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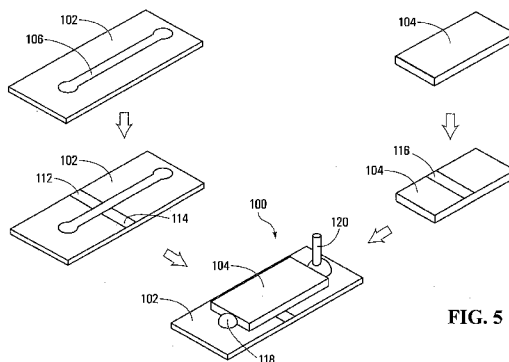


FIG. 5

(57) Abstract: An electronic device has two bonded members. The first member comprises a substrate defining a fluid channel for receiving a fluid, and first and second electrodes formed on the substrate and separated by the fluid channel. The second member comprises a substrate and a semiconductor formed on the substrate. The semiconductor has a modulatable electrical property. The members are bonded with the semiconductor bridging the electrodes across the fluid channel and being proximate to the fluid channel, thus allowing the electrical property of the semiconductor be modulated by the fluid received in the fluid channel. The electrodes and the semiconductor may consist essentially of carbon nanotubes. The substrates may comprise a laminable material. Each substrate may consist essentially of a polymeric material.

ELECTRONIC DEVICE WITH CHANNEL, ELECTRODES AND SEMICONDUCTOR FORMED ON RESPECTIVE BONDED SUBSTRATES

CROSS-REFERENCE TO RELATED APPLICATION

[0001] This application claims the benefit of U.S. provisional application No. 61/136,626, filed September 19, 2008, the entire contents of which are incorporated herein by reference.

FIELD OF THE INVENTION

[0002] The present invention relates generally to electronic devices, and more specifically to electronic devices having channel, electrodes and semiconductor and processes for forming and methods of operating such devices. The electronic devices include field-effect transistors, sensors, memory devices, charge storage devices such as batteries, or the like.

BACKGROUND OF THE INVENTION

[0003] Electronic devices such as field-effect transistors (FET) and biosensors are typically formed in a series of fabrication stages by growing or depositing on a substrate various materials and structures. Various components in the device may be shaped or patterned by masking or etching. For instance, printed electronics based field effect transistors (FETs), circuits, biosensors, and related devices are typically fabricated by sequentially forming the substrate, gate electrode, dielectric, source/drain electrodes, semiconductor, and encapsulation.

[0004] For example, **FIG. 1** shows a schematic of a single-walled carbon nanotube FET (SWCNT-FET) disclosed in Zhou et al., "Supported lipid bilayer/carbon nanotube hydrids," *Nature nanotechnology*, 2007, vol. 2, pp. 185-190 (referred to herein as "Zhou"), and in Rosenblatt et al, "High performance

electrolyte gated carbon nanotube transistors,” *Nano Letters*, 2002, vol. 2, pp. 869-872 (referred herein as “Rosenblatt”). In the technique taught by Zhou and Rosenblatt, a degenerately doped silicon (Si) wafer is capped with a silicon dioxide (SiO₂) capping layer, which is thermally grown. Catalyst islands containing Fe(NO₃)₃•9H₂O, MoO₂(acac)₂, and alumina nanoparticles are formed on the capping layer (not shown). A poly(methyl methacrylate) (PMMA) layer (not shown) is formed and patterned on the catalyst islands as a lift-off mask using photolithography and etching. CNTs are then grown by chemical vapor deposition on the capping layer between the catalyst islands. Gold (Au) source and drain electrodes are next patterned over the catalyst islands using photolithography and a lift-off process, in which the catalyst and PMMA layer are lifted-off. The resulting FET structure is annealed at 600 °C for 45 minutes in an argon environment to improve the contact resistance between the CNTs and the electrodes. The FET structure is then placed inside a polydimethylsiloxane (PDMS) microfluidic channel. The channel contains a fluid which contains a liquid electrolyte. To isolate the electrodes from the electrolyte, the device was covered with an alumina layer except in the regions between the source and drain electrodes containing the nanotubes. This process involves a number of different materials and a sequence of processing steps. A similarly constructed CNT transistor is disclosed in Heller *et al.*, “Identifying the mechanism of biosensing with carbon nanotube transistors,” *Nano Letters*, 2008, vol. 8, pp. 591-595.

SUMMARY OF THE INVENTION

[0005] It is recognized that there is a need for an improved electronic device, an improved process for forming an electronic device, and an improved method of operating the device.

[0006] It is also recognized that it may be advantageous to provide electronic devices such as FETs with enhanced charge carrier mobilities and reduced operation voltages (e.g. < about 2 V).

[0007] Accordingly, in an aspect of the present invention, an electronic device is provided. The device comprises a first member and a second member. The first member comprises a first substrate comprising a first laminable material and defining a fluid channel for receiving a fluid, and first and second electrodes formed on the first substrate and separated by the fluid channel, each of the first and second electrodes comprising a conducting structure such as a nanostructure. The second member comprises a second substrate comprising a second laminable material, and a semiconductor formed on the second substrate and comprising a structure, such as a nanostructure, having a modulatable electrical property. The second member is bonded to the first member, with the semiconductor bridging the first and second electrodes across the fluid channel and the structure of the semiconductor being proximate to the fluid channel to allow the fluid received in the fluid channel to be in proximity to the structure of the semiconductor for modulating the electrical resistance between the first and second electrodes through the semiconductor. The semiconductor may consist essentially of one or more single-walled carbon nanotubes, or any conducting material inclusive of organic materials, inorganic materials, or metallic materials. The semiconductor may comprise graphene, Si, Ge, ZnO, CuO, TiO₂, SnO₂, carbonaceous material, organic semiconductor, or inorganic semiconductor. Each of the first and second electrodes may consist essentially of one or more carbon nanotubes. At least one of the semiconductor and the first and second electrodes may comprise a wire, a tube, a flake, a film, a particle, or any other form that enables its functionality. Each of the first and second substrates may comprise a polymeric material, such as polydimethylsiloxane (PDMS), poly(methyl methacrylate), polycarbonate, or polyepoxide; or organic substrate, glass or ceramic substrate, metallic substrate, or foil. Each of the first and second substrates may consist essentially of PDMS. A third electrode may be formed on the first substrate for applying a modulating voltage to an electrolyte received in the fluid channel. An electrolyte may be disposed in the fluid channel. The electrolyte may comprise a liquid, a gel, or a solid. The second member may comprise a solid electrolyte adjacent the semiconductor and a gate electrode for applying a modulating voltage to the solid electrolyte. Each one of the first and second substrates may comprise a metal coating or ceramic or glass nanoparticles. The metal coating or ceramic or glass

nanoparticles may provide a barrier towards moisture ingress into said device. Thus, the device may be sealed to provide moisture resistant properties and to promote device reliability.

[0008] According to another aspect of the present invention, in a process for forming an electronic device, first and second members are provided. The first member comprises a first substrate and first and second electrodes formed on the first substrate, the first substrate defining a fluid channel for receiving a fluid, the first and second electrodes separated by the fluid channel. The second member comprises a second substrate and a semiconductor formed on the second substrate, the semiconductor having a modulatable electrical property. The first member is bonded to the second member, with the semiconductor bridging the first and second electrodes across the fluid channel and the semiconductor being proximate to the fluid channel thus allowing the electrical property of the semiconductor be modulated by the presence of the fluid in the fluid channel. The semiconductor may comprise a nanostructure or a microstructure. The semiconductor may comprise Si, Ge, ZnO, Cu, TiO₂, SnO₂, graphene, carbonaceous material, . At least one of the semiconductor and first and second electrodes may comprise a tube, wire, flake, film, or particle, or any other form that enables its functionality. Each of the semiconductor and first and second electrodes may consist essentially of one or more carbon nanotubes. At least one of the first and second substrates may comprise polydimethylsiloxane, poly(methyl methacrylate), polycarbonate, or polyepoxide. The first and second members may be laminated by heating. The first member may comprise a third electrode formed on the first substrate for applying a voltage to the fluid received in the fluid channel. The second member may comprise a solid electrolyte adjacent the semiconductor and a gate electrode for applying a modulating voltage to the solid electrolyte. An electrolyte, such as a liquid electrolyte, may be disposed in the fluid channel. Each one of the first and second substrates may comprise a metal coating or ceramic or glass nanoparticles. The metal coating or ceramic or glass nanoparticles may provide a barrier towards moisture ingress into said device.

[0009] In accordance with another aspect of the present invention, there is provided a method of operating the electronic device disclosed herein, or formed by

a process disclosed herein. The method comprises applying a pulsed voltage signal having a peak voltage to a liquid electrolyte received in the fluid channel of the electronic device, wherein a continuous application of the peak voltage to the liquid electrolyte may cause an electrolysis reaction to occur in the liquid electrolyte, the pulsed voltage signal having a pulse profile selected to prevent the electrolysis reaction in the liquid electrolyte; and detecting an electrical signal from at least one of the first and second electrodes of the electronic device in response to application of the pulsed voltage signal.

[0010] Other aspects and features of the present invention will become apparent to those of ordinary skill in the art upon review of the following description of specific embodiments of the invention in conjunction with the accompanying figures.

