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(54) Title: MICROELECTRODE AND METHOD OF FABRICATION THEREOF

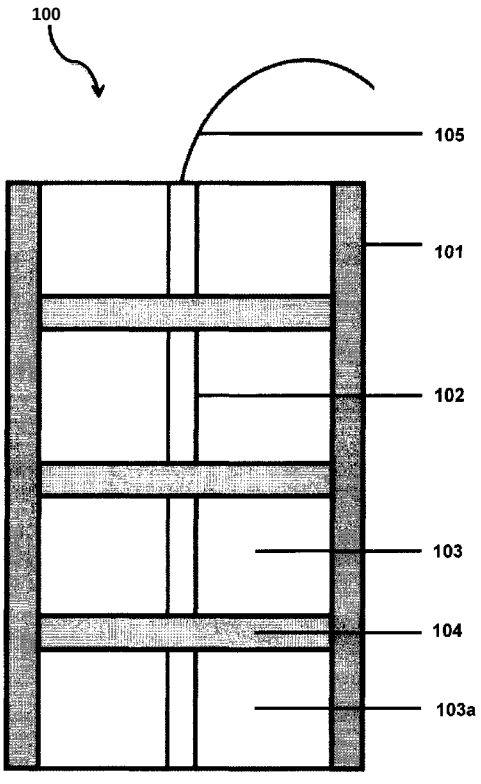


FIGURE 2

(57) Abstract: Microelectrode (100) for use in electrochemical measurements such as amperometric and voltammetric measurement, and analytical sensing, wherein the microelectrode (100) comprises a hollow container (101); a metal wire (102) with micro-diameter for conducting electricity positioned in the center of the hollow container (101) and parallel to the length of the hollow container (101); at least one layer of soluble material (103) for temporarily sealing the metal wire (102); at least one layer of sealing material (104) for securing the position of the metal wire (102) within the hollow container (101); and a wire connector (105) connected to the metal wire (102) at one end of the microelectrode (100) for connecting the microelectrode (100) to external devices. Further, it is detailed herein modifications made to the microelectrode (100), converting the microelectrode into either a micro-wire type microelectrode or a micro-disk type microelectrode. Also, it is presented herein a method for fabricating the aforementioned microelectrode (100).

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MICROELECTRODE AND METHOD OF FABRICATION THEREOF

TECHNICAL FIELD OF THE INVENTION

The present invention relates generally to a microelectrode, and more particularly, to a microelectrode that can be converted into either a micro-wire type microelectrode or a micro-disk type microelectrode.

BACKGROUND OF THE INVENTION

Microelectrode is vastly utilized in the field of electrochemical measurements as it offers numerous advantages such as lower interfacial capacitance, fast mass transport, reduced capacitance, and low current. There are a few kinds of microelectrodes, where disk, band, and ring types being the most typical ones. Out of these three kinds, the disk type microelectrode is the most commonly used microelectrode in analytical and molecular electrochemistry. This is due to a couple of reasons, first one being its simple fabrication process, and second one being the feasibility in controlled surface cleaning.

Nevertheless, the disk type microelectrode poses several problematic issues. One of the main issues is that the current density is not uniform across the surface of the disk, where it is greater at the edge.

Second flaw relates to the method of fabrication of the disk type microelectrode. In general, the method includes insulating metal wire with glass, epoxy, or polymer, and then polishing or electrochemically etching one end. Although the making of disk type microelectrode involves simple steps, however the steps normally include repetitive heavy sawing or grinding of the whole microelectrode in order to expose fresh surface. Often, this repetitive heavy activity causes structural non-idealities of microelectrode. We now refer to Figure 1, which illustrates the most common structural non-idealities of disk type microelectrode. The microelectrode (A) is an elliptical microelectrode caused by the sealing of metal wire at an inappropriate angle within the insulator. The microelectrode (B)

is a protruding and irregular cylindrical microelectrode caused by insufficient mechanical polishing. The defect as shown in microelectrode (B) may also due to the reason that the material used to make the wire metal is softer than the insulator. Further, this particular reason is also capable of causing a recessed microelectrode as illustrated as microelectrode (C) and microelectrode (D). In addition, the flaw of microelectrode (C) and the microelectrode (D) also occurred when the metal wire is polished to a lower level than the surrounding insulator. The microelectrode (E) is a microelectrode with imperfect seal caused by large difference in thermal coefficient of expansion between the metal wire and the insulator. The microelectrode (E) may also be caused by failing to adequately clean the metal wire and the insulator. Appended below is a table indicating the changes in the resistance and capacitance due to these structural non-idealities.

