordingly, only one of these two end sections is tapered (or inclined towards the end).

As used herein opposing ends refer to ends of the at least two adjacent ridges which are located on different sides of the plate-shaped solid oxide electroactive substrate. In other words, each of the ridges has a front end and a rear end (when looked at in the length direction or in the width direction, respectively). When one of the two adjacent ridges has a tapered end section at its front end, the other one of the two ridges has a tapered end section at its rear end.

When the solid oxide electroactive substrate comprises more than two ridges, preferably all ridges have tapered end sections which are formed at alternatingly opposing ends of the ridges. For example, the solid oxide electroactive substrate may comprise a first ridge which has a tapered end section at its front end, an adjacent second ridge which has a tapered end section at its rear end, a third ridge adjacent to the second ridge which has a tapered end section at its frond end, a fourth ridge adjacent to the third ridge which has a tapered end section at its rear end and optionally so forth as the case may be.

It is preferred for the solid oxide electroactive substrate according to the present invention that the at least two adjacent ridges define a groove between them. A groove is herein sometimes also referred to as a channel. The groove or channel is open to one side, namely into the direction of the two ridges, and is closed to the opposite side (has a closed groove bottom). In other words, it is preferred that the solid oxide electroactive substrate is not a mesh, is not a grid, and/or does not have any through-holes. Put differently, the solid oxide electroactive substrate is preferably gas-tight. With such a groove defined between the at least two adjacent ridges, the distribution of gasses in an electrochemical cell can be improved. At the same time, costs of an SOEL can be advantageously lowered when the inventive solid oxide electroactive substrate with such a groove is used in an SOEL. The solid oxide electroactive substrate according to the present invention may comprise more than one groove, for example two or more, three or more, four or more, five or more, six or more, seven or more, eight or more, nine or more, or ten or more adjacent and preferably substantially parallel grooves.

It is preferred for the solid oxide electroactive substrate according to the present invention that the tapered end sections have a height h_t which is lower by ≥ 30% than a maximum height h_r of the ridges, more preferably lower by ≥ 40% and still more preferably lower by ≥ 45%. It is preferred that all tapered end sections comprised by the inventive solid oxide electroactive substrate have the same height h_t. The tapered end sections are inclined towards one of the two ends of the ridge and hence drop from the maximum height h_r to a height h_t. Accordingly, the height h_t is the lowest height which the tapered end section has over the entire ridge. It is preferred that all ridges comprised by the inventive solid oxide electroactive substrate have the same maximum height h_r. In other words, it is preferred that all ridges comprised by the inventive solid oxide electroactive substrate have a constant height of the ridges over their length (over/along their extension) apart from the inclinations forming the tapered end sections. The maximum height h_r corresponds to the maximum height of the cross-section of the corrugation of the corrugated surface. Accordingly, the maximum height h_r regularly includes the thickness of the solid oxide electroactive substrate according to the present invention. With such a relationship between the height of the tapered end sections (h_t) and the height of the ridges (h_r), the distribution of gasses in an electrochemical cell can be further improved. At the same time, costs of an SOEL can be reduced when the inventive solid oxide electroactive substrate embodying such a height relationship is used in an SOEL.

It is preferred for the solid oxide electroactive substrate according to the present invention that the tapered end sections have a height h_t which is lower by not more than 90% than a maximum height h_r of the ridges, more preferably not lower by more than 80% and still more preferably not lower by more than ≥ 70%. Accordingly, it is preferred that $h_t = (0.3\text{-}0.9)h_r$, more preferred that $h_t = (0.4\text{-}0.8)h_r$ and still more preferred that $h_t = (0.45\text{-}0.7)h_r$.

With such a relationship between the height of the tapered end sections (h_t) and the height of the ridges (h_r), the distribution of gasses in an electrochemical cell can be further improved. At the same time, costs of an SOEL can be reduced when the inventive solid oxide electroactive substrate embodying such a height relationship is used in an SOEL.

It is preferred for the solid oxide electroactive substrate according to the present invention that the solid oxide electroactive substrate has a second corrugated surface opposite to the corrugated surface, i.e., opposite to the corrugated surface discussed hereinbefore which may also be referred to as first corrugated surface. With such a second corrugated surface, the inventive solid oxide electroactive substrate can allow for a further improved distribution of gasses in an electrochemical cell, namely on both sides of the solid oxide electroactive substrate. Additionally, the surface area is further increased, and this may allow to further downsize an electrochemical cell using the solid oxide electroactive substrate according to the present invention, especially to downsize such a cell without losing cell performance. This can also lead to a further reduction of costs for an SOEL performed in such a cell.

It is preferred for the solid oxide electroactive substrate according to the present invention that the second corrugated surface comprises at least two adjacent ridges each having two ends, wherein these at least two adjacent ridges each have exactly one tapered end section such that each ridge is inclined towards one of its two ends, wherein these tapered end sections are formed at opposing ends of those at least two adjacent ridges. The solid oxide electroactive substrate according to the present invention thereby provides interconnected channels in its plate-like structure on both of its sides and can hence also advantageously distribute gasses on both of its sides when used in an electrochemical cell. Additionally, the inventive solid oxide electroactive substrate can have a further increased surface area. Consequently, the size of the electrochemical cell may be further reduced without loss of cell performance. Further, the costs of an SOEL can be further lowered when such an inventive solid oxide electroactive substrate is used in an SOEL. The second corrugated surface may comprise more than two adjacent ridges, for example three or more, four or more, five or more, six or more, seven or more, eight or more, nine or more, or ten or more adjacent ridges. The height h_r of the ridge(s) comprised by the second corrugated surface and the height h_t of the tapered end section(s) of the ridge(s) comprised by the second corrugated surface is/are preferably of the same order and preferably have the same relationship as explained for the first corrugated surface. Likewise, the extension of the tapered end section of a ridge comprised by the second corrugated surface is preferably of the same order as explained for the first corrugated surface and are preferably identical for all tapered end sections of ridges comprised by the second corrugated surface.

When the solid oxide electroactive substrate according to the present invention has a second corrugated surface which comprises at least two adjacent ridges, the at least two adjacent ridges regularly define a groove between them. It is preferred that the second corrugated surface comprises at least two adjacent grooves which are connected by a connection formed by a tapered end section.

It is preferred for the solid oxide electroactive substrate according to the present invention that the first corrugated surface and optionally the second corrugated surface comprise(s) at least two adjacent and preferably substantially parallel grooves which are connected by a connection formed by a tapered end section. Thereby, the adjacent grooves form one extended channel which allows for an advantageous distribution of gasses over the corrugated surface of the inventive solid oxide electroactive substrate.

It is preferred that the solid oxide electroactive substrate according to the present invention is a corrugated plate, more preferably such that ridges comprised by the first corrugated surface form grooves of the second corrugated surface and that ridges comprised by the second corrugated surface form grooves of the first corrugated surface. This can allow for a particularly effective production of the solid oxide electroactive substrate, especially by 3D-printing, and for savings in terms of required solid oxide material. Preferably, the corrugated plate has a periodic corrugation.

It is preferred for the solid oxide electroactive substrate according to the present invention that the solid oxide electroactive substrate is obtained by 3D-printing (is 3D-printed). The term 3D-printing (or three-dimensional printing) is used herein as generally understood in the art and refers to a production of the plate-shaped solid oxide electroactive substrate according to a digital model or template. It is preferred that in the 3D-printing, solid oxide material, and potentially other components of the solid oxide electroactive substrate, is deposited, joined or solidified under computer control, with such material being added together, typically layer by layer. The 3D-printing allows for a very precise control of the production of the inventive solid oxide electroactive substrate. The solid oxide electroactive substrate can thereby be made in the form of a very thin, but sufficiently rigid and stable plate for use in an SOEL. Additionally, the dimensions of the plate-shaped solid oxide electroactive substrate can be advantageously controlled. An overview of available 3D-printing techniques is for example provided in Z. Chen et al., Journal of the European Ceramic Society, 39 (2019) 661–687. A preferred 3D-printing can especially be selected from a slurry-based 3D-printing, a powder-based 3D-printing and a bulk solid-based 3D-printing. Here, a preferred slurry-based 3D-printing can be selected from stereolithography (SL), digital light processing (DLP), two-photon polymerisation (TPP), inkjet printing (IJP) and direct ink writing (DIW), a preferred powder-based 3D-printing can be selected from three-dimensional printing (3DP), selective laser sintering (SLS) and selective laser melting (SLM), and a preferred bulk solid-based 3D-printing can be selected from laminated object manufacturing (LOM) and fused deposition modelling (FDM).

Further, when the solid oxide electroactive substrate according to the present invention is obtained by 3D-printing, the 3D-printed solid oxide material can be conductive for electrons and ions such as H^+ and/or O^{2-} ions forming ion and electron conductive material. With 3D-printing, the solid oxide electroactive substrate according to the present invention can thereby be provided with inherent electron and ion conductive properties so that no separate deposition of electrode layers may be required anymore for actual use like in for example a membrane reactor or bi-functional membrane reactor.