BRIEF DESCRIPTION OF THE DRAWINGS

[0011] In the figures, which illustrate, by way of example only, embodiments of the present invention,

[0012] **FIG. 1** is a schematic cross-sectional view of a liquid electrolyte gated field-effect transistor;

[0013] **FIG. 2** a top plan view of an electronic device, exemplary of an embodiment of the present invention;

[0014] **FIG. 3** is a cross-sectional view of the device of **FIG. 2**, along the line A-A;

[0015] **FIG. 4** is a cross-sectional view of the device of **FIG. 2**, along the line B-B;

[0016] **FIG. 5** is a schematic diagram of a process for forming the device of **FIG. 2**, exemplary of an embodiment of the present invention;

[0017] FIG. 6 is a schematic circuit diagram for operating the device of FIG. 2, exemplary of an embodiment of the present invention;

[0018] FIG. 7 is a top plan view of another electronic device, exemplary of an embodiment of the present invention;

[0019] FIG. 8 is a cross-sectional view of a further electronic device, exemplary of an embodiment of the present invention;

[0020] FIG. 9 is a schematic diagram of a process for forming the device of FIG. 8, exemplary of an embodiment of the present invention;

[0021] FIG. 10 is schematic diagram of a process for forming a laminated electronic device, exemplary of an embodiment of the present invention;

[0022] FIG. 11 is a perspective view of the device shown in FIG. 10 during use;

[0023] FIG. 12 is a line graph of measured current-voltage characteristics of an exemplary liquid electrolyte gated electronic device;

[0024] FIG. 13 is a line graph of measured current-time characteristics of the liquid electrolyte gated electronic device of FIG. 12;

[0025] FIG. 14 is a line graph of an exemplary pulse profile for a pulsed voltage signal;

[0026] FIG. 15 is a data graph of measured current-voltage characteristics of an exemplary liquid electrolyte gated electronic device in response to application of a pulsed voltage signal to the liquid electrolyte gate of the device;

[0027] FIG. 16 is a data graph of measured current-voltage characteristics of the liquid electrolyte gated electronic device of FIG. 15, in response to application of a continuous voltage signal to the liquid electrolyte gate;

[0028] FIG. 17 is a data graph of measured current-voltage characteristics of an exemplary liquid electrolyte gated electronic device;

[0029] FIG. 18 is a data graph of measured current-voltage characteristics of an exemplary liquid electrolyte gated electronic device, in the presence of an analyte;

[0030] FIG. 19 is a data graph of measured current-time characteristics of the device of FIG. 18; and

[0031] FIG. 20 is a data graph of measured electrical characteristics of the device of FIG. 18, at different analyte concentrations.

DETAILED DESCRIPTION

[0032] An exemplary embodiment of the present invention relates to an electronic device **100** as schematically illustrated in **FIGS. 2, 3, and 4**.

[0033] Device **100** includes two members bonded to each other. The bottom member includes a bottom substrate **102** and the top member includes a top substrate **104**. Each of substrates **102** and **104** is formed of a material consisting essentially of polydimethylsiloxane (PDMS), and may have a thickness of from about 100 nm to about 5 mm. In different embodiments, substrates **102**, **104** may have different sizes, thickness, and shapes.

[0034] Substrate **102** defines a fluid channel **106** for receiving a fluid, which has a first end **108** and a second end **110**. Fluid channel **106** may have a generally U-shaped cross-sectional profile, with a length from about 100 nm to about 10 cm, a width of about 100 nm to about 1 mm, and a depth of about 50 nm to about 30 μ m. In different embodiments, the dimensions and shape of fluid channel **106** may be different from the above.

[0035] A source electrode **112** and a drain electrode **114** are formed and supported on substrate **102**. Electrodes **112** and **114** are separated by fluid channel **106**. Each of electrodes **112** and **114** consists essentially of carbon nanotubes (CNTs), such as single-walled CNTs (SWCNT). Multi-walled CNTs may also be used to form an electrode **112** or **114**. Each electrode **112**, **114** may have

a thickness of about 15 nm to about 1 mm, but in different embodiments the thickness may vary from this range.

[0036] A semiconductor **116** consisting essentially of a thin film of SWCNT is formed and supported on substrate **104**. The film of SWCNT in semiconductor **116** has a modulatable electrical property, such as a modulatable electrical conductivity. Substrates **102** and **104** are configured and bonded such that semiconductor **116** bridges electrodes **112** and **114** across fluid channel **106**, thus forming a conducting channel between electrodes **112** and **114**. Semiconductor **116** is proximate and exposed to fluid channel **106** to allow a fluid received in fluid channel **106** to contact the SWCNT in semiconductor **116** for modulating the electrical resistance between electrodes **112** and **114** through semiconductor **116**. For example, the resistance (or conductance) of a thin film of SWCNT may be modulated by the changes in its electrostatic environment. In one embodiment, the thin film of SWCNT in semiconductor **116** has an electrical property (such as its conductivity or conductance) that may be modulated by the presence of a modulating substance, such as certain biomolecules, in the proximity to the SWCNT. The resistance (or conductance) of semiconductor **116** may also be modulated when an electrolyte is disposed adjacent to it and a modulating voltage is applied to the electrolyte, as will be further described below. For effective resistance (or conductance) modulation, semiconductor **116** may have a thickness ranging from about 1 nm to about 1 mm.

[0037] Device **100** may be formed according to the process illustrated in **FIG. 5**, exemplary of an embodiment of the present invention.

[0038] As illustrated in **FIG. 5**, two slabs of PDMS of desired sizes and shapes are initially obtained. Suitable PDMS materials may be obtained from commercial sources, or may be prepared according to known techniques.

[0039] The slab of PDMS for substrate **102** is molded to form fluid channel **106**, as shown at the top of the left-hand side in **FIG. 5**. Fluid channel **106** may also be formed with another technique, such as by physical cutting, mechanical machining, embossing, imprinting, or chemical etching.

[0040] SWCNT may be obtained from commercial sources or may be prepared according to known techniques.

[0041] Optionally, pristine SWCNT may be subject to an acid treatment to form carboxylated nanotubes, e.g., according to the technique disclosed in Liu et al., "Fullerene pipes," *Science*, 1998, vol. 280, p. 1253. The presence of a carboxylic group may facilitate interaction between a modulating substance such as biomolecules and the nanotubes, thus improving the modulating effect. The carboxylic group can also form various derivatives in different conditions to provide desirable functional groups such as arene, alkyl halide, alcohol, ether, amine, aldehyde, ester, thiol, or the like. The presence of one or more of these functional groups may provide a desired functionality to the nanotubes depending on the application. However, the presence of carboxylic group is not necessary in some embodiments.

[0042] The SWCNT or carboxylated SWCNT is dispersed in an aqueous solution and is filtered using a suitable filter such as an alumina filter to form a film of SWCNT on the filter. A suitable alumina filter with particle retention size of 0.1 μm may be obtained from Whatman™. The pore sizes of the filter may be smaller than 0.1 μm and other types of filter may be used. For example, a suitable filter may be formed of polytetrafluoroethylene (PTFE), polycarbonate, or the like.

[0043] The thickness of each nanotube film is controlled by adjusting the concentration of the SWCNT in the SWCNT solution and the volume of the SWCNT solution filtered through the filter to form the film. The SWCNT concentration may be reduced by dilution.

[0044] The SWCNT films are then transfer-printed on to the respect PDMS substrates. The transfer-print may be carried out using a suitable stamping technique known to those skilled in the art.

[0045] In different embodiments, the SWCNT may be deposited on to substrates **102** and **104** in a different deposition process. For example a suitable room-temperature deposition process may be used, which may include spraying,

drop-casting, spin-coating, filtration, vacuum-filtration, physical deposition (evaporation), printing, inkjet printing, xerography, or the like.

[0046] A thick (e.g. about 15 to about 50 nm thick) SWCNT film is printed across fluid channel **106** on the bottom PDMS substrate **102**, as shown at the middle on the left-hand side in **FIG. 5**. Conveniently, both electrodes **112** and **114** can be printed with one stamp, as the SWCNT film can break at the edges of fluid channel **106** thus forming a gap between electrodes **112** and **114**. It is also possible to form the electrodes in a different manner, such as by printing them separately.

[0047] A thin (e.g. less than 10 nm thick) film of SWCNT is printed on the top PDMS substrate **104** to form semiconductor **116**, as shown on the right-hand side in **FIG. 5**. Semiconductor **116** may also be transfer-printed.

[0048] Conveniently, each SWCNT film may be deposited on a substrate **102** or **104** by a single transfer-printing or stamping procedure. It is not necessary to perform additional transfer printing of the SWCNT film. As such, the yield loss and film degradation associated with an additional transfer may be avoided. For example, significant yield loss and film degradation may result if the substrate to be used is a silicon-based substrate and the SWCNT is further transfer-printed from the PDMS slab to the silicon-based substrate.

[0049] The CNT film for electrodes **112**, **114** may have a relatively low sheet resistance, such as lower than about 1 k Ω /sq, to provide better electrical contacts. The CNT film for semiconductor **116** may have a relatively high sheet resistance, such as higher than about 300 k Ω /sq. The sheet resistance of a CNT film may be controlled by adjusting the film thickness. A thinner film for semiconductor **116** may allow more effective modulation of the resistance during use.

[0050] The two substrates **102** and **104** are then bonded to each other by lamination, with semiconductor **116** facing fluid channel **106** and positioned to bridge electrodes **112** and **114** across fluid channel **106**, as shown at the bottom of **FIG. 5**.

