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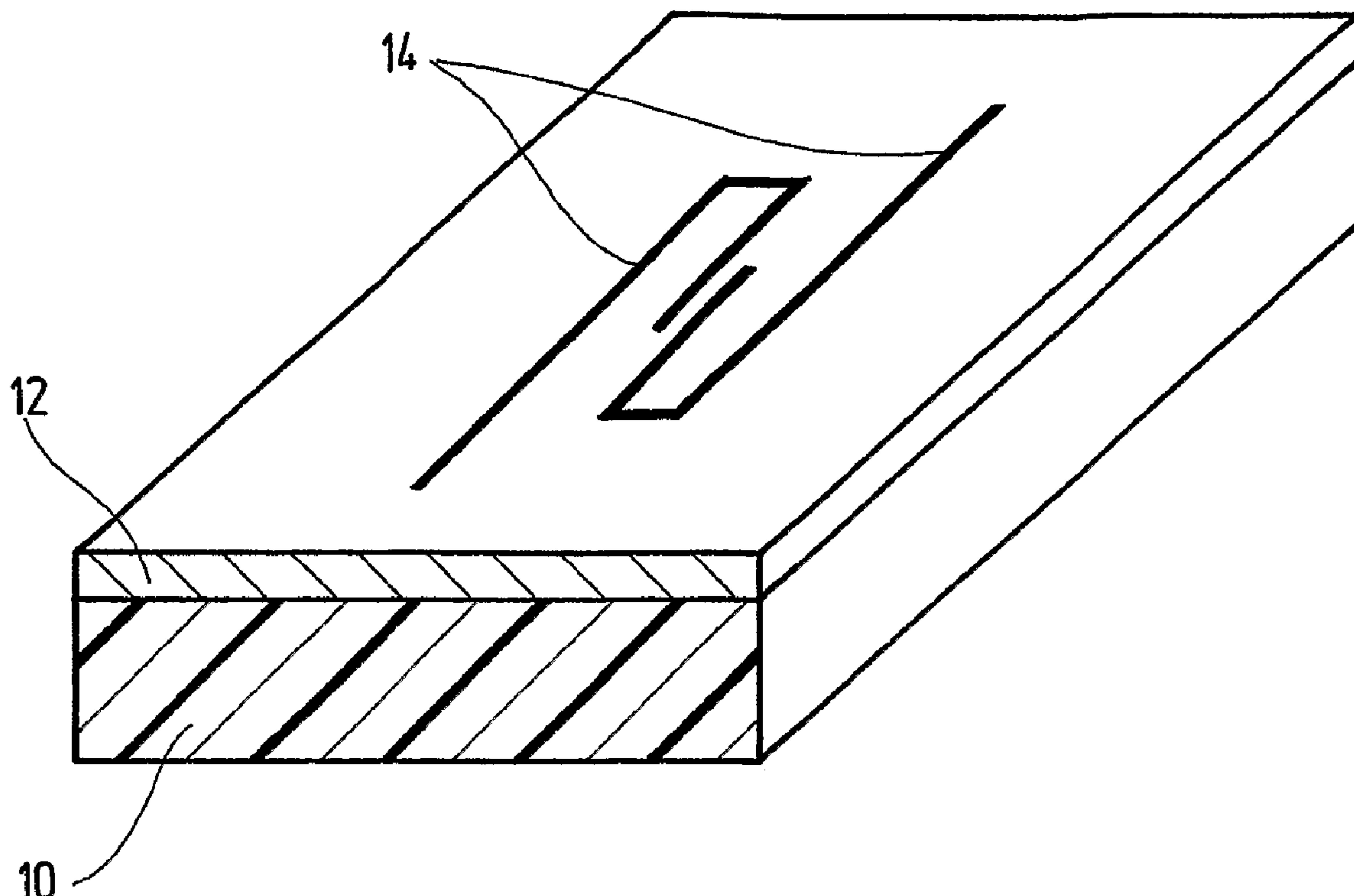
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(54) Titre : PROCEDE DE PRODUCTION D'UN SUBSTRAT HYDROPHILE POURVU D'UNE ELECTRODE EN COUCHE
(54) Title: METHOD FOR THE PRODUCTION OF A HYDROPHILIC SUBSTRATE PROVIDED WITH A LAYER
ELECTRODE



(57) Abrégé/Abstract:

Method for the production of a hydrophilic substrate provided with a layer electrode. The invention concerns a method for producing an insulating substrate provided with a layer electrode especially for an analytical test strip. A combination of the following method steps is disclosed: the layer electrode (14) is formed as a structured surface pattern on the substrate (10) and the hydrophilicity of a cover layer (12) on the substrate (10) is increased by a chemical or physical surface treatment.

