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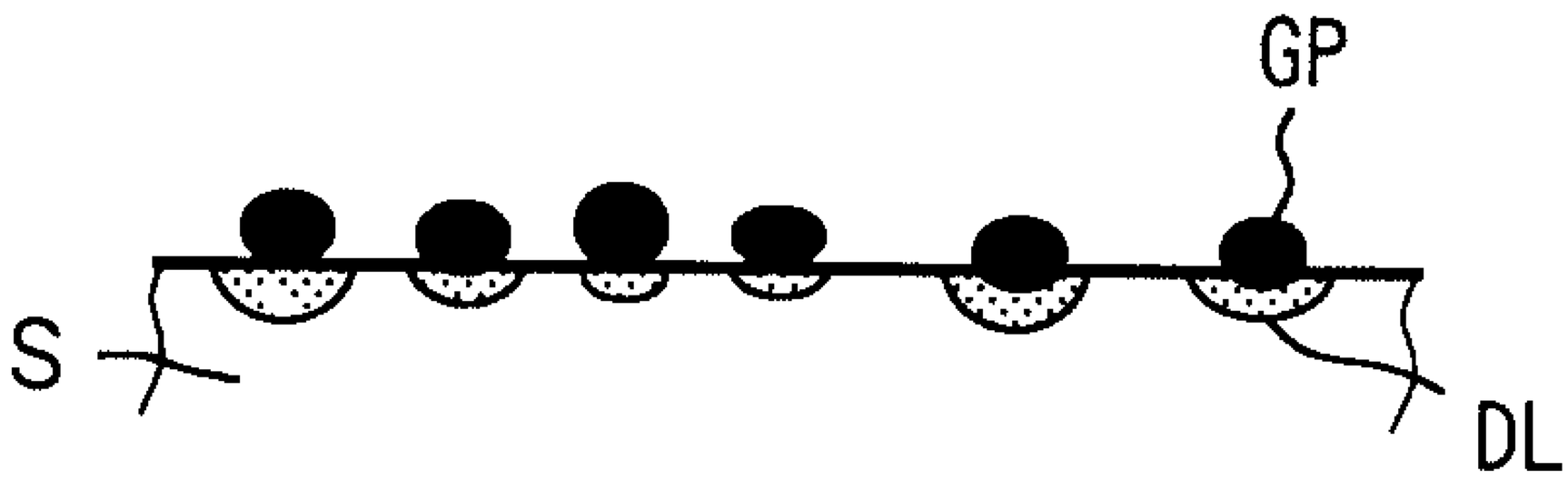
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(54) Title: A SEPARATOR OF A LOW-TEMPERATURE FUEL CELL AND MANUFACTURING METHOD THEREOF



(57) Abrégé/Abstract:
A separator for a low temperature type fuel cell using a stainless steel substrate S having carbon particles, such as graphite powder GP, and an aggregate CA of a carbon black, dispersed and press-bonded to the surface of the substrate in an island form, wherein the carbon particles are preferably bonded to the surface of the substrate S through a diffusion layer DL. The carbon particles such as the carbon aggregate CA and graphite powder GP may be dispersed in a plating layer, or a film component may be thermally decomposed after the carbon particles are dispersed in the film. A contact resistance is low because carbon powder having high conductivity is dispersed in the surface of the stainless steel substrate S having high oxidation resistance, and the occurrence of Joule heat which would otherwise result in a heat loss can be suppressed even when a large number of cells are stacked.

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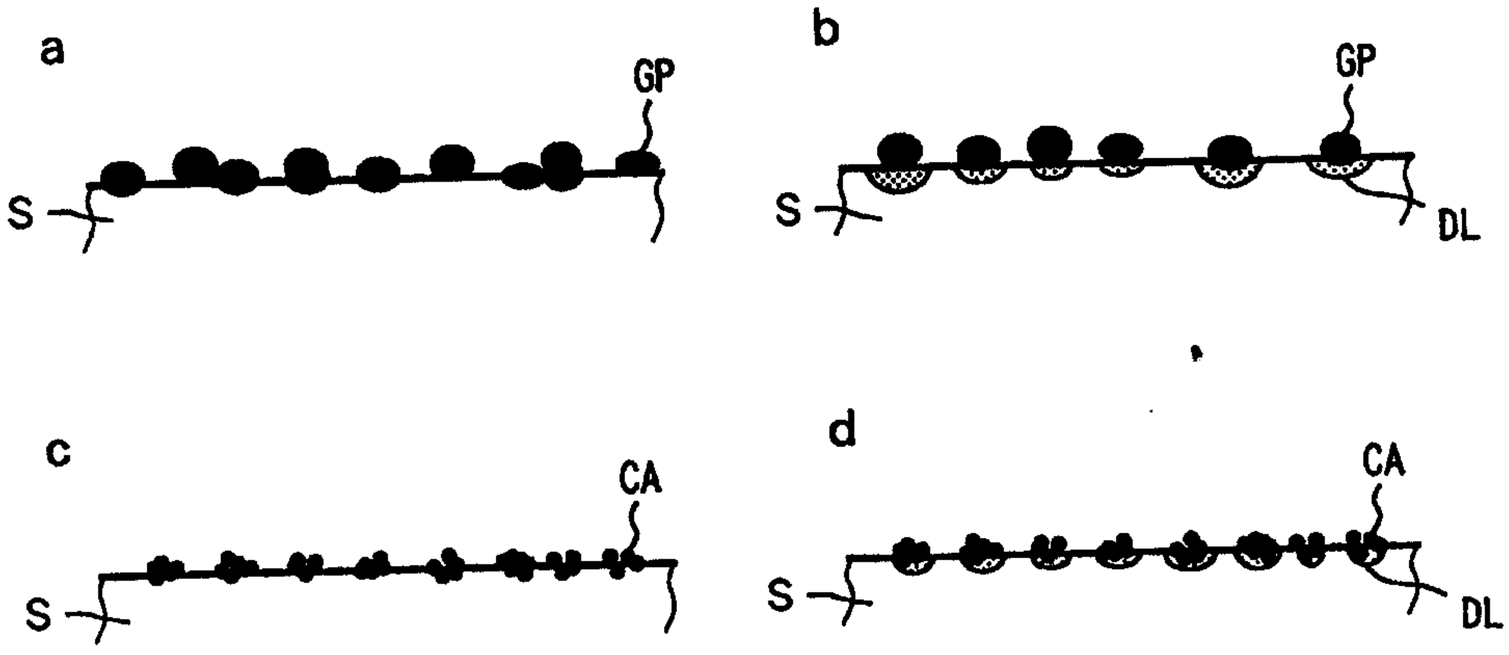
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(54)Title: SEPARATOR FOR LOW TEMPERATURE TYPE FUEL CELL AND METHOD OF PRODUCTION THEREOF

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(57) Abstract

A separator for a low temperature type fuel cell using a stainless steel substrate S having carbon particles, such as graphite powder GP, and an aggregate CA of a carbon black, dispersed and press-bonded to the surface of the substrate in an island form, wherein the carbon particles are preferably bonded to the surface of the substrate S through a diffusion layer DL. The carbon particles such as the carbon aggregate CA and graphite powder GP may be dispersed in a plating layer, or a film component may be thermally decomposed after the carbon particles are dispersed in the film. A contact resistance is low because carbon powder having high conductivity is dispersed in the surface of the stainless steel substrate S having high oxidation resistance, and the occurrence of Joule heat which would otherwise result in a heat loss can be suppressed even when a large number of cells are stacked.

SPECIFICATION

A SEPARATOR OF A LOW-TEMPERATURE FUEL CELL AND MANUFACTURING METHOD THEREOF

BACKGROUND OF THE INVENTION

The present invention relates to a metallic separator of such a fuel cell workable at a relatively low temperature as a solid macromolecular fuel cell and a manufacturing method thereof.

A solid macromolecular fuel cell has such advantages that it works at a temperature below 100°C and that it starts working in a short time, compared with other types of fuel cells. Since such the fuel cell has a structure composed of all solid members, it is easily maintained in an operable state and applicable for various uses subjected to vibrations or impacts. In addition, the fuel cell can be designed to a small size due to high power density. The fuel cell is also good of fuel efficiency with less noise. Accounting these advantages, application of the fuel cell to a motor installed in an electric automobile or the like has been researched and examined so far. If fuel cells which cover a long travelling distance similar to that of a gasoline engine is provided, an automobile which installs such fuel cells therein does not substantially put any harmful influences on the environment due to no generation of SO_x or NO_x with a reduction of CO_2 by half.

A conventional solid macromolecular fuel cell has a solid macromolecular membrane containing a proton-exchanging group in its molecular. The membrane acts as a proton-conductive electrolyte. An interior of the fuel cell is divided to two zones by the membrane. A fuel gas such as hydrogen is supplied to one of the zones, while an

oxidizing gas such as oxygen is supplied to the other zone, in the same manner as other types of fuel cells.

The fuel cell has an inner structure schematically illustrated in Fig. 1A. An air electrode 2 and a hydrogen electrode 3 are coupled to both sides of a solid macromolecular membrane 1, respectively. Both sides of the membrane 1 are faced through gaskets 4 to separators 5. An air-supply hole 6 and an air-discharge hole 7 are formed in the separator 5 at the side of the air electrode 2, while a hydrogen-supply hole 8 and a hydrogen-discharge hole 9 are formed in the other separator 5 at the side of the hydrogen electrode 3.