[0051] Lamination may be effected with any suitable technique such as by heating. A pressure may be applied to the top and bottom members to press them against each other during heating. As PDMS and CNT surfaces can bond to one another without using an external adhesive, it is not necessary to apply an adhesive to the surfaces. Conveniently, a seal may be formed between the surfaces of the top and bottom members, thus preventing fluid or liquid leakage through any gap between the substrates.

[0052] Depending on the technique used, bonding of PDMS and CNT materials can conveniently occur at room temperature.

[0053] In some embodiments, a plasma bonding technique may be used to bond the two members. The plasma bonding may utilize plasma generated from a gas such as air, oxygen or the like. Plasma bonding techniques known to those skilled in the art may be used.

[0054] An exemplary technique for preparing and bonding PDMS substrates with a fluid channel is disclosed in Jo *et al.*, "Three-dimensional micro-channel fabrication in polydimethylsiloxane (PDMS) elastomer," *Journal of Microelectromechanical Systems*, 2000, vol. 9, pp. 76-81.

[0055] Conveniently, the separation between the source/drain electrodes and the length of the conducting channel may be defined by the width of the fluid channel.

[0056] The CNT films for semiconductor **116** and electrodes **112**, **114** may be formed on substrates **102**, **104** such that the external surface of the CNT film is generally flush with the substrate surface, as depicted in **FIGS. 3** and **4**. However, this is not necessary. A CNT film may protrude from the substrate surface to facilitate solid contact between an electrode **112** or **114** and semiconductor **116**. For example, semiconductor **116** may protrude from substrate **104**. Providing a protruding part on one substrate (e.g. semiconductor **116**) and a corresponding recessed portion on the other substrate (e.g. fluid channel **106**) may facilitate the alignment of the substrates during lamination. In any event, the laminated side of

each of the two laminated members may be configured to allow secure bonding between the two members, and to allow convenient alignment.

[0057] Optionally, an electrolyte **118** such as a liquid electrolyte may be disposed in fluid channel **106** as shown in **FIG. 5** as part of the manufacturing process. A liquid electrolyte may be a polar liquid, and may include aqueous-based polar solutions, ethanol, iso-(propyl alcohol), or the like. In some applications, a lower ionic-strength of the aqueous solution may provide better performance. The ionic-strength of the solution can be determined from the solution conductance, as can be understood by those skilled in the art. For example, a phosphate-buffered saline (PBS) solution or a low ionic strength buffer (LISB) solution may be used. The solution may contain an alkali salt such as NaCl or KCl, and may have a relative stable pH such as at about 7.5. The buffer material in the solution may include phosphate.

[0058] Channel ends **108** and **110** of fluid channel **106** may be sealed during storage and transportation to prevent contamination and leakage of electrolyte **118**.

[0059] A liquid electrolyte may also be placed in fluid channel **106** during use as described below. Other types of electrolyte such as solid polyelectrolytes may also be incorporated into device **100** in some embodiments, as will be further discussed below.

[0060] Optionally, a gate electrode **120** may be provided for contacting electrolyte **118** and applying a voltage to electrolyte **118**, as will be further discussed below.

[0061] Conveniently, while not required, the liquid electrolyte may be in contact with source and drain electrodes and it is not necessary to insulate the liquid electrolyte from the source and drain electrodes.

[0062] As now can be appreciated, device **100** may be formed in a parallel processing process and may involve the use of a limited number of different materials. For example, the base unit of device **100** may consist essentially of only PDMS and CNT. Further, as a liquid electrolyte may be disposed in the fluid

channel **106** for modulating the semiconductor resistance, it is not necessary to form any solid dielectric layer on device **100**, thus allowing a simplified fabrication process.

[0063] In an exemplary embodiment, device **100** is operated as a field-effect transistor (FET) device during use as schematically illustrated in **FIG. 6**.

[0064] A fluid electrolyte **118** is placed in fluid channel **106**, to function as a liquid gate (LG). Any suitable liquid electrolyte material as discussed elsewhere herein may be used.

[0065] A gate voltage (V_g) is applied to electrolyte **118** through gate electrode **120**. The gate voltage may be applied using a signal generator **122** such as a function generator. The voltage signal may be continuous or pulsed, as will be further described below. The gate voltage may be monitored using a suitable voltmeter **124** such as an oscilloscope. While depicted separately in **FIG. 6**, the signal generator and the voltage monitor may be integrated.

[0066] A source-drain voltage (V_{sd}) is established between source and drain electrodes **112** and **114** using a suitable signal generator. The source-to-drain electrical current, I_{sd} , is measured using a suitable ammeter. The signal generator and the ammeter may be integrated and provided in ammeter-voltage source device **126** as shown in **FIG. 6**. As the source-drain current is typically in the pico-ampere to micro-ampere range, a pico-ammeter may be used in ammeter-voltage source device **126**.

[0067] Signal generator **122**, gate voltage monitor **124**, and device **126** may be in communication with a computer **128**. Computer **128** includes a processor (not separately shown) and processor readable storage medium (not separately shown). The storage medium may store processor executable program codes that, when executed by the processor, adapt the computer to control the operation of one or more devices connected to computer **128** and to analyze and store data signal received from the various devices connected to computer **128**.

[0068] As can be appreciated by those skilled in the art, when operated as illustrated in **FIG. 6**, device **100** can effectively function as a FET. In particular, due

to the applied source-drain voltage and the electrical conductivity of semiconductor **116**, an electrical current I_{sd} can effectively flow from source electrode **112** to drain electrode **114** (or vice versa) through semiconductor **116**. Electrolyte **118** in fluid channel **106** can function as a dielectric, as a double-layered capacitor can effectively form in the electrolyte when an electrical potential gradient is established within it, as oppositely charged carriers will move towards opposite directions and accumulate at the opposite boundary regions in the electrolyte. However, no significant electrical current will flow through electrolyte **118** and it is not necessary to insulate electrolyte **118** from electrodes **112**, **114** and semiconductor **116**. In some applications, such as when device **100** is used as a FET, resistor, or diode, an insulation layer such as a dielectric layer may be placed between semiconductor **116** and electrolyte **118**.

[0069] In any event, as electrolyte **118** is disposed in the proximity to semiconductor **116**, the formation of electrically charged layers near the interfacial region between semiconductor **116** and electrolyte **118** can modulate the electrical conductance of semiconductor **116**. Thus, the source-drain current I_{sd} at a fixed biasing voltage V_{sd} is modulated by, and is a function of, gate voltage V_g . In other words, an electrical characteristics of the FET, such as the current response to a given applied source-drain voltage, is modulated by the gate voltage through electrolyte **118**. Thus, device **100** can function as a FET device.

[0070] Conveniently, when a liquid electrolyte is used as the gate dielectric, device **100** can properly function with a reduced applied gate voltage as compared to a traditional FET with a solid dielectric between the gate electrode and the transistor channel. Depending on the particular liquid electrolyte used, the effective capacitance of the electrolyte under an applied gate voltage can be more than 1000 times higher than the capacitance in a conventional dielectric layer used in traditional FETs. For example, the effective capacitance in a liquid electrolyte can be more than $1000 \mu\text{F}/\text{cm}^2$. In comparison, the capacitance of a solid polymeric or oxide dielectric typically used in traditional FETs is about $1 \mu\text{F}/\text{cm}^2$. As such, device **100** may be operated with a biasing voltage below about 1 V. In comparison, for a CNT-based transistor with a SiO_2 gate dielectric, the operational gate voltage is typically from 2 to 40 V. The use of liquid electrolyte in device **100**

may also allow a stronger electrical signal, such as the source-drain current, to be generated.

[0071] Device **100** can also be used as a sensor, such as a biosensor, for detecting various analytes, such as biomolecules or other analytes with sizes as small as a nanometer.

[0072] In an exemplary process, to detect the analyte in a sample solution, the sample solution is provided to fluid channel **106**, such as by providing a flow of the sample solution through fluid channel **106**. Conveniently, the sample solution may be mixed with a fluid electrolyte and fed to a channel end **108** or **110**, which can serve as a fluid inlet. Alternatively, the sample solution may be fed to fluid channel **106** separately. A continuous flow of the sample solution may be maintained, such as by withdrawing the sample solution through another end **110** or **108** of fluid channel **106**, which may serve as a fluid outlet. To maintain a flow of the sample solution, a suitable fluid circuit and a pump may be provided. The pump may include a syringe pump, electro-kinetic pump, piezoelectric pump, or the like.

[0073] In any event, when the analyte is present in the sample solution it will be dispersed inside fluid channel **106**, and within the proximity to semiconductor **116**. The presence of the analyte can be detected by monitoring a change in electrical characteristics of device **100**, such as a change in the electrical conductance of semiconductor **116**, as reflected in a change in the current-voltage characteristics or the current-time characteristics of device **100**.

[0074] As is known, the presence of many analytes in the proximity to the SWCNT can affect and modulate the conductance of the SWCNT. Various underlying mechanisms have been proposed for this phenomenon. For example, some analytes, when present, can disturb the electrostatic environment around the SWCNT. Without being limited to any particular theory, it has been suggested that this disturbance can affect the conductance of the nearby CNT. Electrostatic gating may occur due to interaction between biomolecules in the fluid channel. For example, such interactions may include immuno-complex formation, DNA hybridization, or the like. Other possible mechanisms for modulating the semiconductor's conductance reported in the literature include Schottky barrier

modulation, capacitance effect, and mobility change. Depending on the particular mechanisms to be utilized, the device may be modified to more effectively utilize the modulating mechanism. For example, if contact of the analyte with the semiconductor is required for modulating the semiconductor's conductance, the device may be configured to facilitate contact between the analyte and the semiconductor. In other cases, the device may be configured to allow the analyte to move close to semiconductor **116** such as within the Debye length as discussed above.