Abstract

Method for the production of a hydrophilic substrate provided with a layer electrode. The invention concerns a method for producing an insulating substrate provided with a layer electrode especially for an analytical test strip. A combination of the following method steps is disclosed: the layer electrode (14) is formed as a structured surface pattern on the substrate (10) and the hydrophilicity of a cover layer (12) on the substrate (10) is increased by a chemical or physical surface treatment.

Method for the production of a hydrophilic substrate provided with a layer electrode

Description

The invention concerns a method for producing an insulating substrate provided with a layer electrode especially for an analytical test strip and a corresponding product.

Structured electrodes of this type can be used in microfluidic test strips for electrochemical measurements where a capillary channel structure located on the carrier or substrate enables a spatial separation of the sample application site and detection zone using minimal sample volumes. Polymer foils which do not absorb sample liquid and, due to their high hydrophobicity, facilitate coating with an electrode material such as gold but only have a low wettability for the usually aqueous sample liquids such as blood, are frequently used as support materials for the economical mass production of such analytical units. Hence the sample liquids only flow very slowly and inhomogeneously over this material and the uptake of the analyte into the test strip system takes an unacceptably long time for the user.

Arrangements having layers of foils glued on top of one another have already been used as a remedy, for example by gluing a thin masking foil with a hydrophilic surface onto parts of a conductive unstructured electrode layer. However, several complicated production steps are required for this and in such an arrangement steps are formed on the test strip which have to be surmounted by an inflowing liquid. This can stop the sample liquid and thus the analyte may not reach the actual measuring field or only imperfectly.

It is known from WO 99/29435 that a surface layer can be hydrophilized i.e. its affinity for water can be increased by the action of water in order to increase the surface tension of objects. In particular a metallic layer deposited on the object is oxidized beyond its natural oxide layer to such an extent that it loses its metallic appearance and may become completely transparent. The disclosure of WO

99/29435 describes this type of chemical hydrophilization.

The increase in surface tension in this treatment results from an increase in polarity and corresponds to an increased hydrophilicity of the observed surfaces.

Hydrophilicity is the water affinity of a surface. In this connection hydrophilic surfaces are water-attracting surfaces. Aqueous samples also including biological samples such as blood, urine, saliva, sweat and samples derived therefrom such as plasma and serum spread well on such surfaces. One characteristic of such surfaces is that a boundary surface of a water drop forms an acute rim or contact angle on them. In contrast an obtuse rim angle is formed at the interface between a water drop and a surface on hydrophobic i.e. water repellent surfaces.

The rim angle which is a result of the surface tension of the test liquid and of the surface to be examined is a measure of the hydrophilicity of a surface. Water for example has a surface tension of 72 mN/m. If the value of the surface tension of the observed surface is much, i.e. more than 20 mN/m, below this value then the wetting is poor and the resulting rim angle is obtuse. Such a surface is referred to as hydrophobic. If the surface tension approximates the value which is found for water then the wetting is good and the rim angle is acute. If, in contrast, the surface tension is the same as or higher than that of the value found for water, then the drop runs and there is a total spreading of the liquid. It is then no longer possible to measure a rim angle. Surfaces which form an acute rim angle with water drops or on which a total spreading of a water drop is observed are referred to as hydrophilic.

On this basis the object of the invention is to provide a method for producing a substrate provided with a layer electrode and to provide such a product where the known disadvantages are at least reduced or even eliminated and in particular to create an arrangement of electrodes on a support optimized as a transport path for polar liquids in a simple manufacturing process.

In one aspect of the invention, there is provided a process for producing an insulating substrate provided with a layer electrode, the substrate having a capillary channel structure for transporting an aqueous bioliquid into an electrode area in order to carry out electrochemical analyses, comprising the following process steps:

- a) the layer electrode is formed as a structured surface pattern on the substrate,
- b) after forming the layer electrode, the substrate is provided with a cover layer,
- c) water affinity of the cover layer and hence functionality of capillary transport paths is increased by a chemical surface treatment.

In another aspect of the invention, there is provided an insulating substrate provided with a layer electrode, the substrate having a capillary channel structure for the transport of an aqueous bioliquid into an electrode area in order to carry out electrochemical analyses, in which the layer electrode has an electrically conductive surface structure and is arranged under or on a hydrophilic cover layer, wherein an increased water affinity of the cover layer improves functionality of capillary transport paths.

In still another aspect of the invention, there is provided the insulating substrate of the invention for use in an analytical test strip.

Accordingly from a process point of view the following combination of process steps is provided:

- a) the layer electrode is formed as a structured surface pattern on the substrate,
- b) the water affinity of a cover layer of the substrate is increased by a chemical or physical surface treatment.