A plurality of grooves 10 which extend along flowing directions of hydrogen g and oxygen or air o are formed in the separators 5, in order to uniformly distribute hydrogen g and oxygen or air o. Cooling water w is fed through water-supply holes 11, circulated in the separators 5 and discharged through water-discharge holes 12 by water-cooling means provided in the separators 5, so as to release a heat during power generation.

Hydrogen g, which was fed through the hydrogen-supply hole 8 to a gap between the hydrogen electrode 3 and the separator 5, converts to a proton after discharge of an electron. The generated proton permeates through the solid macromolecular membrane 1, accepts an electron at the side of the air electrode 2, and burns with oxygen or air o which passes through a gap between the air electrode 2 and the separator 5. Consequently, an electric power is gained by charging a load between the air electrode 2 and the hydrogen electrode 3.

Since an electro motive force per one fuel cell is very tiny, a plurality of fuel cells are laminated together to gain a voltage

necessary for a practical use, as shown in Fig. 1B. Herein, a solid macromolecular membrane sandwiched between separators is handled as one unit. Due to the constitution that a plurality of fuel cells is laminated together, power-generating efficiency is significantly affected by resistance of the separators 5. A separator material good of electric conductivity with low contact resistance is necessary for improvement of power-generating efficiency. In this regards, graphite separators have been used so far with the same idea as that for a phosphate fuel cell, as disclosed in OHM Vol.83, No.7, p55-61, and FUJI GIHOH Vol.68, No.3, p.164-167).

Such a graphite separator is offered by cutting a graphite block to an objective shape and machining the shaped graphite block to form various holes and grooves. The cutting-machining process excessively consumes graphite material and needs expensive processing fees, so that a fuel cell as a whole is very expensive. The cutting-machining process is also inferior of productivity. Besides, a separator made of brittle graphite is easily broken or damaged by vibrations, impacts and so on. In order to overcome these disadvantages of a graphite separator, JP8-180883 A1 proposed a method of manufacturing a separator from a metal sheet by pressing, punching and so on.

However, when a metal sheet is used as a material for a separator of a fuel cell, there appears another problem. That is, a zone at a side of the air electrode 2 for passage of oxygen or air o is an acid atmosphere with pH 2-3. There has not been realized provision of a metallic material, which sufficiently endures in a strong acid atmosphere and exhibits properties necessary for use as a separator, e.g. superior electric conductivity, low contact resistance with

electrodes and corrosion resistance.

An acid-resistant material such as stainless steel could be used as a metallic material endurable in an acid atmosphere. Such a material exhibits excellent acid-resistance due to a passivated layer formed on its surface, but the passivated layer raises surface or contact electric resistance of the material with hydrogen and air electrodes. Elevation of the contact resistance means generation of a big quantity of a Joule heat at contact planes of separators to the hydrogen and air electrodes. Generation of the Joule heat causes wasteful consumption of an electric power gained by fuel cells, resulting in decrease of power-generating efficiency. Other metal sheets ordinarily also have oxide layers, which raise contact resistance, thereon.

Au is a metal material which does not have a passivated or oxide layer on its surface, and endurable in an acid atmosphere. However, Au is a very expensive material, so that it can not be practically used as a proper material for a separator of a fuel cell. Pt is also a metal material which is resistant to formation of a passivated or oxide layer on its surface and endurable in an acid atmosphere. However, Pt can not be used as a separator material due to its expensiveness.

In addition, a metal material for use as a separator shall be good of workability, since a plurality of grooves 10 or flanges for passages of hydrogen and air are formed by pressing, punching and so on. Workability of the metal material could be improved by applying an organic macromolecular film or a lubricating agent onto a surface of the metal material. However, application of an organic macromolecular film or lubricating agent raises contact resistance of

the metal material, so that a large quantity of a Joule heat would be generated in a power generator having a plurality of fuel cells laminated. Generation of a Joule heat means a loss of an electric power and reduces a power-generating efficiency of the power generator.

After a metal material to which a lubricating agent was applied is worked to an objective shape, the metal material shall be subjected to post-treatment such as degreasing and rinsing. Such post-treatment means an increase of processing steps, and also needs great expenditures for treatment of waste liquids. If the worked metal material is degreased using an organic or flon solvent, the atmosphere would be deteriorated by diffusion of the solvent. When an organic film is applied onto a surface of a metal material, the metal material can be worked to an objective shape without use of a lubricating agent. However, contact resistance of the metal material is raised by the applied organic film, and also the organic film is peeled off or dissolved away from a surface of the metal material due to its poor endurance in an acidic atmosphere.

SUMMARY OF THE INVENTION

The present invention is aimed at provision of a metallic separator which eliminates above-mentioned problems. Excellent electric conductivity and low contact resistance of the metallic separator is ensured without decrease of acid resistance by dotted distribution of carbonaceous particles on a surface of a stainless steel or formation of a metal plating layer or a paint film, in which carbonaceous particles are dispersed, on a surface of a stainless steel.

A first-type separator for a low-temperature fuel cell according

to the present invention is characterized by adhesion of carbonaceous particles onto a surface of a separator made from a corrosion-resistant metal sheet which has an oxide layer preformed in a corrosive atmosphere. A representative metal sheet as a substrate is a stainless steel having a passivated layer on its surface. Carbonaceous particles are preferably applied onto the surface of the substrate sheet with dotted distribution.

Carbonaceous particles are pressed onto a stainless steel sheet by applying the carbonaceous particles onto a surface of the stainless steel sheet and then rolling the stainless steel sheet with a reduction ratio of 0.1-50%, to improve adhesiveness and peeling-resistance of carbonaceous particles onto the stainless steel substrate. The stainless steel sheet may be heat-treated after pressing the carbonaceous particles. A diffusion layer effective for adhesiveness is formed between the carbonaceous particles and the stainless steel substrate by the heat treatment. The carbonaceous particles may be carbon black or graphite particles.

A second-type separator has a stainless steel substrate coated with a metal plating layer in which carbonaceous particles are dispersed in a state exposed to the atmosphere. The plating layer may be a Ni-Cr, Ti, Ta or Ti-Ta layer. The carbonaceous particles to be dispersed in the plating layer may be carbon black or graphite particles. The Ni-Cr plating layer preferably contains 5-60 wt.% Cr and optionally 0.3-40 wt.% Mo.

A third-type separator has a carbon-bonded layer composed of carbonaceous particles bonded through a diffusion layer onto a surface of a stainless steel substrate. Fine granular carbon adheres onto surfaces of the carbonaceous particles in the carbon-bonded layer. The

carbon-bonded layer can be formed by applying a carbonaceous particle-dispersed paint onto the stainless steel substrate, and then decomposing and vanishing organic components with a heat to remain the carbonaceous particles on the surface of the stainless steel substrate. Thermal decomposition of the paint film may be performed by heat-treatment at 300-1150°C in a non-oxidizing atmosphere. Before the heat-treatment, the stainless steel sheet coated with the paint film may be rolled with a reduction ratio of 0.1-50%.

BRIEF EXPLANATION OF THE DRAWINGS

Fig. **1A** is a sectional view illustrating an inner structure of a conventional fuel cell using a solid macromolecular membrane as an electrolyte.

Fig. **1B** is a bird eye's view of the same conventional fuel cell in a dismantled state.

Fig. **2A** is a sectional view illustrating a stainless steel substrate on which graphite particles are applied with a dotted distribution.

Fig. **2B** is a sectional view illustrating a stainless steel substrate having graphite particles bonded through a diffusion layer onto its surface.

Fig. **2C** is a sectional view illustrating a stainless steel substrate onto which aggregates of carbon black are applied with a dotted distribution.

Fig. **2D** is a sectional view illustrating a stainless steel substrate having aggregates of carbon black bonded through a diffusion layer onto its surface.

Fig. **3A** is a sectional view illustrating a stainless steel substrate

coated with a vapor-deposition Ni-Cr layer in which graphite particles are dispersed.

Fig. 3B is a sectional view illustrating a stainless steel substrate coated with an electroplating Ni-Cr layer in which graphite particles are dispersed.

Fig. 3C is a sectional view illustrating a stainless steel substrate coated with a vapor-deposition Ni-Cr layer in which aggregates of carbon black are dispersed.

Fig. 3D is a sectional view illustrating a stainless steel substrate coated with an electroplating Ni-Cr layer in which aggregates of carbon black are dispersed.

Fig. 4A is a sectional view illustrating a stainless steel substrate coated with a metal plating layer in which graphite particles are dispersed.

Fig. 4B is a sectional view illustrating a stainless steel substrate coated with a metal plating layer in which aggregates of carbon black are dispersed.

Fig. 5 is a sectional view illustrating a stainless steel substrate having a carbon-bonded layer to which fine granular carbon adheres.

PREFERRED EMBODIMENT OF THE PRESENT INVENTION

The present invention uses an austenitic or austenite-ferrite dual phase stainless steel superior of acid resistance as a substrate. Since a substrate for a separator of a fuel cell requires endurance from corrosive attacks of non-oxydizing acid as well as oxydizing acid, such a stainless steel contains Ni in addition to Cr as alloying components to improve acid-resistance. Due to excellent acid resistance of the substrate itself, a separator made of such a stainless

steel exhibits sufficient endurance even when pinholes or cracks occur in a plating layer formed on its surface.