[0075] Different modulation mechanisms may be identifiable from the device's electrical response to the presence of the modulating substance. For example, electrostatic gating may cause a shift in the threshold gate voltage. Schottky barrier modulation may result in a decrease in I_{sd} at $V_g < 0$ and an increase in I_{sd} at $V_g > 0$. Capacitance effect may result in a decrease in the gradient of the transfer characteristic at negative and positive V_g . Mobility changes (possibly scattering induced) may cause a decrease in I_{sd} at both positive and negative V_g regions.

[0076] As now can be appreciated, in order to effectively modulate the conductance of semiconductor **116**, the size and thickness of semiconductor **116** should be selected to allow modulation of a sufficient portion of the CNTs in the conducting channel between electrodes **112** and **114** by the presence of the analyte in fluid channel **106**, so that the electrical resistance between electrodes **112** and **114** through semiconductor **116** may be detectably modulated by dispersing an analyte in fluid channel **106**. A more effective modulation may be achieved if a larger percentage of semiconductor **116** is modulatable by the presence of the analyte. In this regard, if semiconductor **116** is too thick, some of the CNTs remote from fluid channel **106** will not be significantly modulatable by the analyte dispersed in fluid channel **106**. Thus, a thinner semiconductor **116** may provide more effective modulation. In some embodiments, semiconductor **116** may be as thin as the size of a single CNT. In some applications, a single nanotube may be used to form the conducting channel.

[0077] The change in electrical conductance of semiconductor **116** affects the electrical characteristics of device **100**, such as its current-voltage (I_{sd} - V_g) characteristics, or current-time (I_{sd} - t) characteristics. Thus, a change in the measured electrical characteristics of semiconductor **116** or device **100**, such as the current-voltage or current-time characteristics, can be correlated to the presence or the quantity of the analyte in the sample solution in some applications.

[0078] The current-voltage characteristics of device **100** may be measured in both the absence and the presence of an analyte. A continuous (DC) V_g signal may be applied, and varied (swept) between -0.8 and 0.8 V. Alternatively, a pulsed V_g signal may be applied, with the amplitude varied (swept) between -2 and 2V. The source-drain bias V_{sd} may be either a constant DC voltage or an AC voltage with a low frequency (< 5 mHz), and may be in the range of 10 - 50 mV. The current-time characteristics may be monitored with similar settings as described above but with the amplitude of gate voltage V_{sd} fixed at a constant value. See Examples for further discussion.

[0079] In addition, by monitoring the changes in different electrical characteristics of device **100** over time, it may be possible to study the reactions and interactions occurring in the solution. For example, the types of reactions occurred may be identified based on the reagents and products detected using device **100**. It is also possible to deduce the underlying modulation mechanism by monitoring the changes in electrical characteristics of device **100**. For instance, decreasing I_{sd} with negative V_g biasing and increasing I_{sd} with positive V_g biasing may indicate modulation of the Schottky barrier as it is characterized by such a trend.

[0080] A liquid electrolyte may be delivered to fluid channel **106** in an aqueous solution together with the sample solution. Since the natural environment for many analytes is aqueous, using liquid electrolyte as the gate material allows the analyte or biomolecular reactions involving the analyte be detected and studied in an environment similar to its natural environment. It is also not necessary to label, such as to dye, the analyte. Further, device **100** does not require that the analyte reacts with the electrolyte in order to detect the presence of the analyte.

This allows more types of analyte be detected and allows more direct detection, which may lead to improved detection sensitivity and accuracy.

[0081] As discussed above, the gate voltage may be lower than about 1 V when a liquid electrolyte is used. However, in some applications, a higher gate voltage may be desirable and may be applied. When a sufficiently high voltage is applied continuously to an electrolyte in an aqueous solution, electrolysis reactions may occur in the solution, which may be undesirable and may affect the proper function of device **100**. Thus, a pulsed voltage signal with a pulse profile selected to prevent the electrolysis reaction in the electrolyte solution may be applied to electrolyte **118**. For example, assume that the continuous application of a voltage above a threshold voltage to a given electrolyte solution will cause an electrolysis reaction to occur in the solution, and the pulsed voltage signal has a peak voltage that is higher than the threshold voltage. Thus, a continuous application of the peak voltage to the electrolyte solution may cause an electrolysis reaction to occur in the electrolyte solution. In such a case, if the pulsed voltage signal has a pulse profile where the voltage is dropped from the peak value to below the threshold voltage and then increased to another peak intermittently, and the duration of each time interval above the threshold voltage is less than the time required to complete the electrolysis reaction, then the electrolysis reaction may be avoided when the pulsed voltage signal is applied to the electrolyte solution. For example, in some embodiments, each pulse may have a pulse length substantially less than 1 ms, or the pulse frequency may be substantially higher than 1 kHz. Tests show that application of a voltage from -1.8 to 0.2 V, or from -2 to 0.5 V, at a frequency of 100 kHz is sufficient to avoid electrolysis in a LISB solution.

[0082] Without being limited to any particular theory, it is expected that an electrolysis reaction takes a finite amount of time to complete at a given applied voltage. Thus, when the applied voltage above the threshold voltage only lasts for a period shorter than the required electrolysis reaction time, then electrolysis may be avoided.

[0083] The pulsed voltage signals may be generated by signal generator **122**. A sample pulsed voltage signal profile is described in the Examples.

[0084] When a pulsed gate voltage signal is employed, the range of the gate voltage may be increased without causing electrolysis reaction in the liquid electrolyte. This may provide certain benefits. For example, the current response to the conductance modulation may be more pronounced when the gate voltage is higher, due to, for instance the contribution from changes in saturation current in addition to the contribution from the change in threshold voltage (I-V shift). At a low gate voltage, the only contribution may be from changes in threshold voltage. As a higher detection signal level can be obtained at a given concentration of the analyte, more sensitive detection can be achieved, and the limit of detection (LOD) may be improved.

[0085] Device **100** may be modified in different embodiments. For example, a substrate **102** or **104** may include a material other than PDMS. Other suitable substrate materials include poly(methyl methacrylate) (PMMA), polycarbonate, polyepoxide (epoxy), or the like. When a list of items is given herein with an “or” before the last item, as in the preceding sentence, any of the listed items or any of the suitable combinations of the listed items may be selected and used.

[0086] The SWCNT film in semiconductor **116** may also be replaced with another nanostructure having a modulatable electrical conductivity. For example, different nanotubes or nanowires may be used. Further suitable nanostructures are described elsewhere herein.

[0087] Further, modulation of the effective electrical resistance of semiconductor **116** may be effected by utilizing various modulation mechanisms, which may include conductance modulation, capacitance modulation, Schottky barrier modulation, charge carrier mobility modulation, or electrostatic gating. Depending on the modulating substance used and the modulation mechanism utilized, it may not be necessary for the modulating substance to contact semiconductor **116** in order to modulate an electrical property of semiconductor **116** such as its apparent electrical resistance. In such a case, semiconductor **116** may be placed proximate to fluid channel **106** so that a fluid received in fluid channel **106** can move sufficiently close to semiconductor **116** to allow the modulation to occur. For example, in some applications, the modulating substance may be

biomolecules which can modulate the electrical properties of a CNT when the distance between the biomolecules and the CNT is less than the Debye length. The Debye length is typically less than 100 nm for many biomolecules. Thus, in some embodiments, it may not be necessary to expose semiconductor **116** to fluid channel **106**, and a layer of a suitable substance (not shown) may be placed between the modulating substance and semiconductor **116**, when a modulation mechanism can still be effectively utilized to modulate an electrical property of semiconductor **116** in the presence of this layer.

[0088] Electrolyte **118** may include a hydrogel electrolyte. For example a hydrogel formed of agarose, vinylpyrrolidone, methacrylamide, N-vinylimidazole, or the like may be used. Other gels may also be used as an electrolyte.

[0089] In addition to the liquid electrolyte, a solid polyelectrolyte may also be used. In liquid electrolytes and polyelectrolytes, the charge mobility may be relatively high, such as up to $0.1 \text{ cm}^2/\text{Vs}$ for liquid electrolyte.

[0090] In different embodiments, an electrode may be formed of a conducting nanostructure, a metal or other suitable conducting materials.

[0091] Additional electrodes may be added. For example, as illustrated in **FIG. 7**, in a modified device **200**, which may be similar to device **100**, a gate electrode **220** is formed on bottom substrate **202**. Device **200** may be otherwise constructed similar to device **100**. Gate electrode **220** extends into fluid channel **206** so that a gate voltage may be applied to the electrolyte received in fluid channel **206** through gate electrode **220**. Gate electrode **220** may be formed of CNT or another suitable material such as Au, Ag, Ag/AgCl, or the like. Device **200** may also be further modified. For example, the electrodes, semiconductor, and substrates in device **200** may be formed of various other suitable materials.

[0092] As illustrated in **FIG. 8**, a further back gate may also be provided to form a dual-gate device **300**. Device **300** may be similar to device **100** or **200** except that a solid electrolyte **322** is formed in top substrate **304** adjacent semiconductor **316**, and a gate electrode **324** is formed in top substrate **304** and in contact with solid electrolyte **322** for applying a gate voltage to electrolyte **322**.

Further, substrates **302**, **304**, electrodes **312**, **314**, and semiconductor **316** in device **300** may be formed of various suitable materials different from those described above with reference to device **100**.