TABLE 1: Effect of non-ideal microelectrode geometry on resistance and capacitance

Geometry	*R* (Ω)	C (μF cm^{-2})	Comment
Elliptical	→	↑	Capacitance increases but resistance decreases relative to ideal due to increased electrode area.
Protruding	↓	↑	Capacitance increases relative to ideal due to increased effective electrode area.
Microcavity Lagooned	↑	→	Resistance typically increases significantly relative to ideal due to difficulties of ion mass transport within pore.
Recessed	↑	→	Resistance may increase moderately but diagnostic abilities of chronoamperometry are limited.
Imperfect seal	↑	↑	Resistance and capacitance both very significantly larger than ideal.

Apart from the above, the repetitive heaving sawing or grinding also causes other unfavorable effect such as rough surface. Further, this particular heavy activity also increases the risk of surface contamination, which dramatically decreases the workability and effectiveness of the microelectrode in producing quality measurements.

In view of the above, it therefore has become the aim of the present invention to solve all of the aforementioned technical issues by providing a novel microelectrode with ideal or near ideal structure and a fabrication method thereof that do not require the heavy sawing or grinding; simplify the polishing process; and decrease or eliminate the risk of contaminating the microelectrode, specifically the surface of the microelectrode.

SUMMARY OF THE INVENTION

The first aspect of the present invention is directed to a microelectrode that can be used in electrochemical measurements such as, but not limited to, amperometric and voltammetric measurements. It is also proposed herein that the microelectrode can be utilized in analytical sensing such as, but not limited to, being the sensor for monitoring hydrogen, nitrogen oxides, sulfur dioxide, and ammonia.

The said microelectrode comprises a hollow container; a metal wire with micro-diameter for conducting electricity positioned in the center of the hollow container, and paralleled to the length of the hollow container; at least one layer of soluble material for temporarily sealing the metal wire; at least one layer of sealing material for securing the position of the metal wire within the hollow container; and a wire connector connected to the metal wire at one end of the microelectrode for connecting the microelectrode to external devices. If the microelectrode has more than one layer of soluble material and more than one layer of sealing material, the layers of soluble material and the layers of sealing material are arranged in an alternate manner within the hollow container.

In an embodiment of the present invention, the hollow container can be made of any non-conductive inert material, in this instance for examples, glass and plastic. The hollow container can be of any geometry shape, in this instance for example, cylindrical geometry shape.

In another embodiment of the present invention, the metal wire can be made of any highly conductive inert metal, preferably the said material is selected from the group of noble metal, in this instance for examples, gold, platinum, copper, and silver.

In a further embodiment of the present invention, the soluble material is polymer that is either malleable near ambient temperature or soluble in organic non-polar solvent, in this instance for examples, wax and plastic. Also, it is very important that the selected soluble material will not cause surface contamination to the metal wire and the sealing material.

In another embodiment of the present invention, the sealing material can be of any non-conductive inert thermosetting polymer, in this instance for example, epoxy resin. Also, it is very important that the selected sealing material will not cause surface contamination to the metal wire.

The aforesaid microelectrode is presented in its raw form. In order words, preparation steps need to be taken and executed on the microelectrode before it can be used. The said preparation steps allow the microelectrode to be converted into either a micro-wire type microelectrode or a micro-disk type microelectrode.

For the micro-wire type microelectrode, firstly, the exposed soluble material at one end of the microelectrode that is without the wire connector is removed. This removal step causes the sealed metal wire and the covered sealing material to be exposed. It also causes the hollow container of the microelectrode to have a protruded part, which is easily removed by sawing or grinding. In other words, heavy sawing or grinding is not necessary as the sealing material, and the metal

wire are not subjected to this particular activity. As a result, the microelectrode is free from defects such as rough surface of sealing material, and imperfect seal between the metal wire and the sealing material. Moreover, contamination due to the activity of sawing or grinding is greatly reduced, if not eliminated. Optionally, the now exposed metal wire can be either left uncut or cut according to desired length.