An austenitic stainless steel suitable for such a purpose contains 14-35 wt.% Cr and 5-60 wt.% Ni. Compositions of the austenitic stainless steel are as follows as an example: 0.008-0.2 wt.% C, 0.05-5.0 wt.% Si, 0.1-5.0 wt.% Mn, 5.0-60 wt.% Ni, 14-35 wt.% Cr and the balance being Fe except optional elements and inevitable impurities.

An austenite-ferrite dual phase stainless steel suitable for such a purpose contains 17-35 wt.% Cr and 2-60 wt.% Ni. Compositions of the dual phase stainless steel are as follows as an example: 0.008-0.2 wt.% C, 0.05-5.0 wt.% Si, 0.1-5.0 wt.% Mn, 2.0-60 wt.% Ni, 17-35 wt.% Cr and the balance being Fe except optional elements and inevitable impurities.

If Cr content in the stainless steel is less than 14 wt.%, a separator would be inferior of endurance in a corrosive atmosphere including an oxydizing acid. If Cr content exceeds 35 wt.% on the contrary, the stainless steel would exhibit great reforming resistance resulting in poor workability during pressing or the like. If Ni content is less than 2 wt.%, a separator would be inferior of acid resistance in a corrosive atmosphere including a non-oxydizing acid. The effect of Ni on acid resistance is saturated at 60 wt.% Ni, and further improvement in acid resistance is not recognized by addition of Ni more than 60 wt.%. Besides, addition of excessive amount of Ni increases a cost of a stainless steel.

Acid resistance of a stainless steel substrate can be further improved by addition of one or more of Mo, Cu and N. When a fuel cell is operated in such the state that a current per a surface unit is

elevated for increase of a power density, a separator is exposed to an acidic atmosphere with a lower pH value. Corrosion attacks under such severe conditions are suppressed by addition of one or more of 0.2-7 wt.% Mo, 0.1-5 wt.% Cu and 0.02-0.5 wt.% N to a stainless steel. Acid resistance of the stainless steel is also improved by addition of a small amount of Ti, Nb and/or Zr as occasion demands.

The first-type separator has a stainless steel substrate onto which carbon particles such as graphite particles or carbon black aggregates directly adhere with dotted distribution. Graphite particles and carbon black are of high purity and superior acid resistance free from faults such as formation of an oxide film or the like caused by impurities. The high-purity graphite particles or carbon black also effectively protects a solid macromolecular membrane of a fuel cell from contamination. If such an unburnt product of petroleum or coke as soot or tar is used as carbonaceous particles, oxide films or the like would be easily formed on the carbonaceous particles due to impurities included therein. Such impurities also cause contamination of a solid macromolecular membrane and inferior performance of a fuel cell itself.

Carbonaceous particles such as graphite particles or carbon black aggregates exhibit low contact resistance and superior acid resistance without formation of an oxide film on their surfaces. In addition, a surface of a stainless steel substrate onto which carbonaceous particles adhere is good of affinity with air and hydrogen electrodes, since these electrodes are mainly made of a carbonaceous material. Consequently, contact resistance of the separator is remarkably lowered, so that a power generator comprising a plurality of fuel cells laminated together efficiently

outputs an electric power with less Joule heat.

Each graphite particles GP can be individually applied onto a surface of a stainless steel substrate S due to its size relatively bigger than carbon black, as shown in Fig. 2A. Application of graphite particles GP with dotted distribution may be performed by scrubbing the stainless steel substrate S with felt impregnated with graphite particles or a roll around which the same felt is wound. Graphite particles GP are pressed onto a surface of the stainless steel substrate S by rolling the stainless steel substrate with a reduction ratio of 0.1-50% after application of graphite particles GP. Carbon black is also applied onto a stainless steel substrate S in the same manner.

After graphite particles GP are pressed onto the stainless steel substrate S, the stainless steel substrate S is preferably heat-treated to form a diffusion layer DL between the stainless steel substrate S and each graphite particle GP, as shown in Fig. 2B. The diffusion layer DL effectively improves adhesiveness of the graphite particle GP to the stainless steel substrate S. Such the graphite particles GP improved in adhesiveness are not peeled off the surface of the stainless steel S, when the stainless steel S is subjected to pressing, corrugating or the like during which the surface of the stainless steel substrate S is scrubbed with dies. The diffusion layer DL is also effective for further reduction of contact resistance, since electric conduction to the stainless steel substrate S is assured through the diffusion layer DL.

Carbon black comprises very fine particles of 1 μm or less in size and likely aggregate. When carbon black is used as a carbonaceous material, carbon black adheres as aggregates CA onto a surface of a stainless steel substrate S, as shown in Fig. 2C. The

carbon black aggregates CA may be also pressed onto the surface of the stainless steel substrate S in the same way as for graphite particle GP. Adhesiveness of the carbon black aggregates CA is also improved by formation of a diffusion layer DL, as shown in Fig. 2D, by heat-treatment after pressing.

The graphite particles GP or the carbon black aggregates CA are preferably dispersed with dotted distribution onto a surface of the stainless steel substrate S, as shown in each of Figs. 2A-2D. The dotted distribution suppresses accumulation of a stress which occur during working such as bending or drawing with reformation, so that the graphite particles GP or the carbon black aggregates CA are prevented from dropping-out or peeling-off. If a surface of the stainless steel substrate S is completely coated with the graphite particles GP or the carbon black aggregates CA in such the state that each particle or aggregate is bonded together, a stress which occurs during machining is not released to anywhere and consequently accumulated in a boundary between the stainless steel substrate S and the graphite particle GP or the carbon black aggregate CA. Due to such accumulation of a stress, the graphite particles GP or the carbon black aggregate CA is easily peeled off or dropped out of the stainless steel substrate S.

A second-type separator for a low-temperature fuel cell has a metal plating layer in which carbonaceous particles are dispersed. For instance, a Ni-Cr plating layer PL formed on a surface of a stainless steel substrate S contains carbonaceous particles such as graphite particles GP or carbon black aggregate CA dispersed therein, as shown each of Figs. 3A-3D. This separator is useful not only for a solid macromolecular fuel cell shown in Fig. 1 but also for an alkali

fuel cell.

The plating layer PL in which the graphite particles GP or carbon black aggregates CA are dispersed is formed by a vapor depositing coating method, an electroplating method or the like. In case of vapor deposition coating, a surface of a stainless steel substrate is activated, carbonaceous particles are applied onto the activated surface, and Ni and Cr vapors with a predetermined flow ratio are introduced on to the surface so as to deposit Ni and Cr. In case of electroplating, a plating solution such as a composite plating solution containing nickel and chromium chlorides and suspending carbonaceous particles therein is used for co-precipitation of the carbonaceous particles during deposition of Ni and Cr.

Graphite particles GP are dispersed in the Ni-Cr plating layer PL in the state that each particle is exposed on a surface of the plating layer PL. The exposure of the graphite particles GP is controlled by conditions for formation of the Ni-Cr plating layer PL. Some amount of Ni or Cr may be deposited on the graphite particles GP during formation of the Ni-Cr plating layer PL. However, a Ni or Cr layer deposited on the graphite particles GP is spontaneously dropped out due to poor adhesiveness, so that the graphite particles GP are exposed on the surface without necessity of any special treatment. Even if the Ni or Cr layer deposited on the graphite particles GP is hardly dropped out, removal of such the Ni or Cr layer is facilitated by brushing the Ni-Cr plating layer as occasion demands.

When graphite particles GP relatively big in size are used, each graphite particle GP is dispersed to a Ni-Cr plating layer PL in an exposed state, as shown in Fig. 3A.

When a size of graphite particles GP is similar to a thickness of a Ni-Cr plating layer PL, each graphite particle GP serves as a direct electroconductive channel between a surface of the Ni-Cr plating layer PL and a stainless steel substrate S, as shown in Fig. 3B. When a thick Ni-Cr plating layer is formed on a stainless steel substrate S, as shown in Fig. 3C, such direct electroconductive channels are decreased in number, but low contact resistance is realized due to exposure of graphite particles GP on a surface of the Ni-Cr plating layer PL. The graphite particles GP dispersed in the Ni-Cr plating layer PL are also effective for improvement of electric conductivity of the Ni-Cr plating layer PL itself.

On the other hand, carbon black, which is very minute particles of 1 μm or less in size and easy to aggregate, is dispersed as carbon black aggregates CA in a Ni-Cr plating layer PL, as shown in Fig. 3C or 3D. Dispersion of such carbon black aggregates CA is also effective for decrease of contact resistance of the Ni-Cr plating layer PL.

An effect of carbonaceous particles such as graphite particles GP or carbon black aggregates CA on decrease of contact resistance or improvement of electric conductivity is clearly noted, when the carbonaceous particles is dispersed in the plating layer PL at a ratio of 0.01-500 mg/m^2. If an amount of the dispersed carbonaceous particles is less than 0.01 mg/m^2, carbonaceous particles exposed on the plating layer PL are reduced in number due to shortage, so as not to sufficiently realize the effect on decrease of contact resistance. If an amount of the dispersed carbonaceous particles exceeds 500 mg/m^2 on the contrary, the effect on decrease of contact resistance is saturated, but the plating layer PL is embrittled and likely peeled off a stainless steel substrate S.