[0093] Solid electrolyte **322** may be a polyelectrolyte. Some exemplary suitable polyelectrolytes are described in Herlogsson et al., "Low-Voltage Polymer FET Gated via a Proton Semiconductor," *Advanced Materials*, (2007), vol. 19, pp. 97-401; and Said et al. "Polymer field-effect transistor gated via a polystyrene sulfonic acid thin film," *Applied Physics Letters*, (2006), vol. 89, p. 143507.

[0094] Device **300** may be formed in a process as illustrated in **FIG. 9**. In particular, the PDMS slab for the top substrate **304** may have a hole. The semiconductor film may be attached to solid electrolyte **322** and they both may be inserted into the hole in the PDMS slab. Gate electrode **324** may be deposited on top of solid electrolyte **322** either before or after lamination. The other components of device **300** may be prepared as described elsewhere herein.

[0095] Conveniently, device **300** may be operated in two different modes and may exhibit two different current-voltage characteristics depending which electrolyte gate is used to modulate the conductance of the semiconductor. For example, a bias voltage may be established across electrodes **312** and **314** to produce a current through semiconductor **316**. The current may be modulated by applying a voltage to liquid electrolyte **318**, or solid electrolyte **322**, or by applying voltages to both electrolytes **318** and **322**. As the two gate voltages may be separately controlled, more complex measurements such as multi-parametric responses may be obtained, as can be understood by those skilled in the art.

[0096] For example, when an analyte is introduced into the fluid channel on substrate **302** and is in the proximity to semiconductor **316**, its presence may be detected by applying a gate voltage to solid electrolyte **322**. A gate voltage may also be applied to liquid electrolyte **318** to detect the analyte. Thus, the detection of the same analyte may be independently verified through the use these two different electrolyte gates. Detection may also be made by using both electrolyte gates concurrently.

[0097] It can be expected that the electrical response of device **300** may exhibit different characteristics when the two different gates are used. For example, the effective capacitor thickness in the solid electrolyte and the liquid electrolyte may be different. The difference between the two response characteristics may provide additional information on the analyte present in the fluid channel. Thus, more sensitive and accurate detection may be obtained in some applications.

[0098] A further embodiment of the present invention relates to a process for forming an electronic device **400** which includes a laminated structure formed of a first member **402** and a second member **404**, as illustrated in **FIG. 10**.

[0099] Member **402** has a surface **406**, which defines a fluid channel **408**. Two electrodes **410** and **412** on surface **406** are separated by channel **408**.

[00100] Member **404** has a surface **414**. A semiconductor **416** is formed on surface **414**.

[00101] The laminated structure is formed by laminating member **402** and member **404** to each other, with surface **406** of member **402** facing surface **414** of member **404**. Lamination may be effected by heating, or another suitable technique. For example, a plasma bonding technique may be used. Lamination may also be effected by pressing members **402** and **404** towards each other. For example, the two opposing polymer surfaces **406** and **414** may form a seal due to hydrophobic interactions between the surface molecules. Surfaces **406** and **414** may be chemically treated or functionalized to facilitate lamination.

[00102] In the laminated structure, semiconductor **416** is in contact with both electrodes **410** and **412**, thus bridging (connecting) them. A portion of semiconductor **416** faces and is proximate to fluid channel **408**. Semiconductor **416** may be exposed to fluid channel **408** in some embodiments.

[00103] Each of members **402**, **404** may include a substrate formed of a material suitable for lamination. For example, each substrate may be formed of polydimethylsiloxane (PDMS), poly(methyl methacrylate) (PMMA), polycarbonate, or polyepoxide (epoxy). Other materials may also be used. The substrates for

members **402**, **404** may be formed of the same or different materials. It may be convenient in some embodiments if both substrates are formed of materials suitable for direct lamination. In some embodiments, a laminating material may be applied to surfaces **406** or **414**, or both, to facilitate lamination between members **402**, **404**. It may also be convenient if, after lamination, surfaces **406** and **414** form a seal to prevent leakage of fluid from channel **408** through any gap between surfaces **406** and **414**. The surface material in channel **408** should be compatible with the fluid to be received therein, as will become clear below. The substrates may have any suitable size or shape, and may be multi-layered. Each of members **402**, **404** may also include different layers depending on the particular application, as illustrated elsewhere herein.

[00104] Channel **408** may be formed by molding, cutting, or etching a groove in the substrate of member **402**. The shape and sizes of channel **408** may vary depending on the application. In some embodiments, channel **408** may have a generally U-shaped cross-sectional profile as illustrated in **FIG. 10**. In other embodiments, channel **408** may have another cross-sectional profile. It is not necessary that channel **408** be elongated. For example, channel **408** may be generally well-shaped in some embodiments. The width and depth of channel **408** may be on the order of micrometer or nanometer. The length of channel **408** may be on the order of micrometer to centimeter. In some embodiments, channel **408** may have a length of about 100 nm to about 10 cm, a width of about 100 nm to about 1 mm, and a depth of about 50 nm to 30 μm . In some embodiments, fluid channel **408** may be a microchannel, with a length from about 0.5 to about 1 cm, a width from about 50 to about 400 μm , and a depth from about 1 to about 20 μm . Channel **408**, however, should be able to receive a sufficient volume of an electrolyte and sufficient analyte for the particular application.

[00105] Electrodes **410** and **412** may be formed of any suitable electrode material. For example, a suitable metal, compound, or composite may be used. Suitable electrode materials may include, e.g., Ag/AgCl, Cr, Au, Pt, nanotubes, nanowires, or the like. Electrodes **410** and **412** may be deposited on surface **406** of member **402** by transfer printing, or any other suitable technique. The electrode

material in a particular application may be selected in view of other materials that may come into contact with the electrode and the desired electrode performance.

[00106] Semiconductor **416** may include any suitable conducting material that has a modulatable electrical conductivity. For example, semiconductor **416** may include a semiconducting material or structure, or a nanostructure having a modulatable electrical property such as modulatable conductivity. Semiconductor **416** may be formed of materials that have different shapes, including films, tubular materials such as nanotubes or nanowires, particulates such as nanoparticles or nanoflakes. A nanostructure has a nanometer-scale characteristic feature that affects its functionality. The nanometer-scale feature typically has a size in the range from about 1 nm to about 100 nm. In other embodiments, semiconductor **416** may have a microstructure, which has a characteristic feature size in the range of micrometers. For example, a nanostructure may include one or more nanotubes, nanowires, graphene (e.g. graphene flakes), or nanoparticles. A unit structure in the nanostructure may have a tubular or cylindrical shape, or may have a generally planar shape (e.g. thin film). Semiconductor **416** may include organic or inorganic materials or metallic materials, and may include materials formed of molecules with various sizes and weights such as polymers, oligomers, and small molecules. For example, organic semiconductors such as poly(3-Hexylthiophene) (P3HT), poly(3,3'-dialkylquater-thiophene) (PQT), pentacene, or the like, may be used in semiconductor **416**. The semiconductor or the nanostructure may have either p-type or n-type charge carriers. For example, the nanotubes may be CNT such as single-walled CNT (SWCNT) or multi-walled CNT (MWCNT). The nanowires may be formed of Si, Ge, ZnO, CuO, TiO₂, SnO₂, or the like. A suitable nanostructure may include any nanostructure that can exhibit different electrical characteristics under a modulating field or in the presence of a modulating material such as an analyte. For example, under suitable conditions the conductance of many conducting nanostructures such as CNTs can be modulated by a modulating electrical field or by the presence of various analytes.

[00107] Semiconductors **416** may be formed on surface **414** of member **404** using any suitable technique. In one embodiment, CNTs may be initially formed (grown) on a separate wafer and then transfer-printed on surface **414**.

Alternatively, the substrate material for member **404** may be a suitable porous/filter material for collecting the nanostructure such as CNTs formed on the separate water. Conveniently, nanostructures such as nanowires may be directly grown on a suitable filter material, such as using a chemical vapor deposition (CVD) technique. An exemplary technique of growing CNT by CVD is described in Kocabas et al., "Spatially Selective Guided Growth of High-Coverage Arrays and Random Networks of Single-Walled Carbon Nanotubes and Their Integration into Electronic Devices," Journal of the American Chemical Society, (2006), vol. 128, pp. 4540-4541.

[00108] Deposition of conducting material on a substrate may also be carried out using any of the following techniques, physical vapor deposition such as evaporation, drop-casting, spin-coating, filtration, vacuum-filtration, printing, inkjet printing, xerograph, or the like.

[00109] The techniques of forming and transferring nanostructures such as CNTs and nanowires on different substrates are known to those skilled in the art. Techniques described elsewhere herein may also be used.

[00110] Member **404** may be multi-layered. For example, nanostructure **416** may be deposited on a sub-layer of member **404**, where the sub-layer is formed of a porous/filter material for collecting nanostructures such as CNTs and nanowires. Member **404** may also include a substrate sub-layer for supporting the porous/filter layer.

[00111] Each of members **402**, **404** may also include an outer protective layer (not shown), as can be understood by those skilled in the art.

