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INVASIVE MULTI-ELECTRODE ELECTROCHEMICAL SENSOR

Abstract

An invasive multi-electrode electrochemical sensor includes a substrate, plural wire electrodes and an axle. Each wire electrode includes an electrically conductive core and an insulating sheath that substantially covers, but exposes a proximal end and a distal end of, the electrically conductive core. Each wire electrode has a substrate section provided on the substrate and an invasion section extending outward from an edge of the substrate. The proximal end and distal end of the electrically conductive core of each wire electrode are located at the substrate section and invasion section of the wire electrode respectively. The invasion sections are wound around the axle spirally. The axle has a terminal end protruding beyond the distal ends of the invasion sections.

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Background/Summary

BACKGROUND OF THE INVENTION

1. Technical Field

[0001] The present invention relates to an invasive electrochemical sensor.

2. Description of Related Art

[0002] Existing electrochemical sensors can be used in fluid detection and in that case are typically structured as test strips. An electrochemical test strip generally has a detection area to which a to-be-tested solution can be transferred in the form of a liquid drop or that can be immersed in the to-be-tested solution. However, existing electrochemical test strips cannot be used to perform invasive detection. When it comes to the continuous monitoring of biological/physiological parameters, an invasive detector is often more suitable than an existing non-invasive electrochemical test strip.

[0003] Therefore, it is an issue worth consideration by those working in the field to which the present invention pertains to provide an invasive electrochemical sensor that also has a relatively low production cost.

BRIEF SUMMARY OF THE INVENTION

[0004] The primary objective of the present invention is to provide a relatively low-cost invasive electrochemical sensor.

[0005] To achieve the above and other objectives, the present invention provides an invasive multi-electrode electrochemical sensor (hereinafter also referred to as the electrochemical sensor for short) that includes a substrate, a plurality of wire electrodes and an axle. Each wire electrode includes an electrically conductive core and an insulating sheath. Each insulating sheath substantially covers the corresponding electrically conductive core while leaving a proximal end and a distal end of the corresponding electrically conductive core exposed. Each wire electrode has a substrate section and an invasion section. The proximal end and the distal end of the electrically conductive core of each wire electrode are located at the substrate section and the invasion section of the wire electrode respectively. The substrate section of each wire electrode is located on the substrate. The invasion section of each wire electrode extends outward from an edge of the substrate, and the invasion sections of the wire electrodes are wound around the axle spirally. The axle has a terminal end protruding beyond the distal ends of the invasion sections.

[0006] The present invention is so designed that the mechanicla strength of the invasion sections are enhanced by the axle. The invasion sections can be inserted into a detection target more easily, thereby increasing the application scenarios of the electrochemical sensor of the present invention.

Description

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

[0007] FIG. **1** is a perspective view of the first embodiment of the present invention.

[0008] FIG. **2** is an exploded view of the first embodiment of the present invention.

[0009] FIG. **3** is another exploded view of the first embodiment of the present invention, showing in particular how each electrically conductive core is electrically connected to the corresponding gold finger by soldering.

[0010] FIG. **4** is a partial enlarged view of the first embodiment of the present invention.

[0011] FIG. **5** shows sectional views of the distal ends of four electrically conductive cores.

[0012] FIG. **6** shows a sectional view of the distal end of an electrically conductive core in another embodiment.