A Ni-Cr plating layer PL in which graphite particles GP are dispersed sufficiently endures against oxidizing and non-oxidizing acids. Due to this superior acid resistance, the Ni-Cr plating layer PL effectively improves durability of a separator which is installed in a low-temperature fuel cell and exposed not only to an oxidizing atmosphere for passage of oxygen or air o but also to an oxygen-free non-oxidizing atmosphere at a gap.

A Ni-Cr plating layer PL preferably contains 5-60 wt.% Cr for acid resistance in both of oxidizing and non-oxidizing atmospheres. If Cr content in the Ni-Cr plating layer PL is less than 5 wt.%, the Ni-Cr plating layer PL is inferior of acid resistance in an oxidizing atmosphere. If Cr content exceeds 60 wt.%, acid resistance in a non-oxidizing atmosphere is inferior due to shortage of Ni content in return. Acid resistance of the Ni-Cr plating layer PL is further improved by addition of 0.3-40 wt.% Mo. An effect of Mo on improvement of acid resistance is clearly noted when Mo content is 0.3 wt.% or more. But, excessive addition of Mo more than 40 wt/% would rather deteriorate acid resistance.

Carbonaceous particles such as graphite particles GP or carbon black aggregates CA exhibit sufficiently low contact resistance and excellent acid resistance without formation of oxide films thereon. In addition, a Ni-Cr plating layer in which graphite particles GP or carbon black aggregates CA are dispersed is good of affinity to air and hydrogen electrodes which are made of carbonaceous material and held in contact with a separator in a fuel cell. Contact resistance is further reduced due to such good affinity. Consequently, when a plurality of cells are laminated together, a power generator superior of performance is constructed while suppressing generation of a Joule

heat.

Carbonaceous particles are prevented from dropping-out from a Ni-Cr plating layer PL during pressing or punching a stainless steel substrate S coated with the plating layer PL, since the carbonaceous particles are firmly retained in the plating layer PL. Due to good workability, the stainless steel substrate S coated with the plating layer PL can be reformed to a predetermined shape suitable for a separator.

A plating layer PL may be a Ti, Ta or Ti-Ta alloy layer instead of the Ni-Cr layer. A Ti, Ta or Ti-Ta alloy plating layer PL, in which carbonaceous particles such as graphite particles GP or carbon black aggregates CA are dispersed, is formed on a surface of a stainless steel substrate S by applying carbon particles onto the surface of a stainless steel substrate S and then introducing one or both of Ta and Ti vapors with a predetermined flow ratio so as to deposit Ta and/or Ti on the surface of the stainless steel substrate S.

Ta or Ti reacts with even a small amount of oxygen and forms a passive film on a plating layer PL. The plating layer PL coated with the passive film exhibits excellent resistance to both of oxidizing and non-oxidizing acids. A Ta-Ti alloy plating layer PL is also superior of acid resistance in the same meaning. In addition, carbonaceous particles are firmly retained in the Ta, Ti or Ta-Ti alloy plating layer PL, since Ta and Ti are reactive with carbon.

Graphite particles GP relatively big in size are dispersed to a plating layer PL in an exposed state, as shown in Fig. 4A. When carbon black very minute in size is used, carbon black aggregates CA are dispersed in a plating layer PL, as shown in Fig. 4B.

Exposure of graphite particles GP or carbon black aggregates

CA is controlled by conditions for formation of a plating layer PL. In this case, a Ta, Ti or Ta-Ti alloy layer deposited on the graphite particles GP or the carbon black aggregates CA is also spontaneously dropped out due to poor adhesiveness, and consequently the graphite particles GP or the carbon black aggregates CA are exposed on a surface of the plating layer PL without necessity of any special treatment.

A Ta, Ti or Ta-Ti alloy plating layer PL formed by a vapor deposition coating method is relatively thin, so that graphite particles GP or carbon black aggregates CA serves as direct electroconductive channels between a surface of the plating layer PL and the stainless steel substrate S.

Carbonaceous particles such as graphite particles GP or carbon black aggregates CA may be dispersed on a surface of a stainless steel substrate S by another method. That is, a paint in which graphite particles GP or carbon black aggregates CA are dispersed is applied onto a surface of a stainless steel substrate S, and then organic components in the paint film are decomposed and vanished with a heat so as to remain the graphite particles GP or the carbon black aggregates CA on the surface of the stainless steel substrate S. A third-type separator for a low-temperature fuel cell produced in this way comprises a stainless steel substrate S, a carbon-bonded layer L_1 formed on a surface of the stainless steel substrate S and an adhesion layer L_2 of granular carbon adhering onto carbonaceous particles of the carbon-bonded layer L_1.

The carbon-bonded layer L_1 and the adhesion layer L_2 each composed of carbonaceous particles can be provided by preparing a paint in which carbonaceous particles are dispersed, applying the

paint onto a stainless steel substrate S and then heating the stainless steel substrate S at 300-1150°C in a non-oxidizing atmosphere.

Organic components of the paint are decomposed by such heat treatment and vanished from the surface of the stainless steel substrate S. In this sense, no restrictions are put on a kind of a paint. Polyester, acrylic, polyolefine, polyurethane or these mixture may be used as a paint component for instance.

A mixing ratio of carbonaceous particles in a paint 100 parts by weight is preferably 0.05-60 parts by weight. If the mixing ratio is less than 0.05 parts by weight, a carbon-bonded layer L_1 in which carbonaceous particles are sufficiently dispersed can not be formed, so that contact resistance of a stainless steel substrate S is not decreased to a lower level. Addition of carbonaceous particles in an excessive amount above 60 parts by weight on the contrary causes poor applicability of a paint and also inferior adhesiveness of a paint film to a stainless steel substrate S.

A carbonaceous particle-dispersed paint film applied to a stainless steel substrate S is preferably of 5 μm or thinner, in order to assure adhesiveness of a carbon-bonded layer L_1 to the stainless steel substrate S. If a thickness of the paint film exceeds 5 μm, too much gasses would generate during heat treatment and cause peeling off of the paint film.

A stainless steel substrate S coated with a paint film in which carbonaceous particles are dispersed is heated at 300-1150°C in a non-oxidizing atmosphere such as N_2, N_2+H_2 or Ar. The carbonaceous particle-dispersed in the paint film remain as such on a surface of the stainless steel substrate S without oxidization due to the heat treatment in the non-oxidizing atmosphere. Carbon is partially

diffused into the stainless steel substrate S by the heat treatment, so as to form a carbon-bonded layer L_1 to which carbonaceous particles are bonded through a diffusion layer DL to the stainless steel substrate S.

Organic components such as resins in the paint film are decomposed by heat treatment and partially left as decomposition residues on the surface of the stainless steel substrate S. Carbonaceous decomposition residues derived from organic components are partially transferred to a carbon-bonded layer L_1, while the remainder is converted to a adhesion layer L_2 bonded to the carbon-bonded layer L_1. Since the carbon-bonded layer L_1 and the adhesion layer L_2 have the structure that each particle is bonded together without covering a whole surface of the stainless steel substrate S, the stainless steel substrate S keeps its good workability after formation of the carbon-bonded layer L_1 and the adhesion layer L_2. The adhesion layer L_2 serves as a lubricating agent during pressing or punching, so as to further improve workability of the stainless steel substrate S.

A stainless steel substrate S coated with a paint film may be rolled with a reduction ratio of 0.1-50% before heat treatment. Such rolling effectively improves adhesiveness of carbonaceous particles to the stainless steel substrate S. Carbonaceous particles improved in adhesiveness by the rolling promotes diffusion reaction to the stainless steel substrate S during heat treatment in the following step. Consequently, a carbon-bonded layer L_1 is firmly bonded to the stainless steel substrate S, so as to effectively decrease contact resistance. The effect of rolling on adhesiveness is clearly noted at a reduction ratio of 0.1% or more. But an excessive rolling ratio above

50% means excessive deformation of the stainless steel substrate S and causes peeling-off of the paint film. Such the effect on promotion of diffusion reaction is saturated at a reduction ratio of 50%.

Since an adhesion layer L_2 of granular carbon on a surface of a stainless steel substrate S serves as a lubricating agent, the stainless steel substrate S is reformed to even a complicated separator shape without rupture or cracking. In addition, a separator obtained in this way exhibits sufficiently low contact resistance due to presence of the carbon-bonded layer L_1 and the adhesion layer L_2 on the surface of the stainless steel substrate S, so that a power generator having such separators installed therein is superior of power generating efficiency and also durability under operational conditions exposed to a severely corrosive acidic atmosphere.

Since a carbon-bonded layer L_1 of carbonaceous particles is bonded through a diffusion layer DL to a stainless steel substrate S, the carbon-bonded layer L_1 is firmly fixed to the stainless steel substrate S. Consequently, even if such deformation as bending or elongation occurs in the stainless steel substrate S during reformation, accumulation of stress in the carbon-bonded layer L_1 is suppressed so that the carbon-bonded layer L_1 is prevented from peeling-off from the stainless steel substrate S. The superior adhesiveness of the carbon-bonded layer L_1 enables reformation of the stainless steel substrate S to a proper shape suitable for a separator. Workability of the stainless steel substrate is further improved by rolling after formation of the carbonaceous particle-dispersed paint film but before heat treatment.