For the micro-disk type microelectrode, firstly, the exposed soluble material at one end of the microelectrode that is without the wire connector is removed. This removal step causes the sealed metal wire and the covered sealing material to be exposed. It also causes the hollow container of the microelectrode to have a protruded part, which is easily removed by sawing or grinding. In other words, heavy sawing or grinding is not necessary as the sealing material, and the metal wire are not subjected to this particular activity. As a result, the microelectrode is free from defects such as rough surface of sealing material, and imperfect seal between the metal wire and the sealing material. Moreover, contamination due to the activity of sawing or grinding is greatly reduced, if not eliminated. The now exposed metal wire is completed cut off. Thereafter, the exposed sealing material is polished to produce a mirror-like finish. The polishing is made simple as the sealing material is not subjected to the activity of sawing or grinding.

The second aspect of the present invention is directed to a method for fabricating the microelectrode as described above. Initially, the metal wire needs to be positioned in the center of the hollow container, and parallel to the length of the hollow container. Subsequently, a layer of soluble material is deposited and cured. Thereafter, a layer of sealing material is deposited and cured. Depending on the user's application, the aforementioned layering steps can be repeated until a desired number of alternating layers of soluble material and sealing material is achieved. Lastly, the wire connector is connected to the metal wire at one end of the microelectrode.

In one embodiment of the present invention, the curing is vacuum-aided curing carried out at mild temperature. It is performed to harden the layers of soluble

material and sealing material, in addition to avoid the formation of permanent bubbles within the layers.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 illustrates the most common structural non-idealities of microelectrode;

Figure 2 illustrates the microelectrode of the present invention;

Figure 3 illustrates the micro-wire type microelectrode of the present invention; and

Figure 4 illustrates the micro-disk type microelectrode of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

The above mentioned and other features and objects of this invention will become more apparent and better understood by reference to the following detailed description. It should be understood that the detailed description made known below is not intended to be exhaustive or limit the invention to the precise disclosed form as the invention may assume various alternative forms. On the contrary, the detailed description covers all the relevant modifications and alterations made to the present invention, unless the claims expressly state otherwise.

The first core aspect of the present invention relates to a microelectrode (100) which can be used in performing various kinds of electrochemical measurement such as, but not limited to, amperometric and voltammetric measurements. The said microelectrode (100) also can be used as sensing media in analytical sensing such as, but not limited to, being the sensing media for monitoring hydrogen, nitrogen oxides, sulfur dioxide, and ammonia.

The microelectrode (100) as introduced herein is an improved version of conventional microelectrode, and has a number of advantages over the conventional type. Specifically, the present microelectrode (100) eliminates the needs for heavy sawing or grinding; simplifies the polishing process; and drastically reduces the risk of the microelectrode (100) or parts of the microelectrode (100) from being contaminated, if not eliminates.

Referring now to Figure 2, the said microelectrode (100) comprises a hollow container (101); a metal wire (102) with micro-diameter for conducting electricity positioned in the center of the hollow container (101), and paralleled to the length of the hollow container (101); at least one layer of soluble material (103) for temporarily sealing the metal wire (102) from exposure and being contaminated; at least one layer of sealing material (104) for securing the position of the metal wire (102) within the hollow container (101); and a wire connector (105) connected to the metal wire (102) at one end of the microelectrode (100) for connecting the microelectrode (100) to an external device. The layer of soluble material (103) that is exposed to the environment is known as exposed soluble material (103a).

In accordance with the above statement, the said microelectrode may comprise more than one layer of soluble material (103) and more than one layer of sealing material (104). In this circumstance, the layers of soluble material (103) and the layers of sealing material (104) are arranged in an alternate manner within the hollow container (101), as illustrated in Figure 2.