[00112] Members **402** and **404** may be bonded using any suitable technique. For example, they may be bonded using a lamination technique known to persons skilled in the art. Lamination may be effected by heating or another technique. For example, a pressure may be applied to facilitate bonding between members **402** and **404**. A plasma bonding technique may be employed. The bonding between PDMS materials may be effected as described above with reference to device **100**. Suitable bonding techniques may include chemical bonding, pressure bonding,

electrostatic bonding, brazing/soldering, mechanical locking, or the like. Bonding between different materials may be effected using similar or different techniques depending on the particular materials used. For example, exemplary techniques for bonding different substrates including polymer substrates are disclosed in Vlachopoulou *et al.*, "A low temperature surface modification assisted method for bonding plastic substrates," *J. Micromech. Microeng.*, 2009, vol. 19, pp. 15007-15012; and Chia-Wen Tsao and Don L. DeVoe, "Bonding Techniques for Thermoplastic Microfluidics," in *Lab-on-a-Chip Technology (Vol. 1): Fabrication and Microfluidics*, Chapter 5, ed. Keith E. Herold and Avraham Rasooly, 2009, Caister Academic Press.

[00113] As can be appreciated by those skilled in the art, the process for forming device **400** may be relatively simple and may involve less processing stages and less intermediate materials, as compared to some conventional processes for forming FET devices. For example, in some embodiments, it is not necessary to transfer-print the nanostructure, or transfer-print the nanostructure more than once. Conveniently, members **402** and **404** may be prepared or formed in parallel (parallel processing) before the lamination stage, thus saving processing time and allowing more flexible arrangement or configuration of the production line or plant and a higher production rate. When the electrodes and the semiconductor are formed of the same or similar materials such as CNT and are bonded to each other by lamination, there may be a relatively low contact resistance between them. Thus, in such a case, it may not be necessary to anneal the device in order to further reduce the contact resistance.

[00114] During use, device **400** may be used as a FET, similarly as discussed above with reference to devices **100**, **200**, and **300**. In particular, channel **408** may be filled with an electrolyte **418** as shown in **FIG. 11**. Electrolyte **418** may be a liquid electrolyte or a hydrogel, as discussed elsewhere herein.

[00115] Electrolyte **418** forms a dielectric volume in channel **408** adjacent semiconductor **416**. An external electrode **420** may be inserted into channel **408** to contact electrolyte **418** to serve as a gate electrode. Electrodes **410** and **412** may be used as source and drain electrodes respectively, as illustrated in **FIG. 11**, and

in a manner similar to electrodes **112** and **114** in device **100**. Device **400** may thus be operated as a FET device. In particular, when appropriate biasing voltages are applied to source electrode **410**, drain electrode **412**, and gate electrode **420**, a current signal may be detected, e.g. using a circuitry similar to that depicted in **FIG. 6**.

[00116] As will be understood by those skilled in the art, a detected electrical signal such as a current signal at an electrode such as electrode **410** or **412** is dependent on the voltage applied at the different electrodes. In some embodiments, the detected signal is also dependent on a modulating electrical field or the presence or absence of a modulating material in the proximity to the transistor channel region which is formed by semiconductor **416**. The modulating field experienced by semiconductor **416** may be produced by a voltage applied to electrolyte **418** through gate electrode **420** and can be further modulated by other substance present in fluid channel **408**.

[00117] As can be understood, a liquid electrolyte can be conveniently used with device **400**. For example, leakage through gaps between surfaces **406** and **414** can be prevented, as discussed above.

[00118] As discussed above, when a liquid electrolyte is used, device **400** may be operated with lower biasing voltages (such as below about 1 V), as compared to traditional FET devices, and may provide more sensitive detection when it is used as a sensor.

[00119] Device **400** may also be used as a sensor or detector for sensing or detecting the presence or the amount of an analyte. When different materials are present in channel **408**, different electrical characteristics, e.g., different current responses to applied voltage (I-V response), may be exhibited by device **400**. Device **400** may therefore be conveniently used to detect the presence or the amount of an analyte in a sample solution.

[00120] For example, the sample may be disposed in channel **408** with electrolyte **418**. A suitable voltage signal is applied to electrolyte **418** such as through an electrode. An electrical response, such as a current signal, is measured

or detected at one or more of electrodes **410**, **412**, **420**. The detected electrical response can then be correlated to the presence and the amount of the analyte in the sample.

[00121] Thus, conveniently, device **400** may be used as a sensor or detector for detecting different analytes.

[00122] As compared to traditional FET or sensor devices, a device described herein may provide more sensitive detection when used as a sensor.

[00123] In devices **100**, **200**, **300** or **400**, the top and bottom members may include a protective, such as moisture-resistive, coating or material (not separately shown). The coating or material may include metal, ceramic, or glass nanoparticles. This material or coating may cover a portion of the device or the entire device to prevent moisture ingress. A completely sealed device may be moisture-resistant and have improved reliability.

[00124] Embodiments of the present invention are further illustrated by the following non-limiting examples.

[00125] Examples:

[00126] EXAMPLE I

[00127] Sample devices were prepared as described above. Both top and bottom substrates were formed of PDMS.

[00128] To prepare semiconductor and source and drain electrodes formed of CNTs, a CNT material (P2-SWNT) was purchased from Carbon Solutions, Inc, and was annealed for 2 hours at 900 °C in Argon environment to minimize the number of carboxyl groups attached to the CNT prior to solution preparation. In some cases, a suspension of 0.1 mg/ml CNT in 1% sodium dodecyl sulfide (SDS) was sonicated and centrifuged for 1 hour at 14000 rpm to remove bundles from the suspension. In other cases, an aqueous dispersion of SWCNT (0.5 mg ml⁻¹) was prepared by dissolving 25 mg of SWCNT in 50 ml of Sodium-dodecyl benzene sulfonate (SDDBS) solution (1 weight%), which was purchased from Sigma-

Aldrich™. The dispersion was then sonicated for approximately two hours for homogenization.

[00129] CNT films were prepared from the CNT solution using a vacuum filtration method as disclosed in Zhou. Briefly, a desired amount of the CNT solution was filtered through an alumina filter (Whatman, Inc., 0.1 μm), rinsed extensively with deionized water to remove any remaining SDS surfactant, and picked up with a PDMS substrate.

[00130] Semiconductors formed of inorganic nanowires (including ZnO and TiO_2 nanowires) were prepared similarly. In particular, the nanowires were dispersed in solutions and deposited on the PDMS substrates under a similar procedure as in the case of CNTs, but without using any surfactant.

[00131] Semiconductors formed of PQT were prepared as follows. PQT was procured from American Dye Source™ (ADS). Si/SiO₂ wafers (n-type) were cleaned according to industry standard procedures. A thin film of PQT was deposited on each wafer by spin-coating 0.3 wt.% PQT in 1,2-dichlorobenzene at 1000 rpm for 140 s, followed by annealing in vacuum at 140 °C, while 10 mg/ml of TEPP-C61 solution in 1,2-dichlorobenzene was prepared to deposit the film at 1000 rpm, 120 s, followed by annealing in N₂ at 120 °C. The PQT film was then picked up on the PDMS stamp by pressing the PDMS stamp to the PQT material and peeling off the PDMS stamp. The thickness of the PQT films for different sample devices ranged from 10 to 100 nm.

[00132] The PDMS substrates/molds used in the sample devices were formed from PDMS materials purchased from Dow Corning, Inc.™ (SLYGARD 184 silicone elastomer kit). To cure the PDMS, the PDMS resin was mixed with a curing agent with a weight ratio of the resin to curing agent of 10:1. The mixture was degassed in vacuum for 1 hour and poured onto a pre-patterned Si master, followed by 1 hour of curing at 100 °C. The PDMS substrate/mold was cooled to room temperature before use.

[00133] The semiconductors and electrodes were transfer-printed on to the respective substrates as described above.

[00134] EXAMPLE II

[00135] The devices formed in Example I were operated as liquid-gate FET (LGFET).

[00136] A low leak flexible reference electrode (3M KCl) (FLEXREF from World Precision Instruments™) was used to apply gate voltage to the liquid electrolyte. A voltage signal from a function generator (Thurlby Thandar Instrument™, TTI TG1304) was applied to the reference electrode, with the drain electrode being grounded. A 10 mV bias voltage was established across source and drain electrodes. The conductance of the device was monitored. In some measurements, an Ag/AgCl reference electrode (World Precision Instruments™) was also used. The applied V_g signal was monitored using a digital oscilloscope (Agilent™ DSO3062A). A voltage source/pico-ampere meter module (Keithley™ 6487) was used to apply the source-drain bias voltage (V_{sd}) and to measure the source-drain current (I_{sd}). All the electrical instruments used were synchronized by a personal computer through a general purpose interfacing bus (GPIB) card (National Instrument) and a programming code written in LabVIEW 7.1.

[00137] The electrolyte was dispersed in a low ionic strength buffer (LISB) composed of 0.5 mM phosphate buffer and 1.5 mM NaCl (pH ~ 7.5). The solution was dispersed in the fluid channel for the measurements.

[00138] Representative results of the current-voltage measurements are shown in **FIG. 12**, and representative results of the current-time measurements are shown in **FIG. 13**.

[00139] As shown in **FIG. 12**, depending on the type of charges present in the sample solution and the type of the semiconductor, the I-V curve shifted and changed after the analyte was introduced.

[00140] **FIG. 13** shows that the current dropped after the analyte was introduced into the fluid channel.

[00141] EXAMPLE III

[00142] The performance of sample devices under pulsed gate voltage was investigated.

[00143] **FIG. 14** shows an exemplary pulse profile for a voltage signal applied to the liquid electrolyte in a sample liquid electrolyte gated device for a current-time measurement. For I-V measurements, the frequency of the pulsed signal was the same but the amplitude of the peak voltage was varied over time.