Carbonaceous particles such as graphite particles GP or carbon black aggregates CA exhibits sufficiently low contact resistance and

excellent acid resistance without formation of oxide films thereon. In addition, the carbon-bonded layer L_1 and the adhesion layer L_2 is superior of affinity to air and hydrogen electrodes which are made of carbonaceous material and held in contact with a separator, so as to further decrease contact resistance. Consequently, a power generator having a plurality of fuel cells laminated together is superior of power generating efficiency with less Joule heat.

Other features of the present invention will be apparent from the following examples. However, these examples do not put any restrictions on a scope of the present invention.

EXAMPLE 1

Stainless steel sheets having compositions shown in Table 1 were used as stainless steel substrates S. Carbon black of 0.05 μm in average size and graphite particles of 3 μm in average size were used as carbonaceous particles.

TABLE 1: STAINLESS STEELS USED IN THE EXAMPLES

steel kind	compositions except Fe (wt.%)							
	C	Si	Mn	Ni	Cr	Mo	Cu	N
A	0.05	0.57	0.91	8.9	18.5	—	—	—
B	0.02	0.48	0.55	25.3	24.5	5.1	0.52	0.15
C	0.01	0.83	0.66	6.1	24.8	3.0	0.45	0.13

Steel A: austenitic stainless steel
Steel B: austenitic stainless steel
Steel C: austenite-ferrite dual phase stainless steel

Carbon black or graphite particles were applied with dotted distribution at an adhesion ratio of 5-10 mg/m^2 by scrubbing a stainless steel sheet with felt impregnated with carbon black or graphite particles. Due to very fine particle size, carbon black was applied as aggregates CA with dotted distribution onto a surface of the stainless steel substrate S. Graphite particles GP were individually applied with dotted distribution free from aggregation on a surface of the stainless steel substrate S.

The stainless steel substrate S was thereafter rolled with a reduction ratio of 2-3% so as to press the carbon black aggregates CA or the graphite particles GP onto the surface of the stainless steel substrate S for improvement of adhesiveness to the stainless steel substrate S. Some of samples were further heated at 700°C for 10 seconds, to generate a diffusion layer DL between the carbon black aggregates CA or the graphite particles GP and the stainless steel substrate S.

After the carbonaceous particles were applied with dotted distribution to the stainless steel substrate S, the stainless steel substrate S was testified for measurement of contact resistance and

acid resistance. Contact resistance was measured by holding the stainless steel substrate S in contact with a carbon electrode with a load of 10 kg/cm^2 and detecting contact resistance therebetween. Acid resistance was measured by dipping the stainless steel substrate S in a sulfuric acid solution of pH 2 at 90°C and measuring a corrosion loss after a predetermined time period. Samples prepared by coating a stainless steel sheet A with a Ni, Cu or Cr plating layer of 5μm in thickness were subjected to the same tests for comparison.

Test results are shown in Tables 2 and 3. Contact resistance and acid resistance of the stainless steel substrates S to which carbon particles were pressed with dotted distribution were shown in Table 2, while contact resistance and acid resistance of the stainless steel substrates S which were further heat-treated to generate diffusion layers DL were shown in Table 3.

It is clearly noted from Table 2 that sample Nos. 1-6, i.e. the stainless steel substrates to which carbon particles were pressed with dotted distribution, were bestowed with such properties as low contact resistance and excellent acid resistance necessary for a separator of a fuel cell. Sample Nos. 13-18 had contact resistance further decreased by the heat treatment to generate diffusion layers DL, as shown in Table 3.

On the other hand, any of sample Nos. 7-9, i.e. stainless steel substrates without adhesion of Carbon Particles, were too high of contact resistance for use as a separator of a fuel cell. Sample Nos. 10, 12, i.e. stainless steel substrates coated with Ni and Cr plating layers, exhibited low contact resistance, but big corrosion losses were detected so that these substrates were inappropriate as a separator of a fuel cell which is subjected to a corrosive atmosphere at a low pH

value. A sample No.11 was inferior both of contact resistance and acid resistance, so inappropriate as a separator of a fuel cell.

TABLE 2: EFFECT OF CARBON PARTICLES PRESSED WITH DOTTED DISTRIBUTION ON CONTACT RESISTANCE AND CORROSION LOSS (WITHOUT DIFFUSION LAYERS)

Sample No.	Steel Kind	Applied Carbon Particles	Contact Resistance $m\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$	Note
1	A	Carbon Black	15	0.00092	Present Invention
2	B	〃	21	0.00012	
3	C	〃	13	0.00015	
4	A	Graphite Particles	32	0.00089	
5	B	〃	33	0.00014	
6	C	〃	25	0.00011	
7	A	Without adhesion	307	0.00090	Comparative Examples
8	B	〃	279	0.00011	
9	C	〃	288	0.00012	
10	—	Ni Plating	26	0.17	
11	—	Cu Plating	120	0.098	
12	—	Cr Plating	36	0.0025	

TABLE 3: CONTACT RESISTANCE AND CORROSION LOSS OF STAINLESS STEEL SUBSTRATES TO WHICH CARBON PARTICLES WERE PRESSED AND THEN SUBJECTED TO HEAT-TREATMENT

Sample No.	Steel Kind	Applied Carbon Particles	Contact Resistance $M\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$
13	A	Carbon Black	5	0.00090
14	B	〃	7	0.00015
15	C	〃	6	0.00018
16	A	Graphite Particles	7	0.00097
17	B	〃	8	0.00013
18	C	〃	7	0.00019

EXAMPLE 2

In Example 2, stainless steel sheets shown in Table 1 were used as substrates. A Ni-Cr plating layer in which carbonaceous particles were dispersed was formed on a surface of each stainless steel substrate by the following vapor deposition coating or electroplating method.

Method 1: a vapor deposition coating method for formation of a Ni-Cr plating layer

Graphite particles of 2μm in average size and carbon black of 0.03μm in average size were used as carbonaceous particles. After a surface of a stainless steel sheet was activated in a vacuum chamber, the surface was scrubbed with felt impregnated with carbonaceous particles so as to apply carbonaceous particles to the surface of the stainless steel at an adhesion ratio of 3-15 g/m^2. Thereafter, Ni and Cr were simultaneously sputtered at a deposition speed of 0.005

μm/second in the same vacuum chamber.

The Ni-Cr plating layer PL formed in this way was of 0.5μm in thickness, and Cr content was 23 wt.%. Graphite particles GP and carbon black aggregates CA were dispersed in each plating layer PL, as shown in Fig. 3A and Fig. 3C, respectively.

Method 2: an electroplating method

for formation of a Ni-Cr plating layer

A plating solution was prepared by suspending carbon black of 0.03μm in average size at a ratio of 300 g/l in an aqueous solution containing 0.6 mol/l $NiCl_2$, 0.9 mol/l $CrCl_2$, 2.2 mol/l NH_4Cl, 0.8 mol/l H_3BO_3, 1.2 mol/l glycine and 3 wt.% a surfactant. A stainless steel sheet having a surface activated was dipped in the plating solution held at 40°C and electroplated at a current density of 10 A/dm^2.

A Ni-Cr plating layer PL formed in this way was of 5 μm in thickness, and Cr content was 40 wt.% on the basis of metal components in the plating layer PL. Graphite particles GP and carbon black aggregates CA were dispersed in each plating layer PL, as shown in Fig. 3B or Fig. 3D, respectively.

Each stainless steel substrate S coated with the plating layer PL in which carbonaceous particles were dispersed was testified for measurement of contact resistance and acid resistance by the same way as Example 1. Test results are shown in Table 4. It is noted from Table 4 that any of the stainless steel substrates S coated with the plating layer PL in which carbonaceous particles were dispersed had sufficiently low contact resistance and superior corrosion resistance, so suitable for a separator of a fuel cell.

TABLE 4: EFFECTS OF CARBONACEOUS PARTICLE-DISPERSED PLATING LAYERS ON CONTACT RESISTANCE AND CORROSION RESISTANCE

Sample No.	Steel Kind	A Plating Layer	Applied Carbonaceous Particles	Contact Resistance $m\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$	Note
1	A	A Ni-Cr Plating Layer containing 23 wt.% Cr	Carbon Black	5	0.00023	Method 1
2	B		〃	5	0.00010	
3	C		〃	4	0.00011	
4	A		Graphite Particles	10	0.00019	
5	B		〃	9	0.00010	
6	C		〃	10	0.00012	
7	A	A Ni-Cr Plating Layer containing 40 wt.% Cr	Carbon Black	6	0.00035	Method 2
8	B		〃	6	0.00018	
9	C		〃	6	0.00018	
10	A		Graphite Particles	12	0.00030	
11	B		〃	11	0.00010	
12	C		〃	11	0.00015	

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EXAMPLE 3

Stainless steel sheets shown in Table 1 were used as substrates, and carbonaceous particle-dispersed plating layers were formed on the stainless steel substrates by a vacuum deposition coating method.

Graphite particles of 2μm in average size and carbon black of 0.03μm in average size were used as carbonaceous particles. After a surface of each stainless steel sheet was activated in a vacuum chamber, the surface was scrubbed with felt impregnated with the carbonaceous particles so as to apply the carbonaceous particles at an adhesion ratio of 5-15 g/m^2 to the surface of the stainless steel substrate. Thereafter, one or both of Ta and Ti were simultaneously sputtered at a deposition speed of 0.004 μm/second in the same vacuum chamber. The plating layers PL formed in this way were of 0.5 μm in thickness. The graphite particles GP or the carbon black aggregates CA were dispersed in each plating layers PL, as shown in Fig. 4A and Fig.4B, respectively.