The 3D printed solid oxide electroactive substrate may also have a function of a support layer preferably with porosity allowing for easy diffusion of gases and may, at least partially and preferably as a whole, function as an electrode. The deposition, and hence presence, of other contacting, electrolyte, barrier, or electrolyte layers is thus also encompassed.

Further, when the solid oxide electroactive substrate according to the present invention functions as an electrolyte, for example in an SOEL, one or two electrode layers typically need to be deposited thereon. A solid oxide electroactive substrate according to the present invention having at least one and preferably two electrode layers (comprising an electron-conductive material like metal, graphite, etc.) deposited thereon is thus also encompassed.

However, when the solid oxide electroactive substrate according to the present invention is obtained by 3D-printing, the 3D-printed solid oxide material can be conductive for electrons and ions such as H^+ and/or O^{2-} ions forming ion and electron conductive material. With 3D-printing, the solid oxide electroactive substrate according to the present invention can thereby be provided with inherent electron and ion conductive properties so that no separate deposition of electrode layers may be required anymore for actual use like in for example a membrane reactor or bi-functional membrane reactor.

It is preferred for the solid oxide electroactive substrate according to the present invention that the ridges comprised by the first corrugated surface and/or the ridges comprised by the second corrugated surface bear protrusions. With such protrusions, the surface area can be further increased. Accordingly, the size of an electrochemical cell comprising such a solid oxide electroactive substrate with protrusions on ridges can be reduced without loss of cell performance. Accordingly, the costs of an SOEL running in such a cell can be lowered.

It is particularly preferred that the protrusions have a conical shape or a frustoconical shape. With the protrusions having a conical shape or a frustoconical shape, the manufacturing precision can be enhanced, especially when the solid oxide electroactive substrate is obtained by 3D-printing. Further, an effective and regular conduction of H^+ and/or O^{2-} ions through the solid oxide electroactive substrate may be possible. Additionally, when the solid oxide electroactive substrate is electroconductive, an effective and regular distribution of electrical charges may be possible.

It is preferred for the solid oxide electroactive substrate according to the present invention that the ridges of the first and/or the second corrugated surface have a curved cross-section defined by an inner circle, wherein the protrusions have a frustoconical shape with a diameter of its circular base of $\leq 1/8$ of the diameter of the inner circle and/or with a diameter of its top base of $\leq 1/12$ of the diameter of the inner circle and/or with a height of $\leq 1/8$ of the diameter of the inner circle. With such dimensions the surface area of the solid oxide electroactive substrate can be particularly increased while keeping or even further reducing the size of the solid oxide electroactive substrate and hence also of a cell comprising the solid oxide electroactive substrate.

It is preferred for the solid oxide electroactive substrate according to the present invention that the corrugated surface has a projected surface area and an effective surface area, wherein the effective surface area is larger than the projected surface area by ≥100%, more preferably larger by ≥120% and still more preferably larger by ≥140%. The projected surface area is the area obtained from a top view of the plate-shaped solid oxide electroactive substrate. In other words, the projected surface area is obtained from a twodimensional area measurement of the three-dimensional solid oxide electroactive substrate by projecting its plate-shape in thickness direction onto a theoretical plane. The effective surface area corresponds to the actual surface of the corrugated surface, i.e., taking the corrugations with optional protrusions thereon into account. With the effective surface area being larger than the projected surface area by ≥100%, the effective surface area is at least double (=100%) or more (>100%) the projected surface area. The surface area is thereby significantly increased, allowing for a downsizing of an electrochemical cell comprising the solid oxide electroactive substrate, and allowing for a reduction of costs for an SOEL performed in such an electrochemical cell.

Generally, the solid oxide electroactive substrate has two surfaces which can be named a top surface and a bottom surface. The perpendicular distance between the top surface and the bottom surface of the solid oxide electroactive substrate defines the thickness of the solid oxide electroactive substrate. It is preferred for the solid oxide electroactive substrate according to the present invention that the solid oxide electroactive substrate has a thickness (also abbreviated “d” herein) of 5 to 1000 µm, more preferred of 50 to 500 µm and still more preferred of 150 to 350 µm. On the one hand, a thickness of ≥ 5 µm can prevent that the solid oxide electroactive substrate easily cracks and can provide stability and rigidity. On the other hand, a thickness of ≤1000 µm can avoid an unnecessary increase of a resistance in the cell, as well as of the cell thickness in which the solid oxide electroactive substrate is used and can avoid an unnecessarily excessive use of material. These effects are respectively more pronounced for a thickness of 50 to 500 µm and still more pronounced for a thickness of 150 to 350 µm.

When the solid oxide electroactive substrate according to the present invention has a preferred thickness of 150 to 350 µm, 3D-printing thereof can provide an improved thickness homogeneity which can avoid the formation of detrimental hot spots in operation caused by the Joule effect when high current densities are passing through thinner regions. An x-y-z resolution as achievable by 3D-printing is regularly not achievable by any other production method.

It may alternatively be preferred for the solid oxide electroactive substrate according to the present invention that the solid oxide electroactive substrate has a thickness (also abbreviated “d” herein) of 120 to 600 µm, more preferred of 150 to 500 µm and still more preferred of 150 to 350 µm. The thickness extends from the top surface to the bottom surface of the solid oxide electroactive substrate according to the present invention.

Accordingly, the thickness is the perpendicular distance between the top surface and the bottom surface. In other words, the thickness is typically the thickness of the plate forming a plate-shaped solid oxide electroactive substrate. On the one hand, a thickness of ≥ 120 µm can prevent that the solid oxide electroactive substrate easily cracks and can provide stability and rigidity. On the other hand, a thickness of ≤600 µm can avoid an unnecessary increase of a resistance in the cell, as well as of the cell thickness in which the solid oxide electroactive substrate is used and can avoid an unnecessarily excessive use of material. These effects are respectively more pronounced for a thickness of 150 to 500 µm and still more pronounced for a thickness of 150 to 350 µm. Due to these effects, loss of cell performance can be avoided when downsizing an electrochemical cell in which the inventive solid oxide electroactive substrate is used.

When the solid oxide electroactive substrate according to the present invention has a preferred thickness of 120 to 600 µm, 3D-printing thereof can provide an improved thickness homogeneity which can avoid the formation of detrimental hot spots in operation caused by the Joule effect when high current densities are passing through thinner regions. An x-y-z resolution as achievable by 3D-printing is regularly not achievable by any other production method. Due to these effects, loss of cell performance can be avoided when downsizing an electrochemical cell in which the inventive solid oxide electroactive substrate is used.

It is preferred for the solid oxide electroactive substrate according to the present invention that the solid oxide electroactive substrate is made from a self-supporting electrode material on which a thin electrolyte may be deposited.

It is preferred for the solid oxide electroactive substrate according to the present invention that the solid oxide electroactive substrate comprises yttria-stabilised zirconia, more preferably an 8 mol% yttria-stabilized zirconia (8YSZ). Such an yttria-stabilised zirconia has an increased O^{2-} conductivity and can improve the effectiveness of an SOEL performed using the inventive solid oxide electroactive substrate.

Subject of the invention is also an electrochemical cell comprising a solid oxide electroactive substrate according to the present invention. The preferred embodiments of the solid oxide electroactive substrate described herein including the claims are likewise preferred for this inventive electrochemical cell in an analogous manner. In the inventive electrochemical cell gasses can be advantageously distributed. Additionally, the size of the inventive electrochemical cell can be reduced while its electrochemical performance is maintained stable. Further, the costs for running an electrolysis in the inventive electrochemical cell can be lower.

The electrochemical cell typically comprises two electrodes and an electrolyte sandwiched between the two electrodes. The electrochemical cell is preferably an electrolytic cell comprising a fuel electrode and an oxygen electrode, or the electrochemical cell preferably operates as a bi-functional membrane reactor. The electrochemical cell is regularly a finished product having the corrugated substrate and added layers so that a smallest fully functional unit is formed which typicvally includes an anode and a cathode as electrodes, an electrolyte and potentially also barrier layers, contacting layers, functional layers and other layers which may have standalone function or can be just a part of the basic cathode, anode, or electrolyte configuration.

The electrochemical cell is preferably configured to run in electrolysis mode, i.e., the electrochemical cell is preferably an electrolyser which consumes electric energy to electrolyse a substance. The electrolysed substance is preferably water (H_2O) or carbon dioxide (CO_2), more preferably H_2O. It is preferred that the electrochemical cell is electrically connected to a renewable energy source, wherein the renewable energy is more preferably selected from energy generated from sunlight, wind, rain, tides, waves and/or geothermal heat, more preferably generated from sunlight or wind. The electrochemical cell can alternatively be configured to run in reverse mode in which case the electrochemical cell is preferably a battery or a fuel cell which outputs electric energy.