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This allows an optimized surface in which the conducting and hydrophilic layer of a low layer thickness adjoin one another while avoiding steps to be created in a process sequence having a few steps that is suitable for mass production.

The substrate is advantageously coated with a starting material for the cover layer and preferably vapour-deposited with aluminium as a starting material. This also enables a hydrophobic substrate material to be provided with a hydrophilic surface in a simple manner without impairing the production of a defined electrode structure.

Another preferred embodiment provides that, after applying the layer electrode, the substrate is preferably provided with the cover layer over its entire surface. It surprisingly turned out that when the cover layer has a suitable thickness, the functionality of the electrode is preserved and the production of the electrode structure on the yet uncoated substrate is considerably facilitated.

Alternatively it is also possible to apply the layer electrode to the cover layer. In this case the cover layer can be hydrophilized before or after applying the cover electrode according to the above-mentioned process step b).

With regard to the process step a) it is advantageous when the uncoated substrate or substrate already provided with the cover layer is covered with an electrode layer to form the layer electrode by a thin layer deposition process in particular by

evaporation coating or sputtering. A geometric structure can be advantageously produced by forming the layer electrode by selectively ablating certain areas preferably by laser ablation of a previously formed electrode layer.

Alternatively it is conceivable that the layer electrode is already structured when it is applied using a mask or stencil in which areas are cut out. This may also be achieved by applying the layer electrode by a printing process.

The layer electrode advantageously has a layer thickness of less than 10 micrometers, preferably of less than 100 nanometers. It is also favourable when the layer electrode consists of a metallic electrode material preferably of gold, platinum, palladium or iridium. However, in principle other conducting materials such as graphite can also be used as electrode material.

In a preferred embodiment the substrate is composed of a hydrophobic insulator material and in particular a polymer foil.

Another advantageous embodiment provides that the cover layer is firstly hydrophobic and becomes hydrophilic due to surface treatment preferably with formation of an inorganic oxide layer. This can be achieved particularly simply in a manufacturing process when the cover layer is surface treated by the action of water in which case the cover layer consists of an inorganic starting material that can be oxidized with water and is hydrophilized by treatment with hot water or water vapour.

A suitable chemical surface modification can also be achieved by hydrophilizing the cover layer by hydrolysis of a phosphoric acid ester.

The cover layer should advantageously have a layer thickness of less than 100 micrometers, preferably less than 50 micrometers.

Another aspect of the invention is a product comprising an insulating substrate provided with a layer electrode especially for an analytical test strip in which the

layer electrode has an electrically conductive surface structure and is arranged under or on a hydrophilic cover layer.

The invention is elucidated in more detail in the following on the basis of an embodiment example shown in the drawing in a schematic manner.

Fig. 1 shows a diagram of a substrate provided with a structured layer electrode and

Fig. 2 shows the process sequence for producing an arrangement according to fig. 1 in a block diagram.

The arrangement shown in fig. 1 is essentially composed of an electrically insulating substrate 10, a hydrophilic cover layer 12 applied to the substrate 10 and a structured layer electrode 14 arranged on or under the cover layer 12. Such an arrangement can be preferably used to construct analytical test strips which have a capillary channel structure on a support foil (substrate) for transporting an aqueous bioliquid into the electrode area in order to carry out electrochemical analyses.

In order to economically manufacture such single-use test strips it is possible to use plastic-based support materials or substrates 10 and in particular polymer foils. The functionality of the capillary transport path that is not shown can be ensured by a hydrophilic cover layer 12 and the electrode structure 14 creates a defined detection zone.

Fig. 2 illustrates the process sequence for manufacturing such electrode supports. Firstly the entire area of one side of a polyester foil 10 as a substrate is coated with a gold layer 16 of ca. 50 nm thickness by vapour depositing or sputtering gold. The coated foil is then bombarded as a target with a laser beam 18 in order to ablate or evaporate certain areas of the gold layer 16. Laser ablation allows a microstructured surface pattern to be exposed and by means of a targeted transfer of energy it is possible to layer-selectively ablate the gold layer.

In the next process step an aluminium layer 20 with a layer thickness of ca. 50 nm or 100 nm is vapour-deposited onto the substrate 10 and the electrode structure 14 which has been generated thereon. Subsequently the hydrophilicity of this layer is increased by chemical treatment or modification. For this purpose the aluminium layer 20 is oxidized by boiling in a water-bath or treatment with water vapour by which means the hydrophilic aluminium oxide / hydroxide layer and cover layer 12 formed in this manner has a permanently high surface tension and polarity in order to achieve good flow properties of a polar liquid sample. In this connection it has turned out that as a result of the low layer thickness and porosity of the oxide layer 12, the functionality of the electrode structure 14 is essentially not impaired.