Each stainless steel substrate S coated with the carbonaceous particle-dispersed plating layer PL was testified for measurement of contact resistance and acid resistance by the same way as Example 1. Test results are shown in Tables 5 and 6. It is noted from these Tables 5 and 6 that any of the stainless steel substrates S coated with the carbonaceous particle-dispersed plating layers PL had sufficiently low contact resistance and superior acid resistance, so properties necessary for a separator of a fuel cell is satisfied.

TABLE 5: EFFECTS OF CARBONACEOUS PARTICLE-DISPERSED VACUUM DEPOSITION TA AND TI LAYERS ON CONTACT RESISTANCE AND CORROSION RESISTANCE

Sample No.	Steel Kind	A Plating Layer	Applied Carbonaceous Powder	Contact Resistance $m\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$	Note
1	A	Ta	Carbon Black	4	0.00025	Present Invention
2	B		〃	3	0.00010	
3	C		〃	3	0.00018	
4	A		Graphite Particles	7	0.00033	
5	B		〃	10	0.00012	
6	C		〃	8	0.00019	
7	A	Ti	Carbon Black	5	0.00045	
8	B		〃	5	0.00018	
9	C		〃	5	0.00010	
10	A		Graphite Particles	9	0.00040	
11	B		〃	7	0.00016	
12	C		〃	7	0.00021	

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TABLE 6: EFFECTS OF CARBONACEOUS PARTICLE-DISPERSED VACUUM DEPOSITION TA-TI LAYERS ON CONTACT RESISTANCE AND CORROSION RESISTANCE

Sample No.	Steel Kind	Ti Content (wt.%) In A Plating Layer	Applied Carbonaceous Powder	Contact Resistance $m\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$
13	A	20	Carbon Black	7	0.00022
14	B		〃	9	0.00011
15	A		Graphite Particles	8	0.00011
16	A		〃	9	0.00025
17	B		〃	9	0.00011
18	C		〃	11	0.00010
19	A		Carbon Black	8	0.00035
20	B	40	〃	6	0.00015
21	C		〃	9	0.00016
22	A		Graphite Particles	11	0.00036
23	B		〃	13	0.00012
24	C		〃	10	0.00019
25	A	70	Carbon Black	8	0.00037
26	B		〃	8	0.00014
27	C		〃	8	0.00010
28	A		Graphite Particles	11	0.00038
29	B		〃	10	0.00011
30	C		〃	14	0.00016

EXAMPLE 4

Stainless steel sheets shown in Table 1 were used as substrates.

Various kinds of paints to be applied to the stainless steel substrates S were prepared by dispersing graphite particles of 1μm in average size or carbon black of 0.05μm in average size as carbonaceous particles in an aqueous polyester urethane paint.

After each paint was applied as a film of 0.2-1.2μm in thickness to a stainless steel substrate S, the stainless steel substrate S was heated at 750°C for 5 seconds in a N_2 atmosphere. Organic substances of the paint were decomposed by the heat treatment. Granular carbon as a decomposition residue was left as a part of an adhesion layer L_2 of 0.1-1.0μm in average thickness on the surface of the stainless steel substrate S. The carbonaceous particles remaining after decomposition of organic substances were converted to a carbon-bonded layer L_1 of 0.01-1.0μm in average thickness which was bonded through a diffusion layer DL to the stainless steel substrate S.

Each stainless steel substrates S coated with the carbon-bonded layer L_1 of carbonaceous particles and the adhesion layer L_2 of granular carbon was testified for measurement of contact resistance and acid resistance by the same way as Example 1. Test results are shown in Table 7. It is noted from Table 7 that any of the stainless steel substrates S coated with the carbon-bonded layer L_1 and the adhesion layer L_2 exhibited sufficiently low contact resistance and superior acid resistance, so suitable for a separator of a fuel cell.

TABLE 7: CONTACT RESISTANCE AND CORROSION LOSS OF VARIOUS SEPARATORS MADEOF STAINLESS STEEL COATED WITH BINDING AND ADHESION LAYERS

Sample No.	Steel Kind	Applied Carbonaceous Particles	Average Thickness Of A Carbon-Bonded Layer (μm)	Contact Resistance $M\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$	Note
1	A	Carbon Black	0.01	5	0.00089	Present Invention
2	B	〃	0.02	4	0.00015	
3	C	〃	0.02	5	0.00018	
4	A	Graphite Particles	0.8	8	0.00093	
5	B	〃	0.8	8	0.00013	
6	C	〃	1.0	7	0.00014	

EXAMPLE 5

After a carbonaceous particle-dispersed paint was applied to each stainless steel sheet B shown in Table 1 under the same condition as Example 4, the stainless sheet was cold rolled at a reduction ratio shown in Table 8. The steel sheet was then heat treated under the same conditions as Example 4, to decompose and vanish organic substances in the paint. An obtained stainless steel substrate S, which was coated with carbon-bonded and adhesion layers L_1, L_2, was testified for measurement of contact resistance and acid resistance by the same way as Example 4. Test results are shown in Table 8.

It is noted from comparison of Table 8 with Table 7 that cold rolling prior to heat treatment is effective for increase of contact resistance, and that a decrease degree of contact resistance become larger as increase of a reduction ratio.

TABLE 8: EFFECTS OF REDUCTION RATIO ON CONTACT RESISTANCE AND CORROSION LOSS

Sample No.	Applied Carbonaceous Particles	Reduction Ratio %	Contact Resistance $m\Omega \cdot cm^2$	Corrosion Loss $g/m^2 \cdot h$
7	Graphite Particles	0.2	7.1	0.00016
8	〃	1.6	5.8	0.00015
9	〃	5.8	5.1	0.00017
10	〃	18.7	4.3	0.00017
11	〃	35.6	3.9	0.00016
12	〃	48.8	3.2	0.00016
13	Carbon Black	26.6	3.6	0.00015
14	〃	49.1	2.9	0.00013

INDUSTRIAL APPLICABILITY

A separator according to the present invention as above-mentioned comprises an acid-resistant stainless steel substrate coated with a carbonaceous particle-dispersed layer to improve electric conductivity. The carbonaceous particle-dispersed layer is formed by pressing carbonaceous particles with dotted distribution onto s surface of the stainless steel substrate or by forming a plating or paint layer in which carbonaceous particles are dispersed on the surface of the stainless steel substrate. A carbonaceous particle-dispersed layer effective for improvement of electric conductivity may be prepared by decomposing and vanishing organic substances of a carbonaceous particle-dispersed paint layer formed on a surface of a stainless steel substrate.

The stainless steel substrate to which carbonaceous particles adhere with dotted distribution is superior both of electric conductivity and corrosion resistance, so useful as a separator of a low-temperature fuel cell. Due to excellent properties, a power generator having the structure that a plurality of low-temperature fuel cells are laminated together keeps its high performance for a long time with less corrosion even in a strong acid atmosphere and also suppresses a heat loss which would be derived from a Joule heat when a plurality of fuel cells are laminated. In addition, the metallic separator is manufactured with good productivity while avoiding increase of material and manufacturing costs.

CLAIMS:

1. A separator for a low-temperature fuel cell comprising a stainless steel substrate, on which an oxide film is generated in a corrosive atmosphere to keep acid resistance, and carbonaceous particles applied onto a surface of said stainless steel substrate.

2. The separator for a low-temperature fuel cell according to Claim 1, wherein the carbonaceous particles are applied with dotted distribution onto the surface of said stainless steel substrate.

3. The separator for a low-temperature fuel cell according to Claim 2, wherein the carbonaceous particles are pressed onto the surface of the stainless steel substrate by rolling said stainless steel substrate at a reduction ratio of 0.1-50%.

4. The separator for a low-temperature fuel cell according to any one of Claims 1-3, wherein the carbonaceous particles are bonded through a diffusion layer to the surface of the stainless steel substrate.

5. The separator for a low-temperature fuel cell according to any one of Claims 1-4, wherein the carbonaceous particles are carbon black or graphite particles.

6. A separator for a low-temperature fuel cell comprising a stainless steel substrate and a Ni-Cr plating layer, in which carbonaceous particles are dispersed, formed on a surface of said stainless steel substrate, wherein said carbonaceous particles are exposed on a surface of said Ni-Cr plating layer.

7. A separator for a low-temperature fuel cell comprising a stainless steel substrate and a Ti, Ta or Ti-Ta alloy plating layer, in which carbonaceous particles are dispersed, formed on a surface of said stainless steel substrate, wherein said carbonaceous particles are exposed on a surface of said Ti, Ta or Ti-Ta alloy plating layer.

8. The separator for a low-temperature fuel cell according to Claim 6 or 7, wherein the carbonaceous particles are carbon black or graphite particles.

9. The separator for a low-temperature fuel cell according to Claim 6, wherein the Ni-Cr plating layer contains 5-60 wt.% Cr.

10. The separator for a low-temperature fuel cell according to Claim 9, wherein the Ni-Cr plating layer further contains 0.3-40wt.% Mo.

11. A separator for a low-temperature fuel cell comprising a stainless steel substrate, a carbon-bonded layer composed of carbonaceous particles bonded through a diffusion layer onto a surface of said stainless steel substrate and an adhesion layer of granular carbon adhering onto a surface of said carbon-bonded layer.