[0013] FIG. **7** shows the electrochemical sensor of the present invention used in combination with an electrochemical sensing relay.
[0014] FIG. **8** shows a way of implementing the invasion sections of the wire electrodes in the present invention.
[0015] FIG. **9** shows another way of implementing the invasion sections of the wire electrodes in the present invention.
[0016] FIG. **10** shows the reproducibility test results of the electrochemical sensor of the present invention.
[0017] FIG. **11** shows cyclic voltammogram traces obtained by immersing the electrochemical sensor of the present invention in aqueous hydrogen peroxide solutions of different concentrations.
[0018] FIG. **12** shows a current-time graph obtained by detecting an aqueous hydrogen peroxide solution with the electrochemical sensor of the present invention by amperometry.
[0019] FIG. **13** shows a linear regression line fitted to the data points on a current-concentration graph obtained by detecting an aqueous hydrogen peroxide solution with the electrochemical sensor of the present invention by amperometry.
[0020] FIG. **14** shows still another way of implementing the invasion sections of the wire electrodes in the present invention.
[0021] FIG. **15** shows yet another way of implementing the invasion sections of the wire electrodes in the present invention.
[0022] FIG. **16** shows another way of implementing the electrochemical sensor of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0023] Please refer to FIG. **1** to FIG. **4** for the first embodiment of the present invention. The electrochemical sensor of the invention can be used to perform invasive detection on a host, and the host may be a human, an animal, or a plant. The electrochemical sensor can detect whether the host's body contains a target analyte, the concentration of the target analyte, and/or other to-be-detected values. The target analyte may be, for example but not limited to, glycated hemoglobin, blood sugar, a heavy metal, a nitrate, a nitrite, an allergen, formaldehyde, dissolved oxygen, uric acid, dopamine, ascorbic acid, potassium ferricyanide, acetaminophen, a halogen ion, a sulfur ion, an aqueous hydrogen peroxide solution, an arsenic (III) ion, a lead ion, a zinc ion, a chromium ion, phenols, an amino acid, or another similar compound. The to-be-detected value may be a physical parameter such as, but not limited to, a pH value or conductivity. The electrochemical sensor of the invention may also be implemented in such a way that it can be used in a non-invasive detection environment, e.g., to test a water sample taken from the environment or other aqueous solutions. In this embodiment, the electrochemical sensor includes a substrate **10**, three wire electrodes **20**, three gold fingers **30**, and a cover plate **40**.
[0024] The material of the substrate **10** may be, but is not limited to, polypropylene, polyethylene terephthalate, polyimide, polyethylene, polyurethane, or polycarbonate.
[0025] Each wire electrode **20** includes an electrically conductive core **21** and an insulating sheath **22** (see FIG. **5**). Each insulating sheath **22** substantially covers the corresponding electrically conductive core **21** while exposing a proximal end **211** and a distal end **212**, or free end, of the corresponding electrically conductive core **21**. In addition, each wire electrode **20** has a substrate section **23** and an invasion section **24**. The proximal end **211** and the distal end **212** of the electrically conductive core **21** of each wire electrode **20** are located at the substrate section **23** and the invasion section **24** of the wire electrode **20** respectively. The substrate section **23** of each wire electrode **20** is provided on the substrate **10**. The invasion section **24** of each wire electrode **20** extends outward from an edge of the substrate **10**, and the length for which each invasion section **24** extends outward from the edge may be greater than 10 mm. In this embodiment, the invasion sections **24** of the three wire electrodes **20** are wound around one another in a spiral manner. Winding the invasion sections **24** together in a spiral manner is advantageous in that the accuracy

of detection is enhanced by the short distance and low electrical resistance among the electrodes. The electrically conductive core **21** in the invasion section **24** of at least one of the wire electrodes **20** is different in material from the electrically conductive cores **21** of the other wire electrodes **20**. In this embodiment, for example, the electrically conductive cores of the three wire electrodes serve separately as a working electrode, an auxiliary electrode, and a pseudo electrode/reference electrode. Depending on the analyte to be detected, the electrically conductive cores of the three wire electrodes may by, but are not limited to, the materials listed in Table 1. The insulating sheaths **22**, on the other hand, are made of an insulating material to prevent the electrically conductive cores of different wire electrodes from direct electrical connection with one another and hence from forming a short circuit.

TABLE-US-00001 TABLE 1 Pseudo electrode/ Working Auxiliary reference electrode electrode electrode Analyte Carbon Carbon Silver Uric acid, dopamine, ascorbic acid, potassium ferricyanide, text missing or illegible when filed Carbon Platinum Silver Uric acid, dopamine, ascorbic acid, potassium ferricyanide, acetaminophen Silver Carbon Silver chloride Halogen ions Nickle Platinum Carbon Sulfur ion Platinum Platinum Silver chloride Aqueous hydrogen peroxide solution Platinum Gold Silver chloride Dissolved oxygen Gold Gold Silver Arsenic(III) ion, lead ion, zinc ion, chromium ion Copper Carbon Silver Phenols, amino acids Bismuth Carbon Silver Arsenic(III) ion, lead ion, zinc ion, chromium ion text missing or illegible when filed indicates data missing or illegible when filed

[0026] The wire electrodes can be viewed as metallic wire ultramicroelectrodes (MWUME) when the diameter Ø of each electrically conductive core **21** is less than or equal to 25 μm, as metallic wire microelectrodes (MWME) when the diameter of each electrically conductive core **21** is greater than 25 μm and less than 1000 μm, or as metallic wire electrodes (MWE) when the diameter Ø of each electrically conductive core **21** is greater than or equal to 1000 μm.