The order of the processes described above can in principle be varied depending on the materials that are used. Thus the substrate 10 can firstly be coated with aluminium and the electrode structure 14 can be generated on the aluminium layer. In this case it is possible to convert the aluminium layer into a hydrophilic cover layer before or after applying the electrode structure where it is basically possible to also use physical methods for the hydrophilization such as plasma treatment.

CLAIMS:

1. Process for producing an insulating substrate provided with a layer electrode, the substrate having a capillary channel structure for transporting an aqueous bioliquid into an electrode area in order to carry out electrochemical analyses, comprising the following process steps:
 - a) the layer electrode is formed as a structured surface pattern on the substrate,
 - b) after forming the layer electrode, the substrate is provided with a cover layer,
 - c) water affinity of the cover layer and hence functionality of capillary transport paths is increased by a chemical surface treatment.
2. Process for producing an insulating substrate provided with a layer electrode, the substrate having a capillary channel structure for transporting an aqueous bioliquid into an electrode area in order to carry out electrochemical analyses, comprising the following process steps:
 - a) the substrate is coated with a starting material of a cover layer,
 - b) the layer electrode is formed as a structured surface pattern on the cover layer,
 - c) before or after forming the layer electrode, water affinity of the cover layer and hence functionality of capillary transport paths is increased by a chemical surface treatment.
3. Process as claimed in claim 2, characterized in that the substrate is vapour-deposited with aluminium as the starting material of the cover layer.
4. Process as claimed in any one of claims 1 to 3, characterized in that the substrate is provided with the cover layer over its entire surface.
5. Process as claimed in any one of claims 1 to 4, characterized in that the substrate is covered with an electrode layer to form the layer electrode by a thin layer deposition process.

6. Process as claimed in any one of claims 1 to 4, characterized in that the substrate already provided with the cover layer is covered with an electrode layer to form the layer electrode by a thin layer deposition process.
7. Process as claimed in claim 5 or 6, characterized in that the thin layer deposition process comprises evaporation coating or sputtering.
8. Process as claimed in any one of claims 1 to 7, characterized in that the layer electrode is geometrically structured by selectively ablating certain areas of a previously formed electrode layer.
9. Process as claimed in claim 8, characterized in that the ablating comprises laser ablation.
10. Process as claimed in claim 8 or 9, characterized in that the entire area of the electrode layer is provided with a reagent film before structuring the layer electrode.
11. Process as claimed in any one of claims 1 to 7, characterized in that the layer electrode is structured when it is applied using a mask.
12. Process as claimed in any one of claims 1 to 7, characterized in that the layer electrode is applied in a structured manner by a printing process.
13. Process as claimed in any one of claims 1 to 12, characterized in that the layer electrode has a layer thickness of less than 10 micrometers.
14. Process as claimed in any one of claims 1 to 12, characterized in that the layer electrode has a layer thickness of less than 100 nanometers.
15. Process as claimed in any one of claims 1 to 14, characterized in that the layer electrode is composed of a metallic electrode material.
16. Process as claimed in claim 15, characterized in that the metallic electrode material is of gold, platinum, palladium or iridium.
17. Process as claimed in any one of claims 1 to 16, characterized in that the substrate is composed of a hydrophobic insulating material.

18. Process as claimed in claim 17, characterized in that the hydrophobic insulating material is a polymer foil.
19. Process as claimed in any one of claims 1 to 18, characterized in that the cover layer is first hydrophobic and is hydrophilized by surface treatment.
20. Process as claimed in claim 19, characterized in that the surface treatment is with formation of an inorganic oxide layer.
21. Process as claimed in claim 2 or 3, characterized in that the surface treatment of the cover layer takes place by the action of water.
22. Process as claimed in any one of claims 1 to 21, characterized in that the cover layer is composed of an inorganic starting material that can be oxidized by water and is hydrophilized by treatment with hot water or water vapour.
23. Process as claimed in any one of claims 1 to 21, characterized in that the cover layer is hydrophilized by hydrolysing a phosphoric acid ester.
24. Process as claimed in any one of claims 1 to 23, characterized in that the cover layer is formed with a layer thickness of less than 100 micrometers.
25. Process as claimed in any one of claims 1 to 23, characterized in that the cover layer is formed with a layer thickness of less than 50 micrometers.
26. An insulating substrate provided with a layer electrode, the substrate having a capillary channel structure for the transport of an aqueous bioliquid into an electrode area in order to carry out electrochemical analyses, in which the layer electrode has an electrically conductive surface structure and is arranged under or on a hydrophilic cover layer, wherein an increased water affinity of the cover layer improves functionality of capillary transport paths.
27. An insulating substrate as claimed in claim 26, for use in an analytical test strip.