12. The separator for a low-temperature fuel cell according to Claim 11, wherein the carbon-bonded layer is composed of carbonaceous particles which remain after decomposition and vanishment of organic substances in a carbonaceous particle-dispersed paint film which was applied onto a surface of the stainless steel substrate and then subjected to heat treatment.

13. A method of manufacturing a separator for a low temperature fuel cell, comprising steps of:

- providing a stainless steel substrate,
- applying a paint in which carbonaceous particles are dispersed onto said stainless steel substrate, and
- heating said stainless steel substrate at 300-1150° C in a non-oxidizing atmosphere.

14. The method of manufacturing a separator for a low-temperature fuel cell according to Claim 13, wherein the stainless steel substrate coated with the paint film is rolled at a reduction ratio of 0.1-50% prior to the heat treatment.

FIG.1A

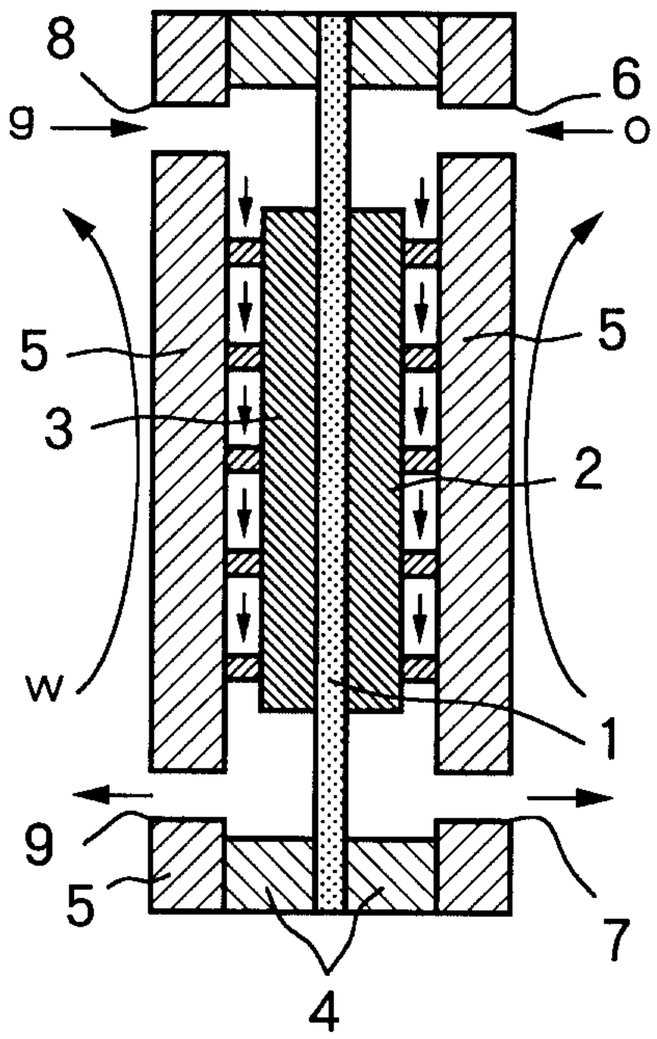
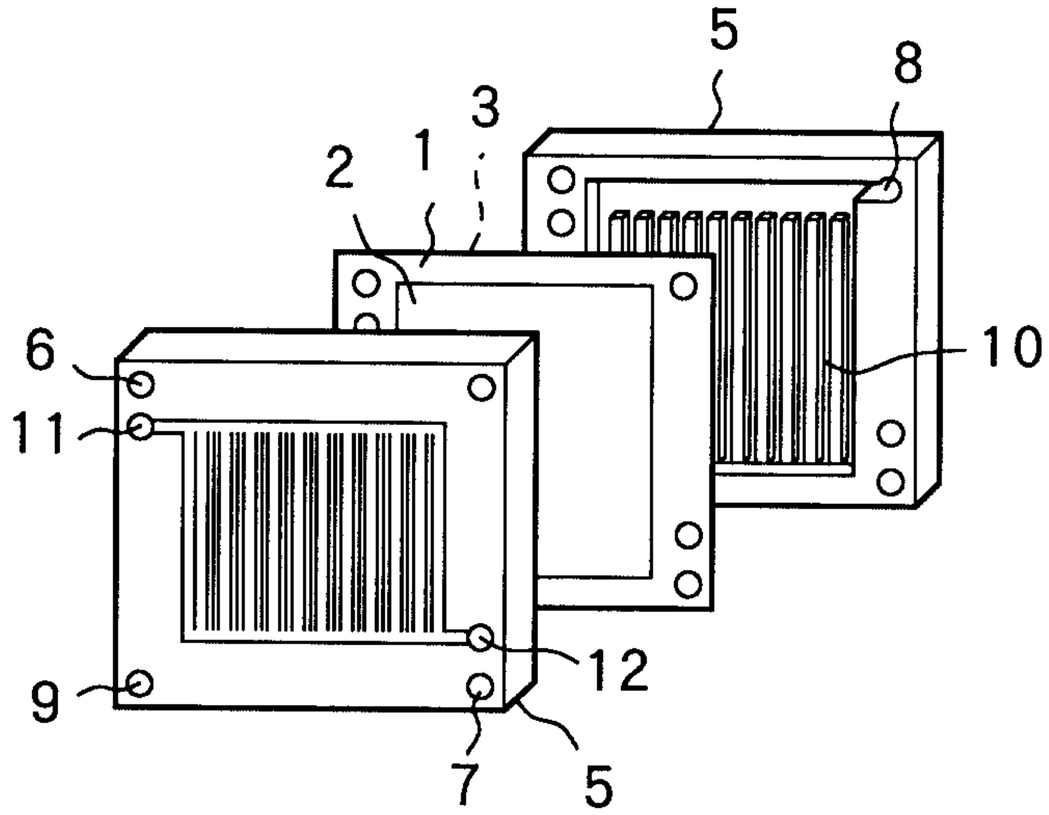


FIG.1B



(PRIOR ARTS)