It is also contemplated that the electrochemical cell is preferably a dual-side or bi-functional reactor or a protonic cell which is conductive for hydrogen ions. When two reactions are simultaneously carried out in one reactor, especially in a catalytic membrane reactor, it may be termed as a bi-functional reactor, especially a bi-functional membrane reactor.

Subject of the invention is also a cell stack comprising at least two electrochemical cells according to the present invention which are adjacent to each other and which are separated by a separation plate. The preferred embodiments of the solid oxide electroactive substrate described herein including the claims are likewise preferred for this inventive cell stack in an analogous manner. The inventive cell stack can allow for distributing gasses in an electrochemical cell in which the substance is electrolysed.

Additionally, the inventive cell stack employs at least two solid oxide electroactive substrates each having an increased surface area. Accordingly, the size of the cell stack for the electrolysis of the substance can be reduced, while maintaining the electrochemical performance of the cell stack on the same level. Further, with the inventive cell stack the costs for electrolysing the substance can be advantageously lowered.

A cell stack comprising at least two electrochemical cells according to the present invention (and hence two solid oxide electroactive substrates according to the present invention) can furthermore especially be a sub-unit of an electrochemical device according to the present invention, i.e., of an electrochemical device comprising a solid oxide electroactive substrate according to the present invention. A cell stack can be separately commercialized and can for example be a substitute for a used and potentially degraded older cell stack. In such a scenario, there is not always a need to replace the entire electrochemical device, but a substitution of the cell stack may be sufficient. In other words, an electroactive substrate according to the present invention, a cell stack according to the present invention and an electrochemical device according to the present invention can in particular be interrelated products. The separate discussions of such interrelated products provided herein shall help to understand the technical field and the overall context of the present invention.

Subject of the invention is also a use of a solid oxide electroactive substrate according to the present invention, or of an electrochemical cell comprising a solid oxide electroactive substrate according to the present invention, or of a cell stack according to the present invention, in an electrolysis of a substance. The electrolysed substance is preferably H_2O and/or CO_2, more preferably H_2O. The preferred embodiments of the solid oxide electroactive substrate described herein including the claims are likewise preferred for this inventive use in an analogous manner. The inventive use can allow for distributing gasses in an electrochemical cell in which the substance is electrolysed. Additionally, the inventive use employs a solid oxide electroactive substrate having an increased surface area. Accordingly, the size of the electrochemical cell for the electrolysis of the substance can be reduced, while maintaining the electrochemical performance of the electrochemical cell on the same level. Further, with the inventive use the costs for electrolysing the substance can be advantageously lowered. Any use of an inventive solid oxide electroactive substrate, or of an electrochemical cell comprising an inventive solid oxide electroactive substrate, or of an inventive cell stack in an electrolysis of a substance may also be considered as a corresponding method of using such an inventive solid oxide electroactive substrate, such an electrochemical cell comprising an inventive solid oxide electroactive substrate, or such an inventive cell stack in an electrolysis of a substance, i.e. for electrolyzing that substance, wherein the substance is preferably H_2O and/or CO_2, and more preferably H_2O.

An electrolysis of H_2O, or electrochemical water splitting, uses electricity to decompose water into oxygen and hydrogen gas. The electrolysis is regularly performed in an electrolytic cell which comprises two electrodes, namely an anode and a cathode, between which an electric field is applied. Between the anode and the cathode, an electrolyte according to the present invention is preferably sandwiched. When operating the electrochemical cell, O^{2-} ions are removed from the water at the cathode to form hydrogen gas, and on the other hand electrons are removed from the O^{2-} ions at the anode to form oxygen gas. Accordingly, H_2 is produced at the cathode according to the following equation:

$$H_2O(g) \rightarrow H_2(g) + O^{2-}$$

At the same time, O_2 is produced at the anode according to the following equation:

$$2O^{2-} \rightarrow O_2(g) + 4e^-$$

The overall cell reaction is then as follows:

$$2\ H_2O(g) \rightarrow 2\ H_2(g) + O_2(g)$$

The efficiency of the electrolysis may be advantageously increased through the use of electrocatalysts. At high temperatures, however, all components of the electrolytic cell including the electrolyte are typically ceramic in nature.

An electrolysis of CO_2, or electrochemical reduction of carbon dioxide, uses electricity to convert CO_2 to more reduced chemical species. The electrolysis is regularly performed in an electrolytic cell which comprises two electrodes, namely an anode and a cathode, between which an electric field is applied. Between the anode and the cathode, an electrolyte according to the present invention is preferably sandwiched. When operating the electrochemical cell, O^{2-} ions recombine at the anode to form oxygen (O_2). Alternatively, the O^{2-} ions might participate in a reaction with a gas, e.g., methane and/or other hydrocarbons, CO, COS, H_2, H_2S, sulphur, N_2O, NO, NH_3 or a mixture of such gases fed to the anode. Gas preferably fed to the anode also includes exhaust gas with the remnants of unburnt compounds and/or soot, natural gas, sour gas, fuel gas, biogas, syngas etc.

At the cathode, CO_2 is reduced to CO or carbon according to the following reactions:

$CO_2 + 2e^- \rightarrow CO + O^{2-}$

$CO_2 + 4e^- \rightarrow C + 2\,O^{2-}$

Brief description of the drawings

Fig. 1 schematically shows a corrugated solid oxide electroactive substrate with one corrugated surface without tapered end sections of ridges.

Fig. 2 schematically shows a corrugated solid oxide electroactive substrate with one corrugated surface including tapered end sections of ridges.

Fig. 3 schematically shows another corrugated solid oxide electroactive substrate with one corrugated surface including tapered end sections of ridges.

Fig. 4 schematically shows a corrugated solid oxide electroactive substrate with two opposite corrugated surfaces without tapered end sections of ridges.

Fig. 5 schematically shows a corrugated solid oxide electroactive substrate with two opposite corrugated surfaces including tapered end sections of ridges.

Fig. 6 schematically shows a superposition of a corrugated solid oxide electroactive substrate according to Fig. 4 and a corrugated solid oxide electroactive substrate according to Fig. 5.

Fig. 7 schematically shows a top view of a plate-shaped solid oxide electroactive substrate having two opposite corrugated surfaces.

Fig. 8 schematically shows a corrugated solid oxide electroactive substrate having ridges which bear protrusions.

Fig. 9 shows an angled side view of a corrugated solid oxide electroactive substrate with one corrugated surface including tapered end sections of ridges.

Fig. 10 shows a corrugated solid oxide electroactive substrate having a corrugated surface including tapered end sections at opposing ends of adjacent ridges.

Fig. 11 shows the corrugated solid oxide electroactive substrate of Fig. 10 with level curves.

Fig. 12 shows another corrugated solid oxide electroactive substrate having a corrugated surface including tapered end sections at opposing ends of adjacent ridges.

Fig. 13 shows the corrugated solid oxide electroactive substrate of Fig. 12 with level curves.

Exemplary embodiments

Embodiment 1

In Fig. 1, a plate-shaped corrugated solid oxide electroactive substrate 1 having one corrugated surface is shown. The solid oxide electroactive substrate 1 has a corrugated surface 2 which is formed from adjacent ridges 3 and adjacent grooves 5. Also shown is a separation plate 9 which is usually electrically conductive. As seen from Fig. 1, the adjacent ridges 3 abut the separation plate 9 and thus have the same height. According to the present invention, the height of the ridges 3 is however not constant over the entire length of the ridges. Rather, at one end of the ridges 3 a tapered end section 4 is formed as illustrated in Fig. 2. Note that Fig. 2 shows a cross-section of one end, or edge, of the plate-shaped substrate 1 at which every second ridge 3 has, or is formed with, a tapered end section 4. At the not shown opposite end or edge of the plate-shaped substrate 1 those ridges of Fig. 2 which abut the separation plate 9 have, or are formed with, a tapered end section 4. Hence, the tapered end sections 4 are formed at opposing ends of every two adjacent ridges 3.

Embodiment 2

In Fig. 3, another plate-shaped corrugated solid oxide electroactive substrate 1 having one corrugated surface is shown. Again, the solid oxide electroactive substrate 1 has a corrugated surface 2 which is formed from adjacent ridges 3 and adjacent grooves 5, wherein the ridges 3 abut a separation plate 9. In this embodiment, tapered end sections 4 are formed at the end of every second ridge 3, but these tapered end sections 4 have a further reduced height. This is illustrated by showing the entire height of the ridges 3 indicated by “h_r”, and the height of the tapered end sections indicated by “h_t”. Even though Fig. 3 is basically schematic, it is seen that the height h_t of the tapered end sections 4 is lower by ca. 30% or more than the maximum height h_r of the ridges 3.