[00144] Representative measured I-V data under the application of a pulsed voltage signal is shown in **FIG. 15**. In comparison, the measured I-V data from the same device but under a continuous voltage signal is shown in **FIG. 16**. As can be seen, using pulsed gate voltage signal significantly improved the performance of the device.

[00145] Test results also showed that when the gate voltage was increased to above about 1 V, application of a continuous voltage signal resulted in a significant reduction in source-drain current, while the current remained relative stable with the pulsed voltage signal. Further, the current-time measurements showed that using a pulsed voltage signal did not change the current-time characteristics of the device as compared to a continuous voltage signal, which indicated that application of the pulsed voltage signal did not alter the behavior of the device itself.

[00146] It was estimated that the time required for electrolysis reaction to complete in pure water is about 1 ms. Thus, a pulse frequency of higher than about 1 kHz may prevent electrolysis in pure water. In different solutions, the electrolysis reaction time may vary, and the threshold frequency may correspondingly vary.

[00147] **FIG. 17** shows the I-V characteristics of the sample device having a PQT semiconductor, where the PQT film was transfer printed from a SiO₂ substrate. Similar results (not shown) were obtained from devices with PQT semiconductor formed by direct spin coating on the PDMS substrate.

[00148] EXAMPLE IV

[00149] Poly-L-Lysine (PLL) was used as the analyte for detection by a sample liquid electrolyte gated electronic device with CNT semiconductor/electrodes and PDMS substrates. The resistance of the CNT films for forming the semiconductor and the source/drain electrodes were adjusted by controlling the filtration volume and hence the density of the nanotube films. The CNT film for the semiconductor (transistor channel) had a sheet resistance of larger than 300 k Ω /sq. The CNT films for the electrodes had a sheet resistance of less than 1 k Ω /sq. The surfaces of the PDMS substrates were kept clean before lamination to ensure self-sealing between the laminated PDMS surfaces. The liquid channel was a microfluidic channel with a width of 400 μ m. The transistor channel length (distance between the source and drain electrodes) was defined by the width of the microfluidic channel and was thus also about 400 μ m. It was found that when the length of the transistor channel was 2000 μ m (thus giving a width/length ratio of 2000/400), the resistance of the CNT semiconductor film was 20-25 k Ω .

[00150] Initially system stability was examined by drawing LISB into the microchannel (fluid channel) while recording the signal levels. Only small signal disturbances were recorded that might be associated with renewal and stabilization of the electrical double layer capacitance in the liquid electrolyte as new electrolyte was added.

[00151] PLL was purchased from Sigma Aldrich™ (p-4707). A series of diluted solutions of PLL were prepared in de-ionized water (*MilliQ*) at increasing concentrations: (1) 0.2 aM, (2) 2 aM, (3) 20 aM, (4) 200 aM, (5) 2 fM, (6) 20 fM, (7) 200 fM, (8) 2 pM, (9) 20 pM, (10) 200 pM, (11) 2 nM, (12) 20 nM, (13) 200 nM.

[00152] An optimum incubation time of PLL was calculated and used in the tests. The PLL solutions were introduced into the fluid channel using a syringe. For each measurement, after flushing the fluid channel several times with LISB, a PLL with a given concentration (e.g. 1 nM) was injected into the electrolyte reservoir. A signal fluctuation was observed after injection of the PLL solution, followed by restoration, which indicated that the transfer characteristic reflected the

response at the semiconductor-electrolyte interface at the middle of the fluid channel.

[00153] The interaction between the PLL and the nanotubes in the fluid channel was studied electrically through I_{sd} - V_g and current-time measurements with and without application of a pulsed gate bias. In the case of I_{sd} - V_g measurement, the V_g bias was swept across the range of -0.8 to 0 V (simple DC sweep) or the range of -5 to 0 V (pulsed-gate, frequency of 100 kHz). The real-time interaction between the PLL and the nanotubes was studied through current-time measurements, whereby the V_g bias was kept constant at -0.5 and -2 V for simple DC bias and pulsed-gate, respectively, and the current I_{sd} was measured as a function of time. When the leakage current was small, the current measured at the drain electrode (I_d) (or source electrode) was used to approximate the source-drain current (I_{sd}).

[00154] Upon drawing PLL into the microchannel, the I_{sd} - V_g curve shifted to the negative direction, e.g. by about 0.1 V with 1 nM of PLL.

[00155] **FIGS. 18, 19, and 20** show representative data obtained.

[00156] As shown in **FIG. 18**, the I_{sd} - V_g curve shifted towards left (down) after 90 nM PLL was injected into the buffer (LISB) solution.

[00157] **FIG. 19** shows the real-time measurement of I_{sd} , taken at gate potential of -100 mV (corresponding to the dashed line shown in **FIG. 18**). The changes in semiconductor conductance at various times due to changes occurring in the fluid channel can be observed, as indicated in **FIG. 19**. It was also found that PLL attachment to the semiconductor is an irreversible process. Tests also showed that the sample devices and comparison devices in which the CNT semiconductor was grown using a CVD process exhibited similar changes in I-V characteristics when exposed to PLL.

[00158] **FIG. 20** shows representative measured electrical responses from the sample device at different PLL concentrations. As can be seen, both the I-V response and the I-t response can be correlated to the quantity (concentration) of

PLL in the fluid channel. Thus, the sample device could be used to detect the quantity of the PLL analyte in a sample solution.

[00159] The measured data indicated that the dominant conductance modulating mechanism in this case was electrostatic gating. It was expected that the highly positively charged PLL (pH ~ 7.5) was absorbed into the CNT film and induced a negative charge screening on the CNT surface, thus shifting the I-V curve towards a negative gate voltage and leading to a reduction in I_{sd} .

[00160] It was observed that when the concentration of the analyte (PLL) was below about 10 pM, the signal change was linearly proportional to the analyte concentration, but when the analyte concentration increases to above 10 pM, this linear relationship was lost (referred to as signal saturation). This signal saturation may imply that the CNT film surface was completely covered by PLL molecules at about 10 pM concentration, and possible steric hindrance effects may prevent any further absorption of PLL when the PLL concentration is further increased.

[00161] The test results show that the sample laminated liquid electrolyte gated CNTFET was a practical electronic device with certain benefits as described herein. For example, the sample device exhibited highly sensitive response to the presence of PLL. The limit of detection (LOD) of the sample devices for PLL was found to be approximately 1 pM.

[00162] It can be expected that the substrates may be replaced with other suitable laminable polymers, such as polydimethylsiloxane (PDMS), poly(methyl methacrylate), polycarbonate, or polyepoxide, as long as the substrate material is compatible with the liquid electrolyte and the possible analyte, and does not negatively interfere with the function of the electrical components of the device. Suitable substrate materials may also include organic materials, glass, ceramic materials, metallic materials, foils, or the like.

[00163] It is also expected that the semiconductor material may be replaced with other suitable semiconductor materials that have an electrical property that is modulatable by a modulating substance such as a liquid electrolyte or an analyte in the proximity to the semiconductor. The modulatable electrical property may be

modulatable by any suitable modulation mechanism including conductance modulation, capacitance modulation, Schottky barrier modulation, charge mobility modulation, electrostatic gating, or the like. For example, organic or inorganic semiconductors in thin film, nanowire, or particulate forms may be suitable replacement material for the semiconductor.

[00164] It is expected that the electrodes may be formed of any suitable conducting materials including organic, inorganic, or metallic materials in any suitable form.

[00165] Both the conducting and semiconducting materials may have nano-sized or micro-sized structures, which may in the form of films, particles, flakes, wires, rods, tubes, or the like. The conducting or semiconducting materials may include carbonaceous materials.

[00166] Embodiments of the electronic devices described herein may also be used as, or in, various other devices such as memory devices, or batteries and other charge storage devices.

[00167] Of course, the above described embodiments are intended to be illustrative only and in no way limiting. The described embodiments are susceptible to many modifications of form, arrangement of parts, details and order of operation. The invention, rather, is intended to encompass all such modification within its scope, as defined by the claims.

WHAT IS CLAIMED IS:

1. A device comprising:

a first member comprising

a first substrate comprising a first laminable material and defining a fluid channel for receiving a fluid, and

first and second electrodes formed on said first substrate and separated by said fluid channel, each of said first and second electrodes comprising a conducting structure; and

a second member comprising

a second substrate comprising a second laminable material, and

a semiconductor formed on said second substrate and comprising a structure having a modulatable electrical property;

wherein said second member is bonded to said first member, with said semiconductor bridging said first and second electrodes across said fluid channel and said structure of said semiconductor being proximate to said fluid channel to allow said fluid received in said fluid channel to be in proximity to said structure of said semiconductor for modulating the electrical resistance between said first and second electrodes through said semiconductor.

2. The device of claim 1, wherein said semiconductor comprises Si, Ge, ZnO, CuO, TiO₂, SnO₂, graphene, carbonaceous material, organic semiconductor, or inorganic semiconductor.
3. The device of claim 1 or claim 2, wherein at least one of said semiconductor and said first and second electrodes comprises a tube, wire, flake, film, or particle.
4. The device of any one of claims 1 to 3, wherein said semiconductor consists essentially of one or more single-walled carbon nanotubes.
5. The device of each one of claims 1 to 4, wherein each of said first and second electrodes consists essentially of one or more carbon nanotubes.