FIG.2A

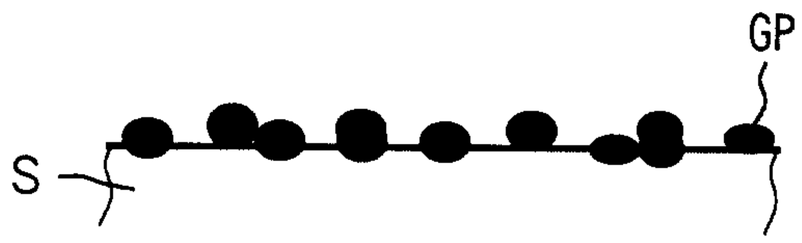


FIG.2B

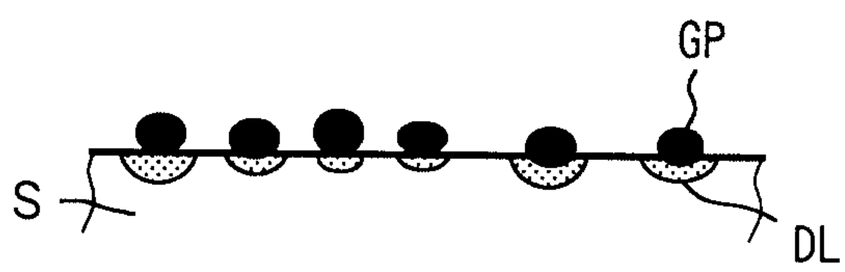


FIG.2C

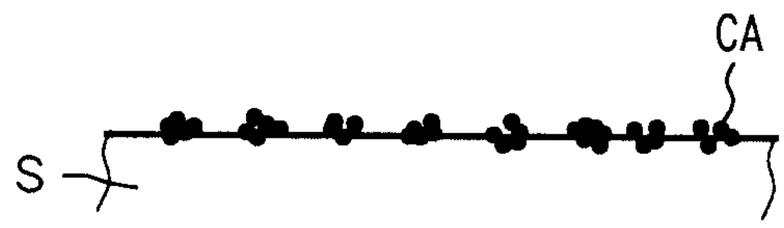


FIG.2D

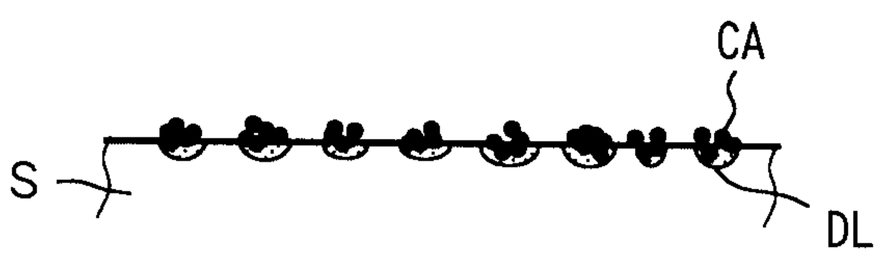


FIG.3A

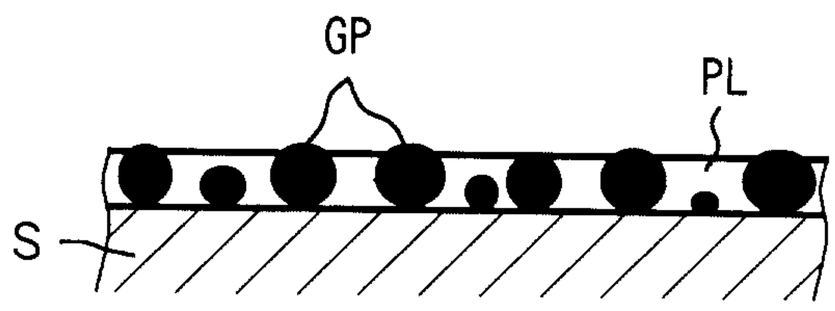


FIG.3B

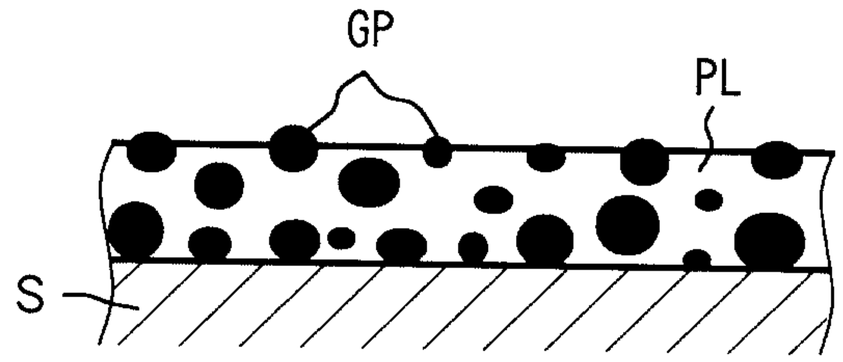


FIG.3C

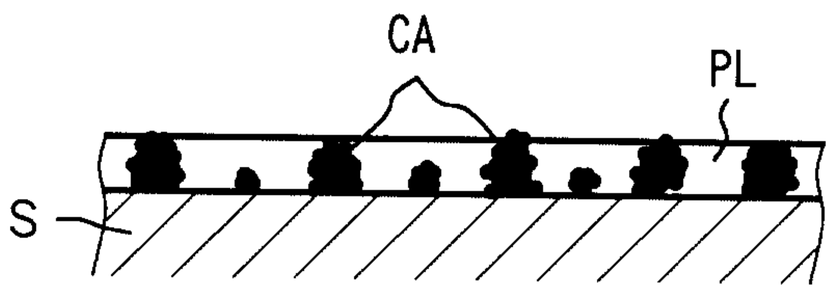


FIG.3D

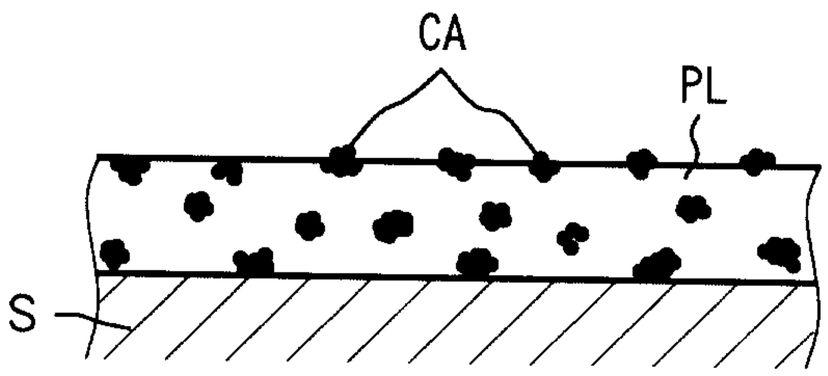


FIG.4A

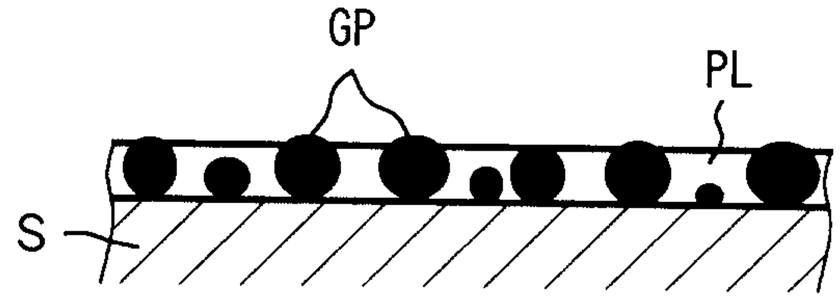


FIG.4B

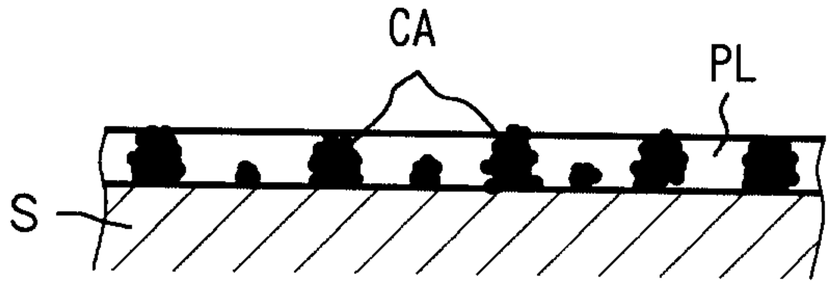
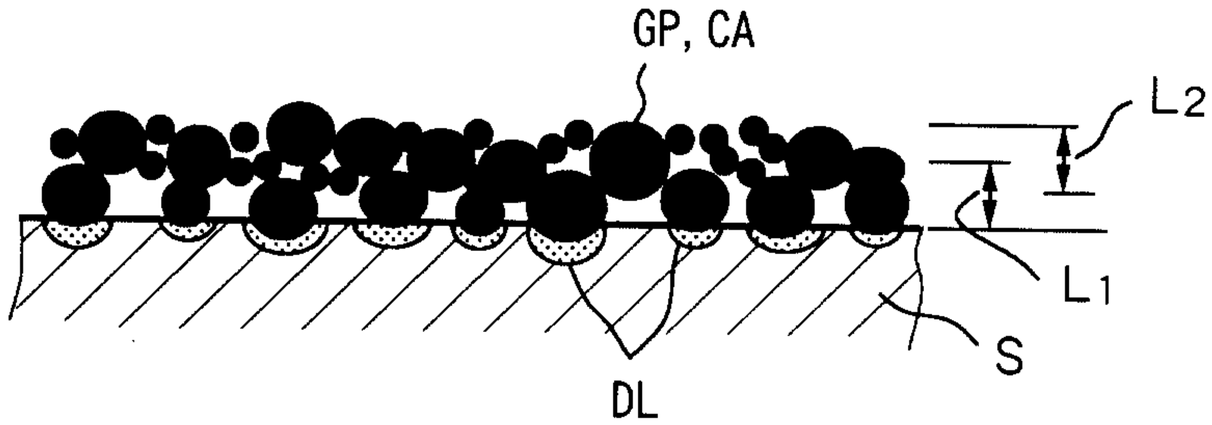
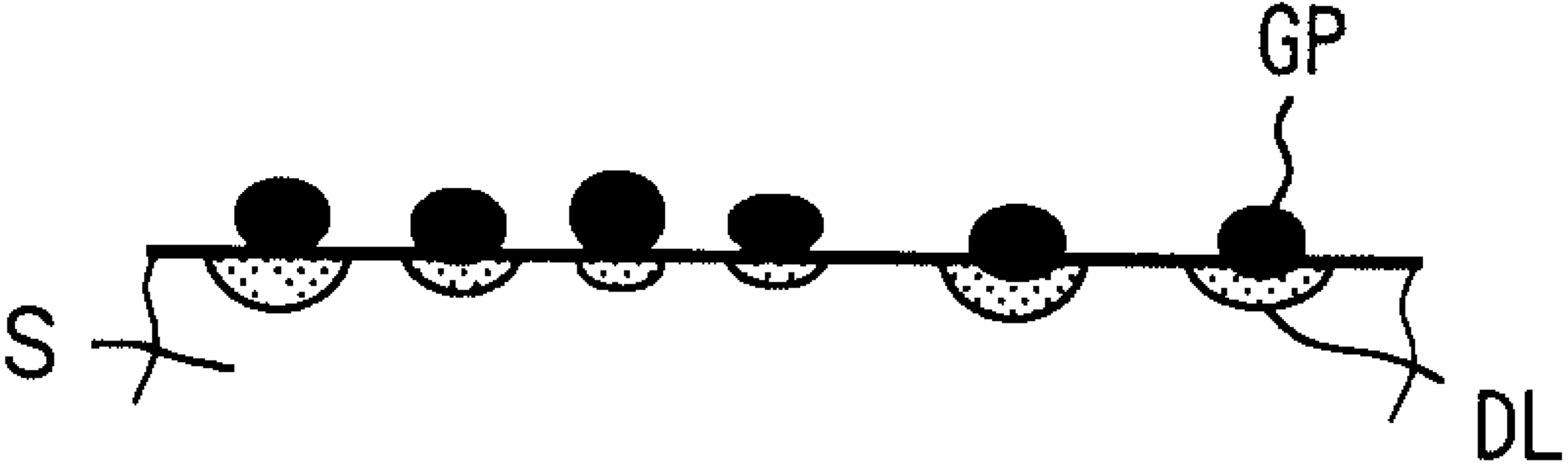


FIG.5




GP
S
DL