The hollow container (101) is made of non-conductive inert material. Preferably, it (101) is made of non-conductive inert material that is transparent. For examples, but not limited to, the hollow container (101) can be made of glass or plastic. Further, the hollow container (101) can be of any geometry shape. Preferably, it (101) is of, but not limited to, cylindrical geometry shape.

The metal wire (102) of the present invention is made of highly conductive inert metal. Preferably, the highly conductive inert material is selected from the group

of noble metal. For examples, but not limited to, gold, platinum, copper and silver are the preferred choice to be used as the metal wire (102) of the present invention. As previously mentioned, the diameter of the metal wire (102) is in micro-measurement. Determination of the exact diameter of the metal wire (102) to be used in the present invention is solely based on the user's application.

The soluble material (103) in this case can be of any polymer that is either malleable near ambient temperature or soluble in organic non-polar solvent. Also, it is very important that the chosen polymer does not cause surface contamination to the metal wire (102) and the sealing material (104). For examples, but not limited to, wax and plastic with low melting point are good polymer that can be used as the soluble material (103) for temporarily sealing the metal wire (102).

As for the sealing material (104), it (104) can be of any thermosetting polymer that possesses the characteristics of non-conductive and inert. Similarly to the soluble material (103), it (104) should not cause surface contamination to the metal wire (102). In the present invention, epoxy resin is a preferred polymer to be used as the sealing material (104). However, it should be noted that the sealing material (104) is not limited to epoxy resin only.

The above-mentioned microelectrode (100) is described and presented in its raw form. In other words, it (100) is in its pre-processed form and certain preparation methods needs to be performed on the microelectrode (100) before it (100) can be used. Two preparation methods will be described hereunder. Each method converts the microelectrode (100) in its raw form into different types of microelectrode (100), specifically, a micro-wire type microelectrode and a micro¬disk type microelectrode, as depicted in Figure 3 and Figure 4, respectively.

We now refer to Figure 2 and Figure 3. For the micro-wire type microelectrode, the preparation method, firstly, comprises a step of removing the exposed soluble material (103a) at one end of the microelectrode (100) that is without the wire connector (105). This first step causes the sealed metal wire (102) and the

covered sealing material (104) to be exposed resulting in an exposed metal wire (102a) and an exposed sealing material (104a). In the instance wax is used as the soluble material (103), the wax can be easily removed by subjecting it to minimal heat in order to effectively and completely remove the same. In addition, this first step also causes the hollow container (101) of the microelectrode (100) to have a protruded part, which is compulsory to be removed via an easy and simple process of sawing or grinding. In other words, heavy sawing or grinding is not necessary as the sealing material (104), and the metal wire (102) are not subjected to this particular heavy activity. As a result, the micro-wire type microelectrode is free from defects such as rough surface of exposed sealing material (104a), and imperfect seal between the metal wire (102) and the sealing material (104). Moreover, contamination due to the activity of sawing or grinding is greatly reduced, if not eliminated. Optionally, the now exposed metal wire (102a) can be either left uncut or cut according to desired length. The final form of micro-wire type microelectrode is represented in Figure 3.

We now refer to Figure 2, Figure 3, and Figure 4. For the micro-disk type microelectrode, the preparation method, firstly, comprises a step of removing the exposed soluble material (103a) at one end of the microelectrode (100) that is without the wire connector (105). This first step causes the sealed metal wire (102) and covered sealing material (104) to be exposed resulting in an exposed metal wire (102a) and an exposed sealing material (104a). In the instance wax is used as the soluble material (103), the wax can be easily removed by subjecting it to minimal heat in order to effectively and completely remove the same. In addition, this first step also causes the hollow container (101) of the microelectrode (100) to have a protruded part, which is compulsory to be removed via an easy and simple process of sawing or grinding. In other words, heavy sawing or grinding is not necessary as the sealing material (104), and the metal wire (102) are not subjected to this particular heavy activity. As a result, the micro-disk type microelectrode is free from defects such as rough surface of exposed sealing material (104a), and imperfect seal between the metal wire (102) and the sealing material (104). Moreover, contamination due to the activity of sawing or grinding is greatly reduced, if not eliminated. The now exposed metal

wire (102a) is completed cut off. Thereafter, the exposed sealing material (104a) is polished to produce a mirror-like finish, which can be easily achieved by employing polishing disks, micro-cloth, or polishing paste. The polishing is made simple as the sealing material (104) is not subjected to the activity of sawing or grinding. The final form of micro-disk type microelectrode is represented in Figure 4.