Embodiment 3

In Fig. 4, a plate-shaped corrugated solid oxide electroactive substrate 1 as in principle known from Pesce et al. is shown which has two corrugated surfaces on opposite sides of the plate-shaped substrate 1. More specifically, the solid oxide electroactive substrate 1 has a corrugated surface 2 which is formed from adjacent ridges 3 and adjacent grooves 5. The shown structure also has a second corrugated surface 6 with ridges 7 and grooves 8. In Pesce et al. all ridges are formed in a uniform manner without any tapered or inclined sections. In order to form stacks, the gas distribution would have to be assured via a dedicated channel plate. Otherwise, the groves 5 would not be interconnected so that the grooves would form a number of separated channels. According to embodiment 3 the cells are stacked by using two separation plates 9 from the top and the bottom. Fig. 4 thus represents a cut through the middle part of the plate-shaped corrugated solid oxide electroactive substrate 1.

In Fig. 5, another corrugated solid oxide electroactive substrate 1 according to the present invention is shown which also has two corrugated surfaces. As seen from Fig. 5, the solid oxide electroactive substrate 2 according to the present invention comprises a ridge 3 which has a tapered end section 4. In the depicted embodiment, the corrugation is decreased to about one half, thereby interconnecting two adjacent grooves 5 so that one long channel results. At the opposite side, the corrugation is likewise decreased to about one half, thereby interconnecting two adjacent grooves 8. A decrease to one half means that the height of the tapered end section h_t is half the height of the ridge h_r, or $h_t = 0.5h_r$, as shown in Fig. 5.

In Fig 6, the corrugated structure of Fig. 4 is shown together with a superimposed structure of an embodiment of a corrugated solid oxide electroactive substrate with tapered end sections of Fig. 5.

Fig. 7 shows a top view of the present embodiment of a corrugated solid oxide electroactive substrate 1 having two opposite corrugated surfaces. The thick black line connects the highest points (maxima) of the corrugated surface 2. These highest points form ridges 3. The lowest points (minima) are connected by grey lines. These lowest points form grooves 5. By decreasing the corrugation at the end of the ridges 3 and the grooves 5, it is possible to interconnect the two neighbouring grooves 5. In other words, by forming the tapered end sections 4 on the first corrugated surface 2, connections 10 between the grooves 5 are formed. This allows gas to pass through. Accordingly, this structure effectively creates a path for reactants and products which is very similar to the standard path in a channel plate. Hence, while achieving the same advantages as achieved with currently used channel plates, the corrugated solid oxide electroactive substrate according to the present invention further brings about advantages associated with the significantly increased effective surface area, namely the possibility to downsize the electrochemical cell for an SOEL, and to reduce the costs for an SOEL.

In the embodiment shown in Fig. 7, the solid oxide electroactive substrate 1 has a second corrugated surface 6 opposite to the surface 2 which surface 6 has ridges 7 and grooves 8. The ridges 7 basically correspond to the grooves 5 of the first surface 2. The tapered end sections 4 of the first surface 2 also form tapered end sections 4 in the second corrugated surface 6. By decreasing the corrugation at the end of the ridges 7 and the grooves 8 of the second surface, it is possible to interconnect the two neighbouring grooves 8. In other words, by forming the tapered end sections 4, connections 11 between the grooves 8 on the second corrugated surface 6 are formed. This allows also the gas on the opposite side to pass through. With two similarly corrugated surfaces 2 and 6, the effects of the solid oxide electroactive substrate 1 described above are basically doubled.

Embodiment 4

The lateral and height resolution of the 3D-printing method (stereolithography, SLA) is typically about 25 µm. This allows to create small protrusions 12 on the surface of the solid oxide electroactive substrate 1 as shown in Fig. 8. The protrusions 12 further increase the effective surface area of the solid oxide electroactive substrate 1. The higher surface area is beneficent as it further decreases the specific area resistance and allows to reach higher yields. In the shown embodiment, the protrusions 12 are conical frustum structures, i.e., the protrusions 12 have a frustoconical shape. More specifically, the conical frusta have a diameter of the circular base of 75 µm, a diameter of the top base of 25 µm, and a height of 100 µm. Compared to the structures reported in Pesce et al., who report an increase of the effective surface area of 57%, the frustoconical protrusions 12 lead to a further increase of the effective surface area by another 90%, making overall 147% rise in effective surface area in comparison to the flat design (i.e., in comparison to the projected surface area).

In the example of the cell used in Pesce et. al, the original projected surface area of 2 cm^2 would be increased to 4.95 cm^2 by this surface roughening. At the same time, the surface remains accessible everywhere to deposit thin electrodes and for future access of reactant gasses and escape of product gasses, for example in an SOEL. In case of a thick self-supporting electrolyte shown in Fig. 8, the electrodes are deposited by painting the surface with/dipping the cell in commercially available electrode solutions. The resulting electrode layer is thus very thin around 25-50 µm.

Illustration of basic geometry

Fig. 9 illustrates the formation of tapered end sections on ridges of a plate-shaped solid oxide electroactive substrate 1’ which has a corrugated surface 2’. A ridge 3’ is shown which has a constant height over its length apart from the end section shown in front, which thereby forms a tapered end section 4’. The ridge has two ends with only the front end being shown in Fig. 9. At this front end, the tapered end section 4’ is formed such that the ridge 3’ is inclined towards one of its two ends, namely its front end. Fig. 9 shows inclinations of adjacent ridges at the same end, but only for illustration purposes. In a plate-shaped solid oxide electroactive substrate according to the present invention the tapered end sections are formed at opposing ends of the at least two adjacent ridges, as for example shown in the top view of Fig. 7. Fig. 9 is therefore not meant to present an embodiment of the invention, but shows and further explains the geometry of ridges and tapered end sections, respectively, of electroactive substrates of the present invention.

Embodiment 5

Figs. 10 and 11 show an embodiment of a solid oxide electroactive substrate 1 with a corrugated surface 2 according to the present invention. Between two adjacent ridges 3 a groove 5 is formed. The two adjacent ridges 3 each have two ends, one pointing forward and one pointing backwards (observer’s perspective). In other words, each ridge 3 has one front end and one rear end. At opposing ends, i.e., at one forward pointing end and at one backwards pointing end, there are inclinations such that two opposing tapered end sections 4 are formed. In other words, one of the two adjacent ridges 3 has a tapered end section 4 at its front end, while the other one of the two ridges 3 has a tapered end section 4 at its rear end. Each ridge 3 has only one such tapered end section 4, that is each ridge 3 has two ends and has exactly one tapered end section 4. In Fig. 11 level curves (or contour lines) are added to further illustrate a constant height of the ridges 3 over their length, or extension, apart from the inclinations forming the tapered end sections 4.

Embodiment 6

Figs. 12 and 13 show another embodiment of a solid oxide electroactive substrate 1 with a corrugated surface 2 according to the present invention. Accordingly, a groove 5 is present between two adjacent ridges 3 each of which has two ends and consequently also two end sections. Of these two end sections, only one is tapered such that the ridge is inclined towards exactly one of its two ends, thereby forming exactly one tapered end section 4 one each ridge 3. In line with the present invention, the respective tapered end section 4 is formed, or arranged, at opposing ends of the two adjacent ridges 3. In Fig. 12, from the observer's perspective one tapered end section 4 is formed at a forward pointing end (or front end) of a first ridge 3, while the tapered end section 4 of a second and directly neighbouring (or adjacent) ridge 3 is formed at a backwards pointing end (or rear end) thereof. Fig. 13 further shows level curves which show that ridges 3 have a constant height of the over their length which only drops towards one of its two end sections, thereby creating a tapered end section 4. Figs. 12 and 13 additionally show a flow path 13 which illustrates how fluid travels along the grooves 5 (channels) and passes from one groove 5 via a tapered end section 4 into a neighbouring groove 5. In this way, interconnected channels or grooves 5 are provided in the plate-shaped solid oxide electroactive substrate 1. This interconnection allows for an advantageous distribution of fluid, in particular of gasses, especially when the solid oxide electroactive substrate 1 is used in an electrochemical cell.

List of reference signs

1: solid oxide electroactive substrate
1': illustrative solid oxide electroactive substrate
2: corrugated surface
2': illustrative corrugated surface
3: ridge
3': illustrative ridge
4: tapered end section
4': illustrative tapered end section
5: groove
6: second corrugated surface
7: ridge
8: groove
9: separation plate

10:	connection
11:	connection at the opposite side
12:	protrusion
13:	flow path
d:	thickness of substrate
h_t:	height of tapered end section
h_r:	height of ridge

<u>Further disclosure</u>

The present invention further provides the following items:

1. A plate-shaped solid oxide electroactive substrate (1) having a corrugated surface (2) comprising at least two adjacent ridges (3), wherein the at least two adjacent ridges (3) each have one tapered end section (4), wherein the tapered end sections (4) are formed at opposing ends of the at least two adjacent ridges (3).

2. The solid oxide electroactive substrate (1) according to item 1, wherein the at least two adjacent ridges (3) define a groove (5) between them.

3. The solid oxide electroactive substrate (1) according to item 1 or 2, wherein the tapered end sections (4) have a height h_t which is lower by ≥ 30% than a maximum height h_r of the ridges (3).

4. The solid oxide electroactive substrate (1) according to any of the preceding items, wherein the solid oxide electroactive substrate (1) has a second corrugated surface (6) opposite to the corrugated surface (2).