[0027] The gold fingers **30** are provided on the substrate **10**. The electrically conductive core **21** in the substrate section **23** of each wire electrode **20** (e.g., the proximal end **211** of each electrically conductive core **21**) is electrically connected to the corresponding gold finger **30**. The electrical connection between each electrically conductive core **21** and the corresponding gold finger **30** may be formed by, for example but not limited to, soldering or application of electrically conductive tape. The gold fingers **30** may be printed on the substrate **10** by screen printing for example, and the material of the gold fingers **30** may be printed carbon paste or printed silver paste for example. The printed carbon paste or printed silver paste may further receive a surface treatment by, for example, being sputter-coated with a metal such as platinum, gold, copper, or silver. In this embodiment, the gold fingers **30** extend to an edge of the substrate **10**.

[0028] The cover plate **40** is provided on the substrate **10** such that the entire substrate sections **23** of the wire electrodes **20** are fixed between the cover plate **40** and the substrate **10**.

[0029] Referring to FIG. **5**, the distal end of each electrically conductive core **21** may receive a surface treatment so as to have the configuration of being flush with the corresponding end face of the corresponding insulating sheath **22**, or protruding from the corresponding end face of the corresponding insulating sheath **22**, or having an irregular surface, or being sunken into the corresponding end face of the corresponding insulating sheath **22**. One way of implementing the wire electrodes is to make the distal end **211** of each electrically conductive core **21** sunken into an end face **221** of the corresponding insulating sheath **22** while satisfying the relationship $Hw/Ø<50$, where Hw is the depth to which the distal end **211** is sunken into the end face **221**. This sunken configuration leaves room for subsequent chemical modification of the distal ends of the electrically conductive cores. For example, the recess formed at the aforesaid end face of each insulating sheath may be filled with an enzyme layer (not shown). Besides, referring to FIG. **6**, the distal end **211** of at least one of the electrically conductive cores **21** is made of a material different from the material of the other portion of the electrically conductive core **21**. For example, the distal end of this at least one electrically conductive core is made of carbon while the other portion of the

electrically conductive core is made of copper in order to adapt to different detection environments.
[0030] Referring to FIG. **7**, the electrochemical sensor according to the embodiment shown in FIG. **1** to FIG. **4** can be used in combination with an electrochemical sensing relay **1**. The electrochemical sensing relay **1** is configured to be electrically connected to each and every gold finger of the electrochemical sensor **2** and to relay the signals sensed by the electrochemical sensor to a remote receiving unit (e.g., a smartphone, a computer, or a cloud server) in order to perform further computation and/or display the computation result.
[0031] It should be pointed out that the number of the wire electrodes can be adjusted. For example, in the embodiment shown in FIG. **8**, the spirally wound invasion sections of the four wire electrodes **20** can be used to detect more analytes at the same time than when fewer wire electrodes are provided, and as shown in FIG. **9**, the invasion sections of six wire electrodes may be spirally wound around the invasion section of a central wire electrode **20***c* that extends along a straight line, i.e., with the invasion section of at least one wire electrode that extends along a straight line serving as an axis around which the invasion sections of the other wire electrodes are spirally wound.
[0032] Referring to FIG. **10**, a reproducibility test was conducted in which a plurality of electrochemical sensors as shown in FIG. **1** took turns being immersed in each of two different aqueous solutions for three times. The test results show that the detection result of each aqueous solution was highly reproducible with the electrochemical sensor of the present invention.
[0033] Referring to FIG. **11**, an electrochemical sensor with three wire electrodes was immersed in 0.1 M PBS (phosphate-buffered saline), 500 μM aqueous hydrogen peroxide (H.sub.2O.sub.2) solution, and 1000 μM aqueous hydrogen peroxide solution separately. The material of the major portions of the electrically conductive cores of the wire electrodes was carbon while the material of the distal ends of the electrically conductive cores was platinum. The test results show that the electrochemical sensor of the present invention was indeed capable of detecting different oxidation and reduction potentials in aqueous hydrogen peroxide solutions of different concentrations.
[0034] Referring to FIG. **12**, the applicant used amperometry to test the performance of the electrochemical sensor of the present invention in detecting aqueous hydrogen peroxide solutions. The electrochemical sensor used in the test had three wire electrodes, with the material of the major portions of the electrically conductive cores of the wire electrodes being carbon, and the material of the distal ends of the electrically conductive cores being platinum. The applicant used amperometry together with the working parameters in Table 2 to detect aqueous hydrogen peroxide solutions of different concentrations. More specifically, the concentration of an aqueous hydrogen peroxide solution was adjusted every 50 seconds. Each of the first ten adjustments involved increasing the concentration by 100 μM, and each of the last five adjustments involved increasing the concentration by 200 μM. The final concentration was 2000 μM. The oxidation voltage was fixed at 600 mV. The test results show that the electrochemical sensor of the invention was able to detect the changes in concentration of the aqueous hydrogen peroxide solution sensitively, and that the detected current values are highly linearly correlated to the changes in concentration (see FIG. **13**), indicating that the electrochemical sensor of the invention has high accuracy.
TABLE-US-00002 TABLE 2 Elapsed Concentration of aqueous hydrogen time (s) peroxide solution (μM) 0 0 50 100 100 200 150 300 200 400 250 500 300 600 350 700 400 800 450 900 500 1000 550 1200 600 1400 650 1600 700 1800 750 2000
[0035] The embodiment shown in FIG. **14** is similar to the embodiment in FIG. **9**, the major difference being that the electrochemical sensor in FIG. **14** includes an axle **50**, and that the central wire electrode **20***c* in FIG. **9** is replaced by the axle **50** while the invasion sections **24** of the other wire electrodes **20** are wound around the axle **50** spirally. A portion of the axle is configured to be fixed to the substrate (not shown). The portion of the axle **50** around which the invasion sections **24** are spirally wound extends along a straight line. The axle **50** has a terminal end **51**, and the terminal end **51** protrudes beyond the distal ends **212** of the invasion sections **24**. In this embodiment, the terminal end **51** is solid and pointed and serves to change the structural strength of the invasion