The second core aspect of the present invention relates to a method for fabricating the microelectrode (100) as introduced above. The method comprises the first step of positioning and suspending the metal wire (102) in the center of the hollow container (101) as well as parallel to the length of the hollow container (101). Thereafter, a layer of soluble material (103) is deposited and cured within the hollow container (101). Once the layer of soluble material (103) is cured, a layer of sealing material (104) is deposited and cured within the hollow container (101) and on top of the previously cured layer of soluble material (103). The curing process is preferably a vacuum-aided curing process carried out at mild temperature, which is beneficial in terms of effectiveness in hardening the layers of soluble material (103) and sealing material (104), as well as preventing the formation of permanent bubbles within the layers, especially the layer or layers of sealing material (104). These steps of deposition and curing of layers of soluble material (103) and sealing material (104) can be optionally repeated depending on the user's application or until a desired number of alternating layers of soluble material (103) and sealing material (104) is achieved. Ultimately, a wire connector (105) is connected to the metal wire (102) at one end of the microelectrode (100).

CLAIMS

1) A microelectrode (100) for electrochemical measurements and analytical sensing comprises:

a) a hollow container (101);

b) a metal wire (102) with micro-diameter for conducting electricity positioned in the center of the hollow container (101), and paralleled to the length of the hollow container (101);

c) at least one layer of soluble material (103) for temporarily sealing the metal wire (102);

d) at least one layer of sealing material (104) for securing the position of the metal wire (102) within the hollow container (101); and

e) a wire connector (105) connected to the metal wire (102) at one end of the microelectrode (100) for connecting the microelectrode (100) to external devices;

wherein in the event that the microelectrode (100) has more than one layer of soluble material (103), and more than one layer of sealing material (104), the layers of soluble material (103) and the layers of sealing material (104) are arranged in an alternate manner within the hollow container (101);

wherein the soluble material (103) does not cause surface contamination to the metal wire (102) and the sealing material (104);

wherein the sealing material (104) does not cause surface contamination to the metal wire (102).

2) A microelectrode (100) in accordance with Claim 1, wherein the hollow container (101) is made of non-conductive inert material.

3) A microelectrode (100) in accordance with Claim 1, wherein the hollow container (101) is of any geometry shape.

4) A microelectrode (100) in accordance with Claim 1, wherein the metal wire (102) is made of highly conductive inert metal.

5) A microelectrode (100) in accordance with Claim 1, wherein the soluble material (103) is polymer that is malleable near ambient temperature or soluble in organic non-polar solvent.

6) A microelectrode (100) in accordance with Claim 1, wherein the sealing material (104) is non-conductive inert thermosetting polymer.

7) A preparation method for converting the microelectrode (100) of Claim 1 into and to be used as a micro-wire type microelectrode comprises the steps of:

a) removing an exposed soluble material (103a) at one end of the microelectrode (100) that is without the wire connector (105), to expose the metal wire (102) and the sealing material (104) resulting in an exposed metal wire (102a) and an exposed sealing material (104a);

b) removing the protruded part of the hollow container (101) caused by the step a); and

c) optionally, cutting the exposed metal wire (102a) to desired length.

8) A preparation method for converting the microelectrode (100) of Claim 1 into and to be used as micro-disk type microelectrode comprises the steps of:

a) removing an exposed soluble material (103a) at one end of the microelectrode (100) that is without the wire connector (105), to expose the metal wire (102) and the sealing material (104) resulting in an exposed metal wire (102a) and an exposed sealing material (104a);

b) removing the protruded part of the hollow container (101) caused by the step a);

c) completely cutting off the exposed metal wire (102a); and

d) polishing the exposed sealing material (104a) to produce a mirror-like finish.