5. The solid oxide electroactive substrate (1) according to item 4, wherein the second corrugated surface (6) comprises at least two adjacent ridges (7), wherein the at least two adjacent ridges (7) each have one tapered end section (4), wherein the tapered end sections (4) are formed at opposing ends of the at least two adjacent ridges (7).

6. The solid oxide electroactive substrate (1) according to any of the preceding items, wherein the solid oxide electroactive substrate (1) is obtained by 3D-printing.

7. The solid oxide electroactive substrate (1) according to any of the preceding items, wherein the ridges (3) and/or the ridges (7) bear protrusions (12).

8. The solid oxide electroactive substrate (1) according to item 7, wherein the protrusions (12) have a conical shape or a frustoconical shape.

9. The solid oxide electroactive substrate (1) according to item 8, wherein the ridges (3) and/or the ridges (7) have a curved cross-section defined by an inner circle, and wherein the protrusions (12) have a frustoconical shape with a diameter of its circular base of ≤ 1/8 of the diameter of the inner circle and/or with a diameter of its top base of ≤ 1/12 of the diameter of the inner circle and/or with a height of ≤ 1/8 of the diameter of the inner circle.

10. The solid oxide electroactive substrate (1) according to any of the preceding items, wherein the corrugated surface (2) has a projected surface area and an effective surface area, wherein the effective surface area is larger than the projected surface area by ≥100%.

11. The solid oxide electroactive substrate (1) according to any of the preceding items, wherein the solid oxide electroactive substrate (1) has a thickness of 5 to 1000 µm.

12. The solid oxide electroactive substrate (1) according to any of the preceding items, wherein the solid oxide electroactive substrate (1) comprises yttria-stabilised zirconia.

13. An electrochemical cell comprising a solid oxide electroactive substrate (1) according to any of the preceding items, wherein the electrochemical cell is preferably an electrolytic cell comprising a fuel electrode and an oxygen electrode or wherein the electrochemical cell preferably operates as a bi-functional membrane reactor.

14. A cell stack comprising at least two solid oxide electroactive substrates (1) according to any of items 1 to 12, which are adjacent to each other and which are separated by a separation plate (9).

15. Use of a solid oxide electroactive substrate (1) according to any of items 1 to 12, or of an electrochemical cell according to item 13, or of a cell stack according to item 14, in an electrolysis of a substance, preferably in an electrolysis of H_2O and/or CO_2.

Claims

1. A plate-shaped solid oxide electroactive substrate (1) having a corrugated surface (2) comprising at least two adjacent ridges (3) each having two ends, wherein the at least two adjacent ridges (3) each have exactly one tapered end section (4) such that each ridge (3) is inclined towards one of its two ends, wherein the tapered end sections (4) are formed at opposing ends of the at least two adjacent ridges (3).

2. The solid oxide electroactive substrate (1) according to claim 1, wherein the at least two adjacent ridges (3) define a groove (5) between them.

3. The solid oxide electroactive substrate (1) according to claim 1 or 2, wherein the tapered end sections (4) have a height h_t which is lower by ≥ 30% than a maximum height h_r of the ridges (3).

4. The solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the solid oxide electroactive substrate (1) has a second corrugated surface (6) opposite to the corrugated surface (2).

5. The solid oxide electroactive substrate (1) according to claim 4, wherein the second corrugated surface (6) comprises at least two adjacent ridges (7) each having two ends, wherein the at least two adjacent ridges (7) each have exactly one tapered end section (4) such that each ridge (7) is inclined towards one of its two ends, wherein the tapered end sections (4) are formed at opposing ends of the at least two adjacent ridges (7).

6. The solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the solid oxide electroactive substrate (1) is obtained by 3D-printing.

7. The solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the ridges (3) and/or the ridges (7) bear protrusions (12).

8. The solid oxide electroactive substrate (1) according to claim 7, wherein the protrusions (12) have a conical shape or a frustoconical shape.

9. The solid oxide electroactive substrate (1) according to claim 8, wherein the ridges (3) and/or the ridges (7) have a curved cross-section defined by an inner circle, and wherein the protrusions (12) have a frustoconical shape with a diameter of its circular base of ≤ 1/8 of the diameter of the inner circle and/or with a diameter of its top base of ≤ 1/12 of the diameter of the inner circle and/or with a height of ≤ 1/8 of the diameter of the inner circle.

10. The solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the corrugated surface (2) has a projected surface area and an effective surface area, wherein the effective surface area is larger than the projected surface area by ≥100%.

11. The solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the solid oxide electroactive substrate (1) has a thickness of 5 to 1000 µm.

12. The solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the solid oxide electroactive substrate (1) comprises yttria-stabilised zirconia.

13. An electrochemical cell comprising a solid oxide electroactive substrate (1) according to any of the preceding claims, wherein the electrochemical cell is preferably an electrolytic cell comprising a fuel electrode and an oxygen electrode or wherein the electrochemical cell preferably operates as a bi-functional membrane reactor.

14. A cell stack comprising at least two electrochemical cells according to claim 13, which are adjacent to each other and which are separated by a separation plate (9).

15. Use of a solid oxide electroactive substrate (1) according to any of claims 1 to 12, or of an electrochemical cell according to claim 13, or of a cell stack according to claim 14, in an electrolysis of a substance, preferably in an electrolysis of H_2O and/or CO_2.