sections **24** so that the invasion sections **24** can be easily inserted into a detection target (e.g., soil or a plant) without the help of an additional placing device. The embodiment shown in FIG. **15** is similar to the embodiment in FIG. **14**, the major difference being that the terminal end **51** of the axle **50** in FIG. **15** is hollow and pointed and serves also to change the structural strength of the invasion sections **24**. The hollow terminal end **51** helps disturb the tissue into which it is inserted, so that the tissue can contact the distal ends **212** of the invasion sections **24** easily through capillary action for example, thereby facilitating detection. The axle **50** may be a non-metallic, non-conductor wire, whose material may be, for example, ceramic, silicon dioxide, plastic, acrylic, or a polymer such as polypropylene (PP), polycarbonate (PC), or polyethylene (PE).

[0036] Referring to FIG. **16**, the embodiment shown therein is similar to the embodiment in FIG. **1**. The portion of each gold finger **30** that is not covered by the cover plate **40** is defined as an exposed section **31**. The exposed sections **31** are parallel to one another and extend in an insertion direction L. The gold fingers **30** are provided on a working surface **11** of the substrate **10**. The invasion section **24** of each wire electrode has a front-side portion **241**, and the distal end **212** of the electrically conductive core of each wire electrode is located at the front-side portion of the invasion section of the wire electrode. The front-side portions **241** extend in a direction that is not parallel to the insertion direction L or the working surface **11**. In this embodiment, the front-side portions **241** are perpendicular to the working surface **11**. Another possible way of implementing the electrochemical sensor is to provide the invasion sections **24** with flexibility so that a user can adjust the extending direction of the front-side portions **241** according to the geometric properties of the detection target.

Claims

1. An invasive multi-electrode electrochemical sensor, comprising: a substrate; a plurality of wire electrodes, wherein each said wire electrode comprises an electrically conductive core and an insulating sheath, each said insulating sheath substantially covers a corresponding one of the electrically conductive cores while exposing a proximal end and a distal end of the corresponding one of the electrically conductive cores, each said wire electrode has a substrate section and an invasion section, the proximal end and the distal end of the electrically conductive core of each said wire electrode are located at the substrate section and the invasion section of the each said wire electrode respectively, the substrate section of each said wire electrode is provided on the substrate, the invasion section of each said wire electrode extends outward from an edge of the substrate; and an axle, the invasion sections of the wire electrodes being wound around the axle spirally, the axle having a terminal end protruding beyond the distal ends of the invasion sections.

2. The invasive multi-electrode electrochemical sensor of claim 1, wherein the terminal end is pointed.

3. The invasive multi-electrode electrochemical sensor of claim 1, wherein the terminal end is solid.

4. The invasive multi-electrode electrochemical sensor of claim 1, wherein the terminal end is hollow.

5. The invasive multi-electrode electrochemical sensor of claim 1, further comprising a cover plate provided on the substrate such that the substrate sections of the wire electrodes are at least partially fixed between the cover plate and the substrate.

6. The invasive multi-electrode electrochemical sensor of claim 1, wherein a length for which each of the invasion sections extends outward from the edge is greater than 10 mm.