9) A method for fabricating the microelectrode (100) of Claim 1 comprises the following steps:

a) positioning the metal wire (102) in the center of the hollow container (101), parallel to the length of the hollow container (101);

b) depositing the layer of soluble material (103) into the hollow container (101), and curing the layer of soluble material (103);

c) depositing the layer of sealing material (104) into the hollow container (101), and curing the layer of sealing material (104);

d) optionally, repeating steps b) to c) until a desired number of alternating layers of soluble material (103) and sealing material (104) is achieved; and

e) connecting the wire connector (105) to the metal wire (102) at one end of the microelectrode (100).

10) A method in accordance with Claim 8, wherein the curing is vacuum-aided curing carried out at mild temperature to harden the layers of soluble material (103) and sealing material (104), and preventing the formation of permanent bubbles within the layers (103) (104).

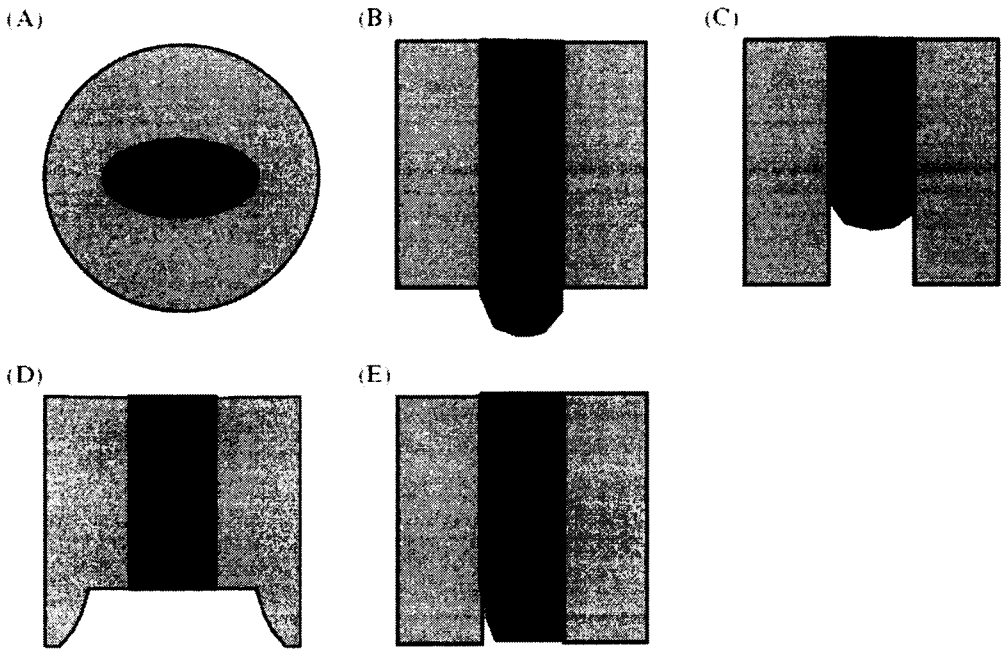


FIGURE 1

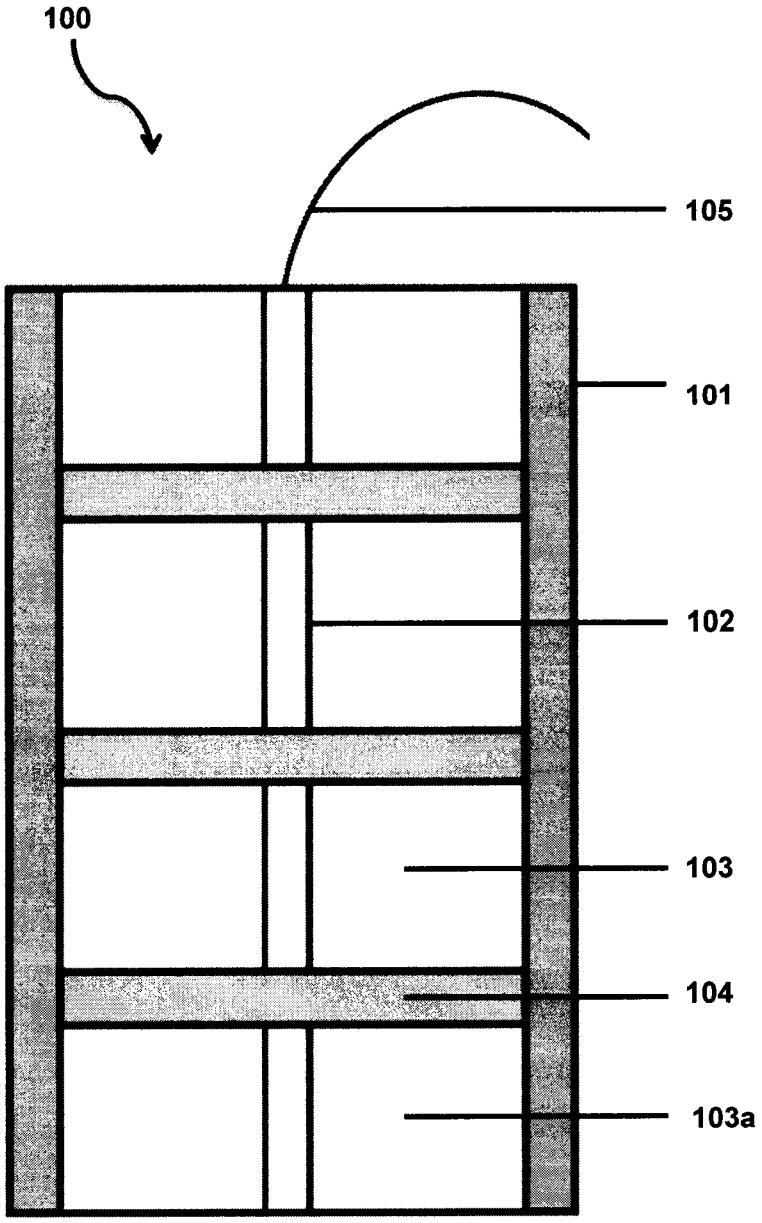


FIGURE 2

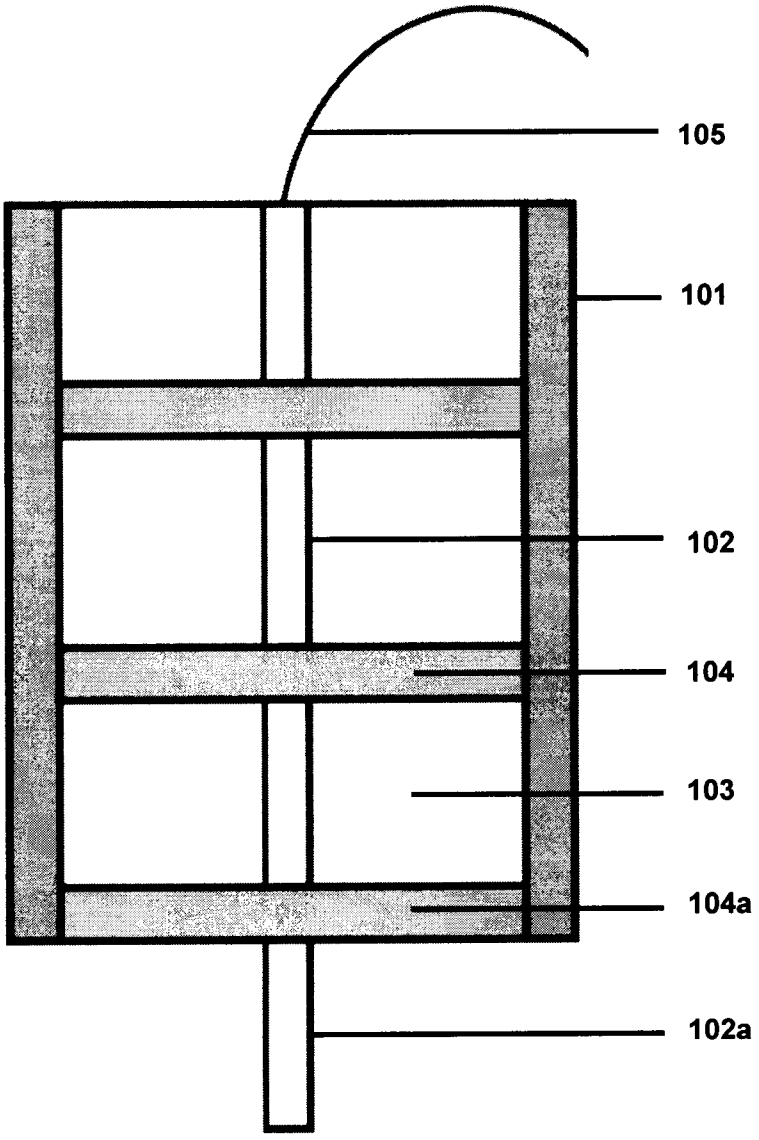


FIGURE 3

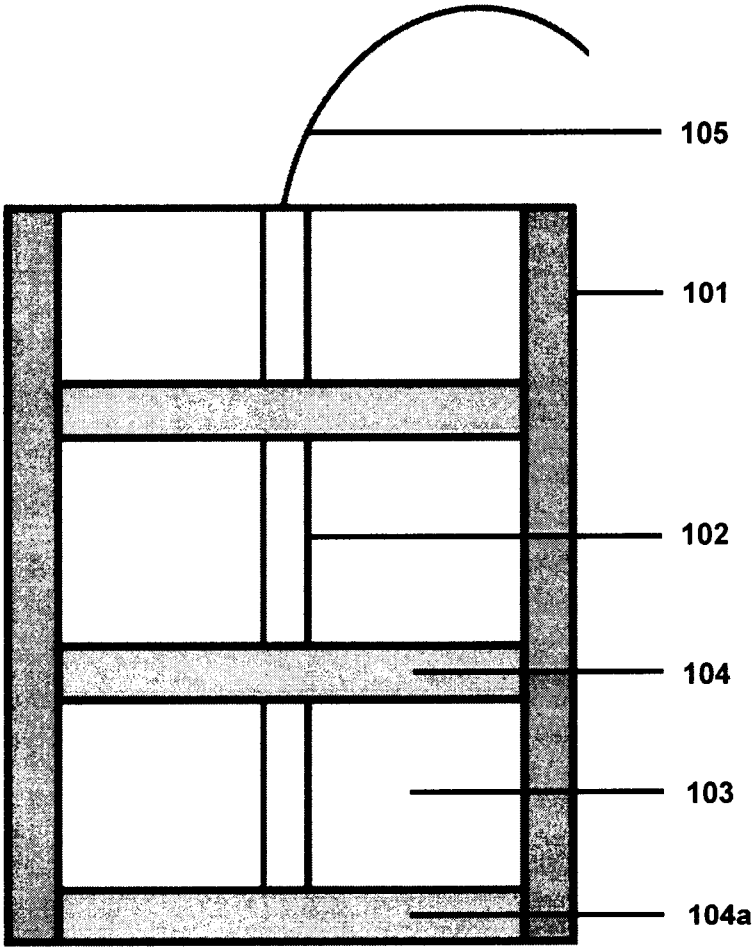


FIGURE 4

INTERNATIONAL SEARCH REPORT

International application No
PCT/MY2014/0OQ061

A. CLASSIFICATION OF SUBJECT MATTER
INV. G01N27/30
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
G01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal , WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 833 825 A (OTTEN SI EF [US] ET AL) 10 November 1998 (1998-11-10) the whole document	1-6
X	US 5 580 431 A (ISAACS HUGH S [US] ET AL) 3 December 1996 (1996-12-03) the whole document	1-4
A	DE 198 42 735 AI (VOS TORSTEN [DE] ; THELEMANN TORSTEN [DE]) 23 March 2000 (2000-03-23) the whole document	1-10
A	DE 35 41 806 CI (MANNESMANN AG; K0LB GUSTAV) 19 February 1987 (1987-02-19) the whole document	1-10

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[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
20 August 2014	28/08/2014

Name and mailing address of the ISA/	Authorized officer
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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 196 52 596 Al (HERAEUS ELECTRO NITE INT [BE]) 2 July 1998 (1998-07-02) the whole document -----	1-10
A	US 5 792 329 A (CURE OMER PAUL IVO [BE] ET AL) 11 August 1998 (1998-08-11) the whole document -----	1-10

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International application No
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