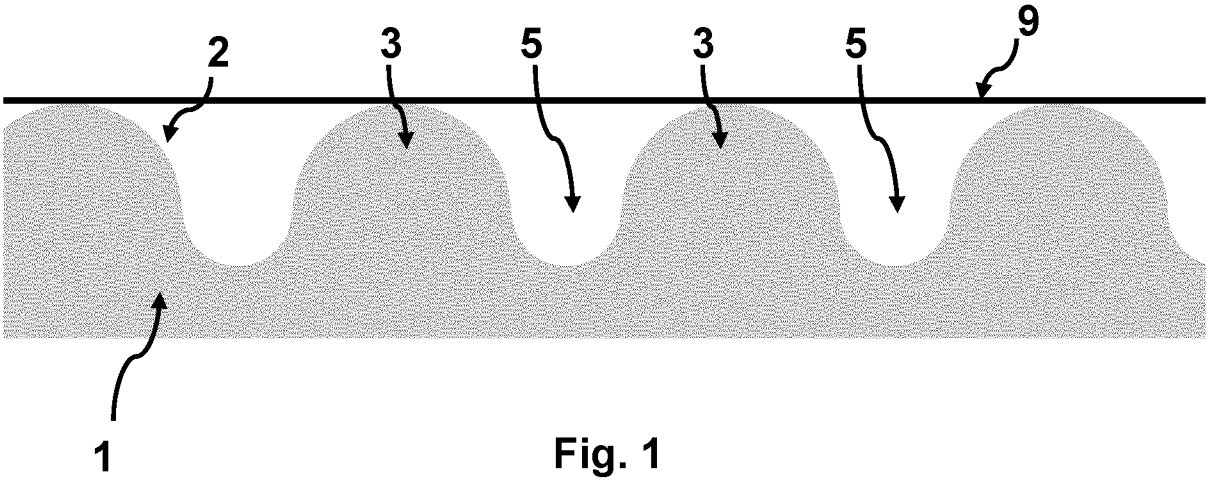

Fig. 1

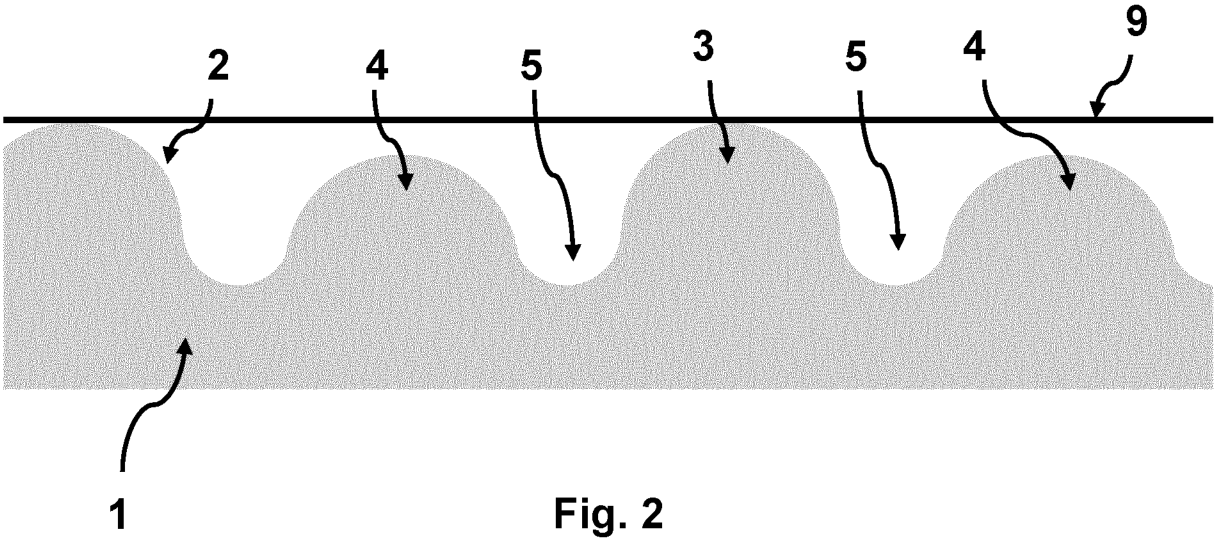

Fig. 2

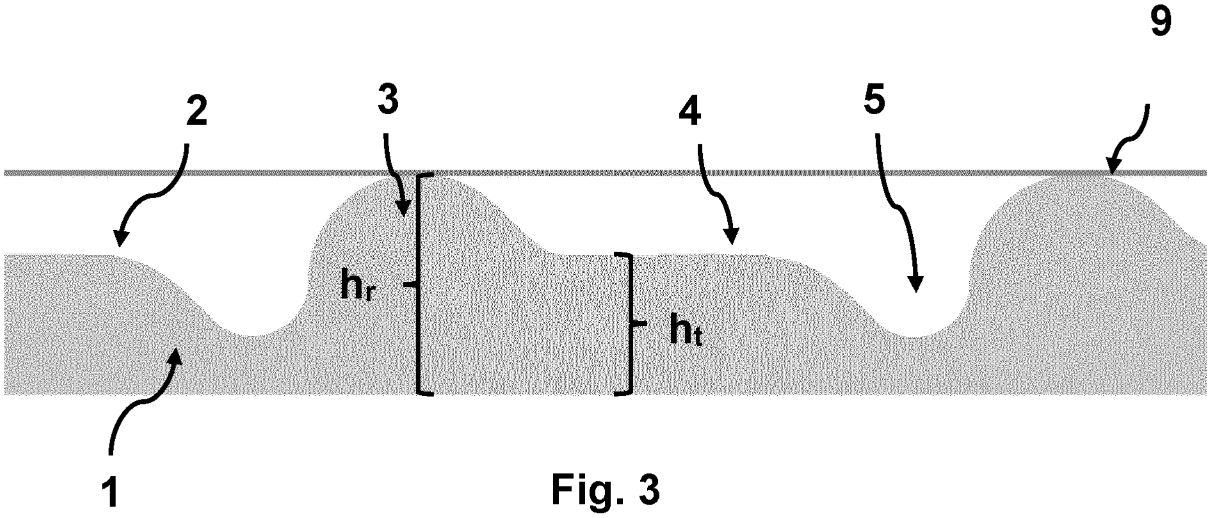

Fig. 3

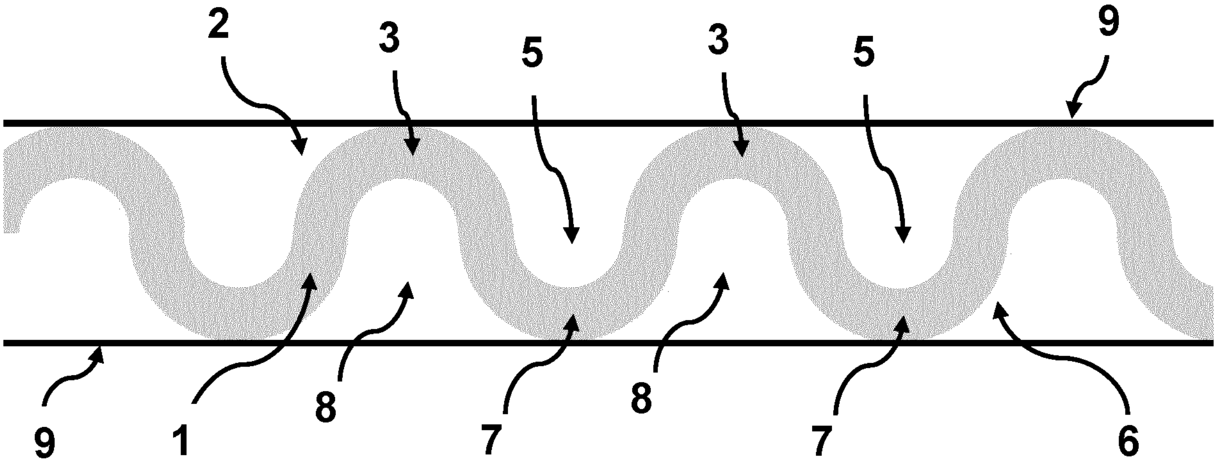

Fig. 4

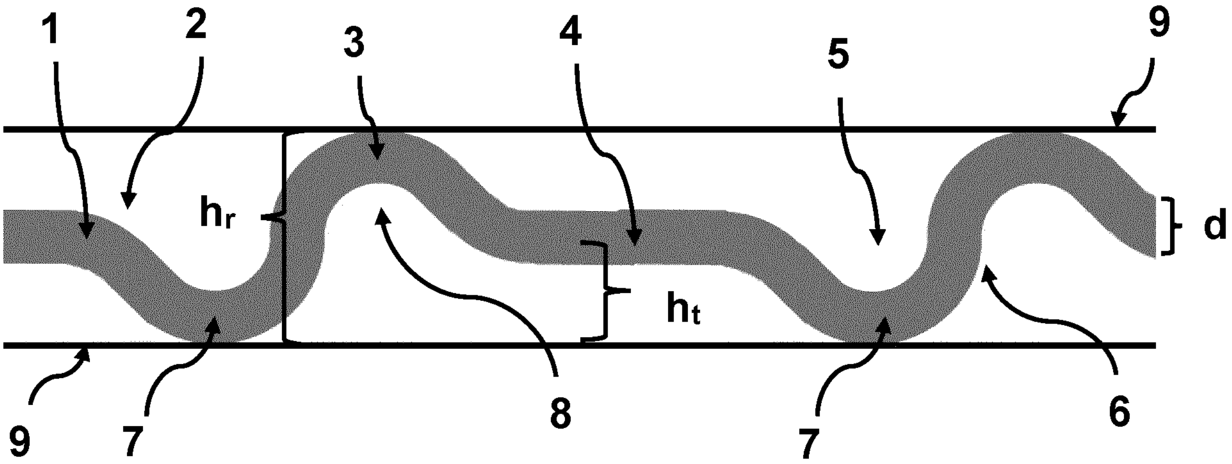

Fig. 5

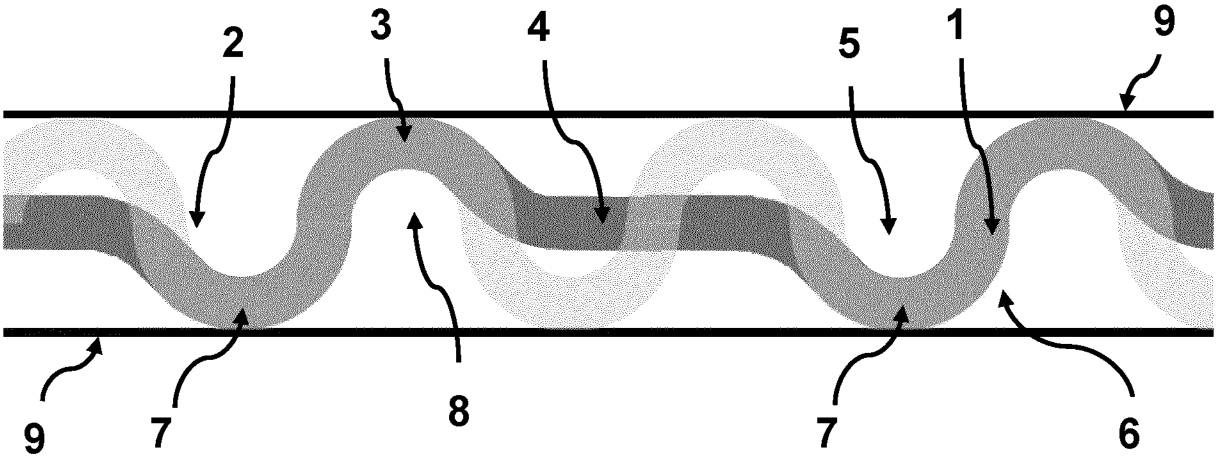

Fig. 6

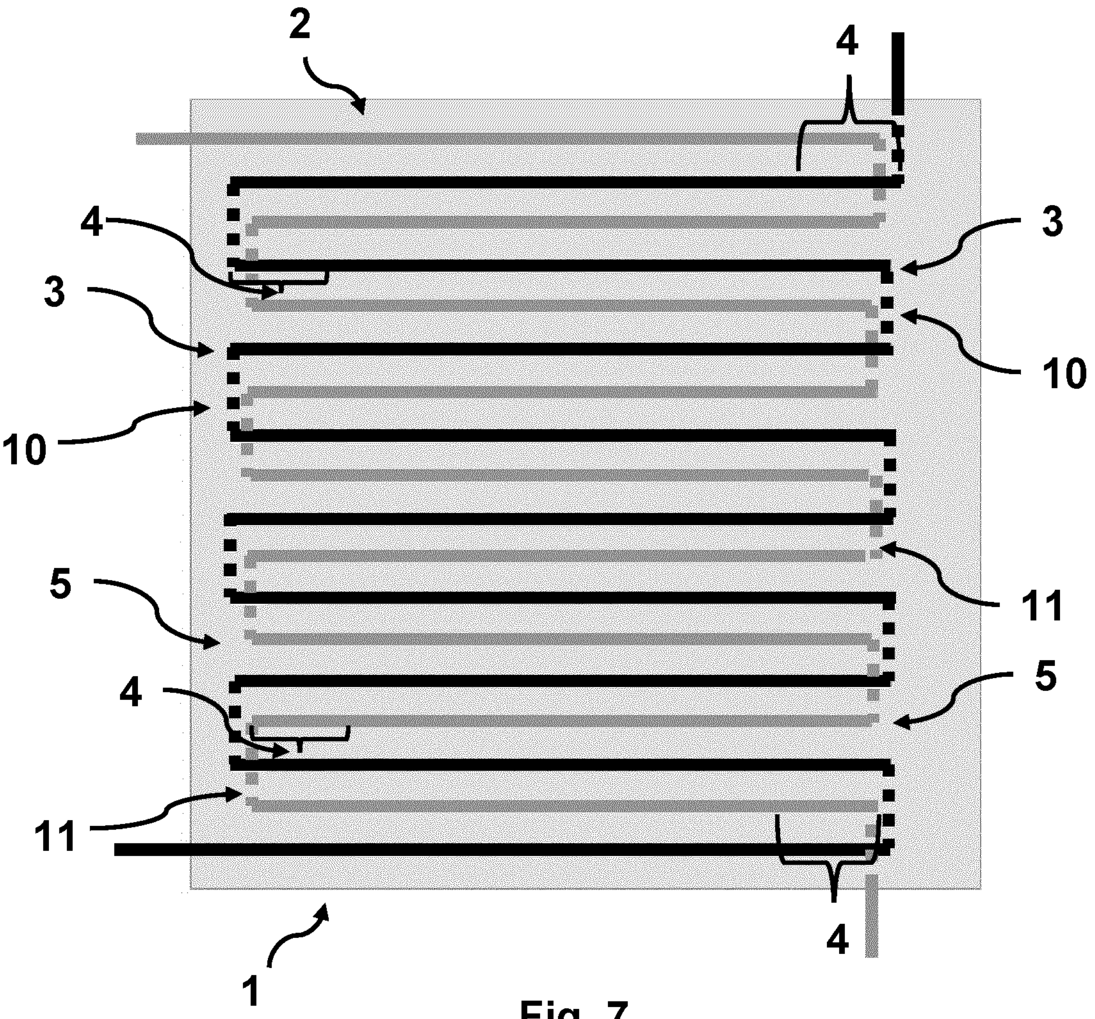

Fig. 7

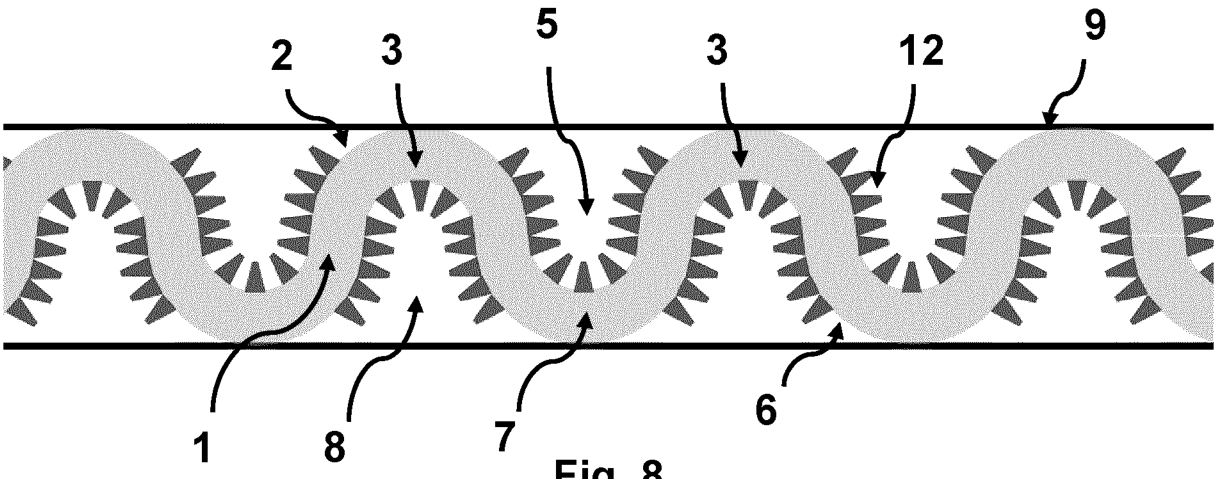

Fig. 8

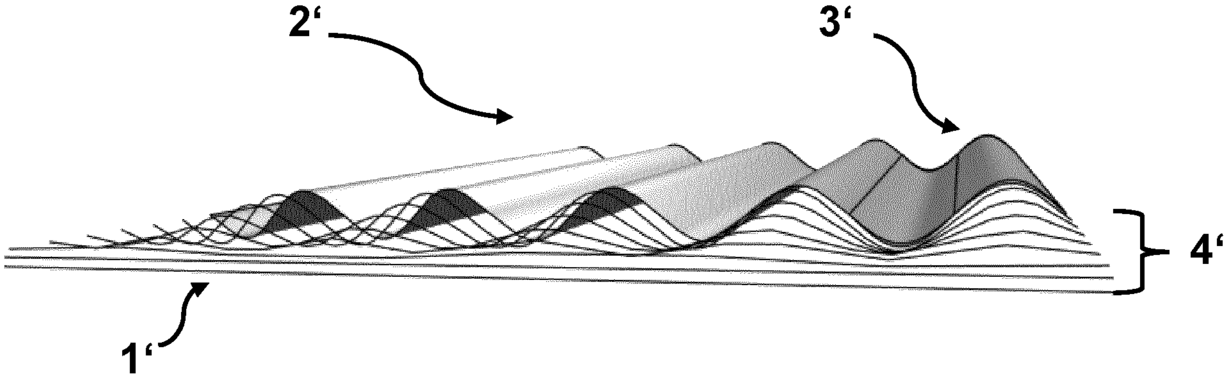

Fig. 9

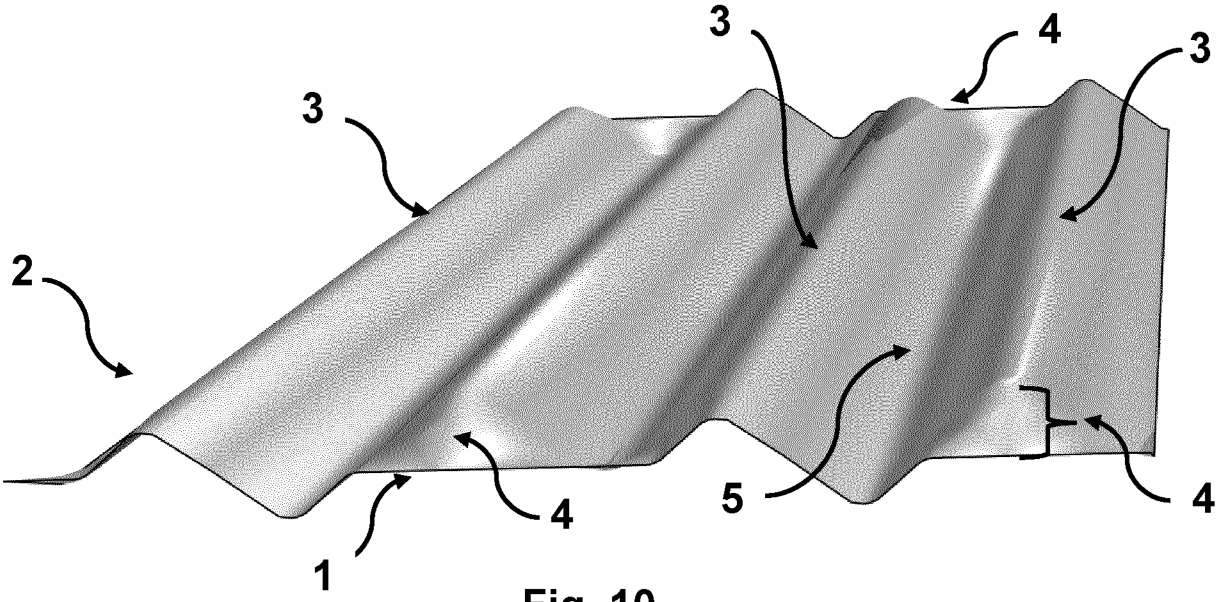

Fig. 10

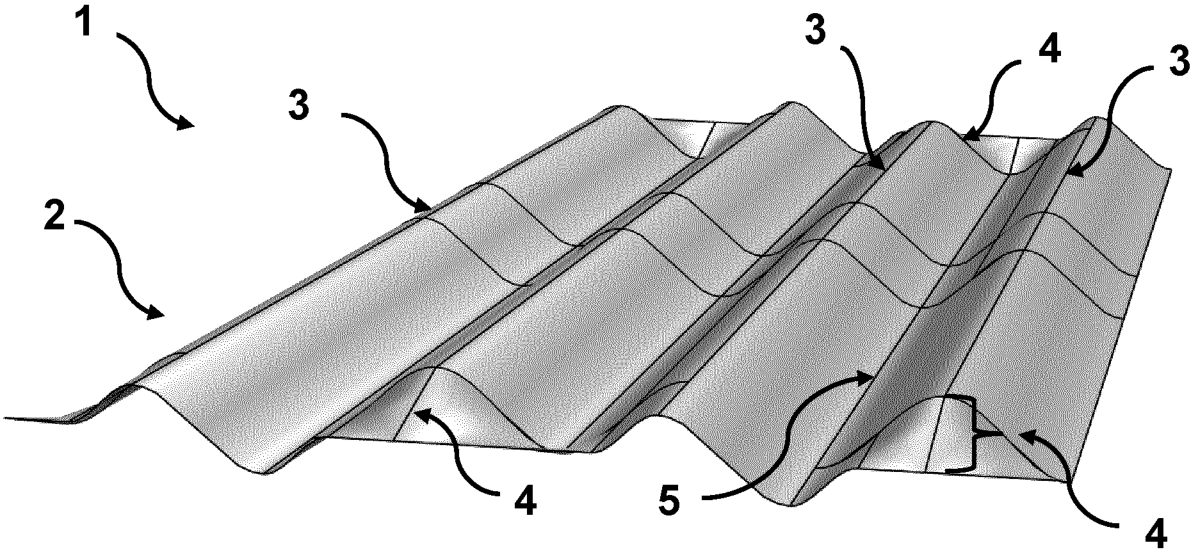

Fig. 11

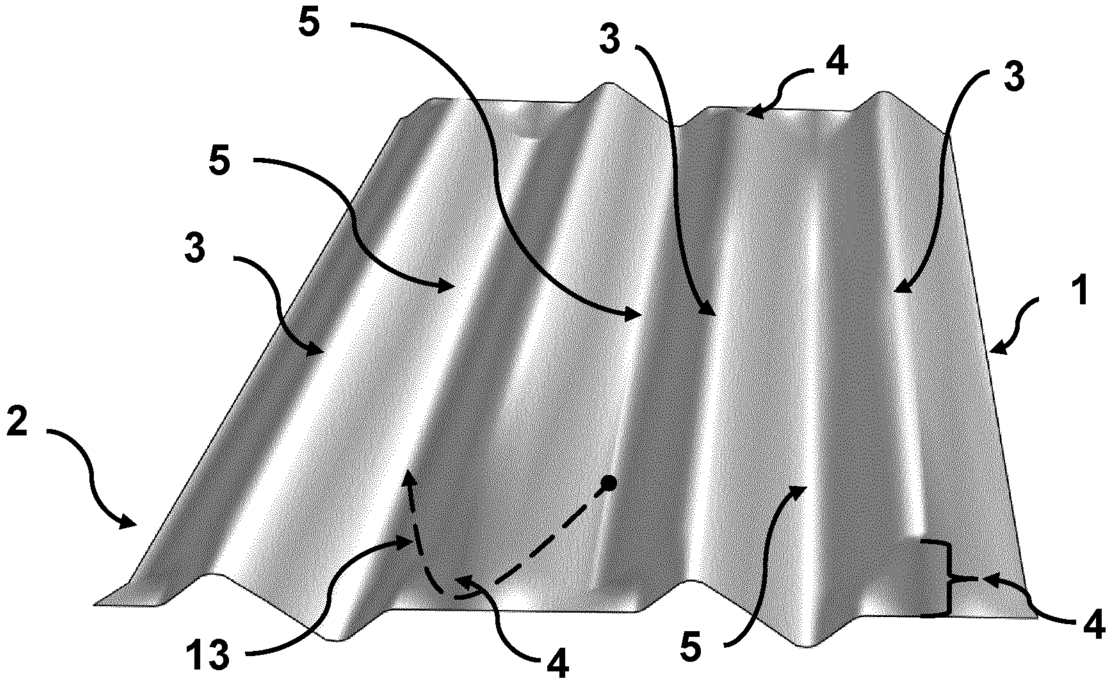

Fig. 12

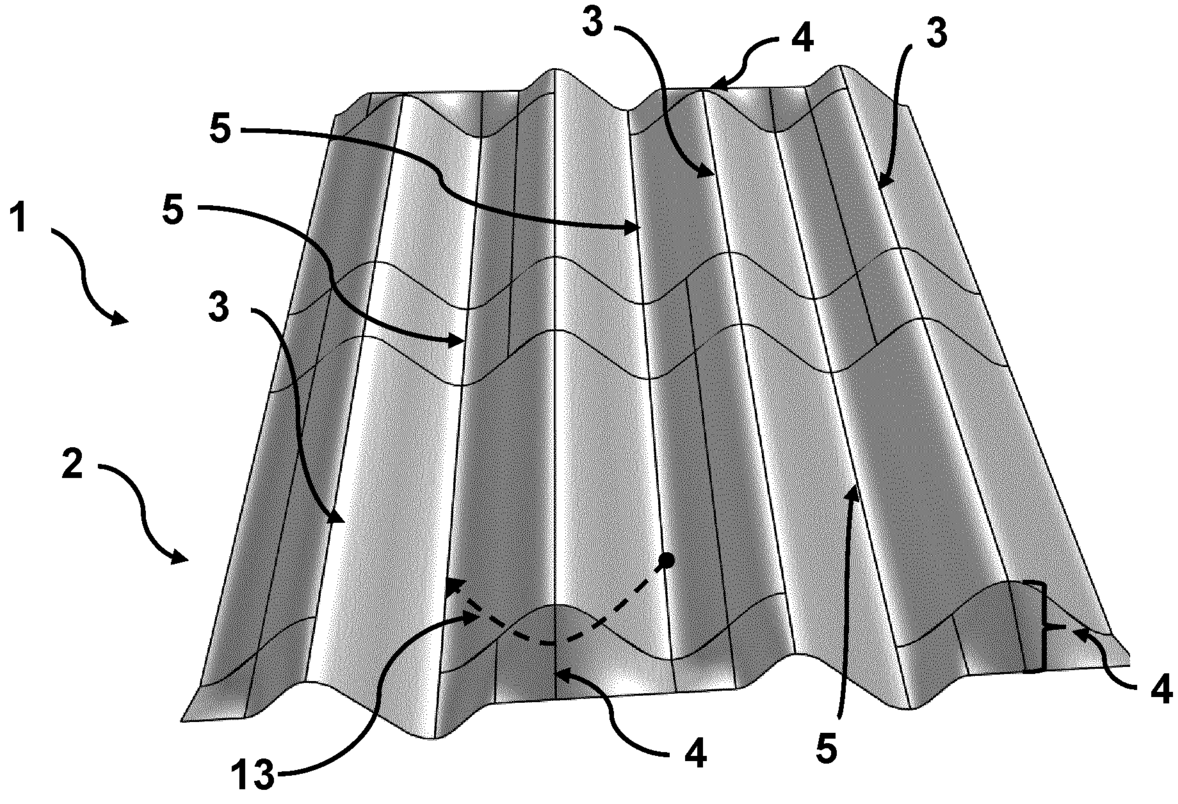

Fig. 13

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2024/051213

A. CLASSIFICATION OF SUBJECT MATTER
INV. C25B1/04 C25B1/042 C25B3/26 C25B13/02 C25B13/07
H01M8/1253
ADD.

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)
C25B H01M

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 practicable, search terms used)

EPO-Internal, WPI Data, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 2007/030106 A1 (CORNING INC [US]; KETCHAM THOMAS D [US] ET AL.) 15 March 2007 (2007-03-15) abstract; figures 1-5, 9-13B paragraph [0030] - paragraph [0053]; claims 1-14 -----	1-15
A	EP 1 892 787 A2 (NGK INSULATORS LTD [JP]) 27 February 2008 (2008-02-27) abstract; claims 1-7; figures 2, 17 paragraph [0019] - paragraph [0023] paragraph [0044] -----	1-15
A	EP 2 413 408 A1 (NIPPON CATALYTIC CHEM IND [JP]) 1 February 2012 (2012-02-01) abstract; claims 1-13; figures 1, 2, 10 paragraph [0025] - paragraph [0027] ----- -/--	1-15

☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search 12 April 2024	Date of mailing of the international search report 22/04/2024
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