6. The device of any one of claims 1 to 5, wherein each of said first and second substrates comprises polydimethylsiloxane, poly(methyl methacrylate), polycarbonate, polyepoxide, organic substrate, glass or ceramic substrate, metallic substrate, or foil.
7. The device of claim 6, wherein each of said first and second substrates consists essentially of polydimethylsiloxane.
8. The device of any one of claims 1 to 7, comprising a third electrode formed on said first substrate for applying a modulating voltage to an electrolyte received in said fluid channel.
9. The device of any one of claims 1 to 8, comprising an electrolyte disposed in said fluid channel, said electrolyte comprising a liquid, a gel, or a solid.
10. The device of any one of claims 1 to 9, wherein said second member comprises a solid electrolyte adjacent said semiconductor and a gate electrode for applying a modulating voltage to said solid electrolyte.
11. The device according to any one of claims 1 to 10, wherein each one of said first and second substrates comprises a metal coating or ceramic or glass nanoparticles, said metal coating or ceramic or glass nanoparticles providing a barrier towards moisture ingress into said device.
12. A process for forming an electronic device, comprising:
 - providing a first member comprising a first substrate and first and second electrodes formed on said first substrate, said first substrate defining a fluid channel for receiving a fluid, said first and second electrodes separated by said fluid channel;
 - providing a second member comprising a second substrate and a semiconductor formed on said second substrate, said semiconductor having a modulatable electrical property; and
 - bonding said first member to said second member, with said semiconductor bridging said first and second electrodes across said fluid channel, said semiconductor being proximate to said fluid channel thus allowing the

electrical property of said semiconductor be modulated by the presence of said fluid in said fluid channel.

13. The process of claim 12, wherein said semiconductor comprises a nanostructure or a microstructure.
14. The process of claim 12 or claim 13, wherein at least one of said semiconductor and said first and second electrodes comprises a tube, wire, flake, film, or particle.
15. The process of any one of claims 12 to 14, wherein said semiconductor comprises Si, Ge, ZnO, CuO, TiO₂, SnO₂, graphene, carbonaceous material, organic semiconductor, or inorganic semiconductor..
16. The process of any one of claims 12 to 15, wherein each of said semiconductor and said first and second electrodes consist essentially of one or more carbon nanotubes.
17. The process of any one of claims 12 to 16, wherein at least one of said first and second substrates comprises polydimethylsiloxane, poly(methyl methacrylate), polycarbonate, polyepoxide, organic substrates, glass or ceramic substrates, metallic substrates, or foils.
18. The process of any one of claims 12 to 17, wherein said bonding comprises laminating said first and second members.
19. The process of any one of claims 12 to 18, wherein said first member comprises a third electrode formed on said first substrate for applying a voltage to said fluid received in said fluid channel.
20. The process of any one of claims 12 to 19, wherein said second member comprises a solid electrolyte adjacent said semiconductor and a gate electrode for applying a modulating voltage to said solid electrolyte.
21. The process of any one of claims 12 to 20, comprising disposing an electrolyte in said fluid channel.

22. The process of claim 21, wherein said electrolyte disposed in said fluid channel is a liquid electrolyte.

23. The process of any one of claims 12 to 22, wherein each one of said first and second substrates comprises a metal coating or ceramic or glass nanoparticles, said metal coating or ceramic or glass nanoparticles providing a barrier towards moisture ingress into said device.

24. A method of operating the electronic device of any one of claims 1 to 11, said method comprising:

applying a pulsed voltage signal having a peak voltage to a liquid electrolyte received in the fluid channel of said electronic device, wherein a continuous application of said peak voltage to said liquid electrolyte may cause an electrolysis reaction to occur in said liquid electrolyte, said pulsed voltage signal having a pulse profile selected to prevent said electrolysis reaction in said liquid electrolyte; and

detecting an electrical signal from at least one of the first and second electrodes of said electronic device in response to application of said pulsed voltage signal.

25. A method of operating an electronic device formed according to the process of any one of claims 12 to 23, said method comprising:

applying a pulsed voltage signal having a peak voltage to a liquid electrolyte received in the fluid channel of said electronic device, wherein a continuous application of said peak voltage to said liquid electrolyte may cause an electrolysis reaction to occur in said liquid electrolyte, said pulsed voltage signal having a pulse profile selected to prevent said electrolysis reaction in said liquid electrolyte; and

detecting an electrical signal from at least one of the first and second electrodes of said electronic device in response to application of said pulsed voltage signal.

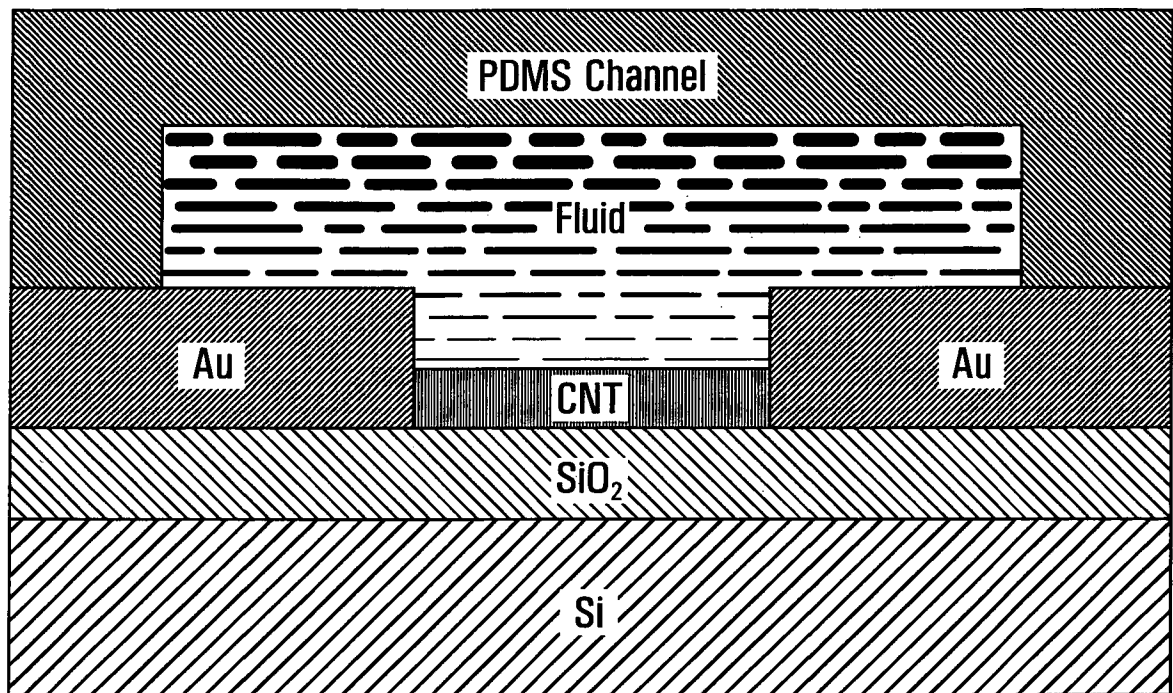
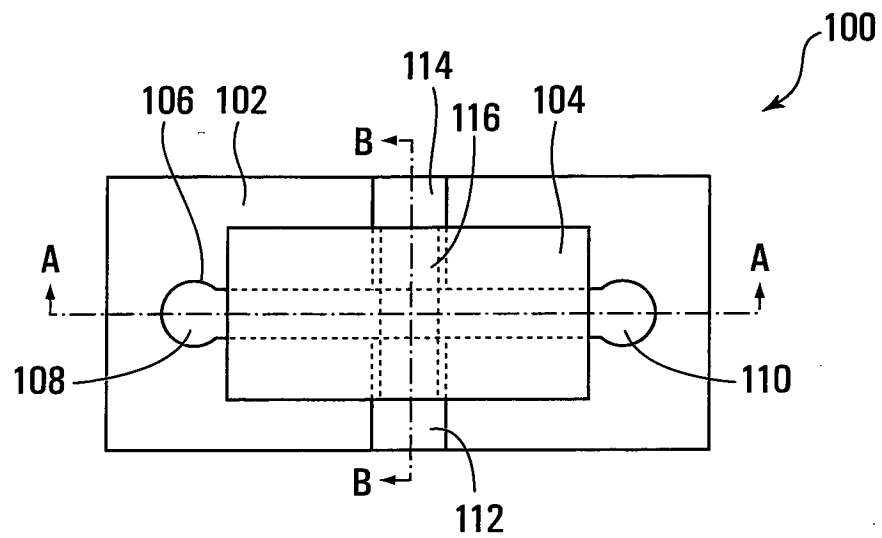
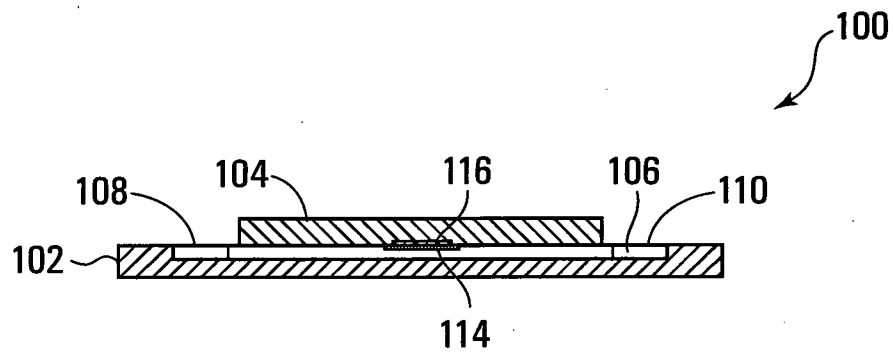
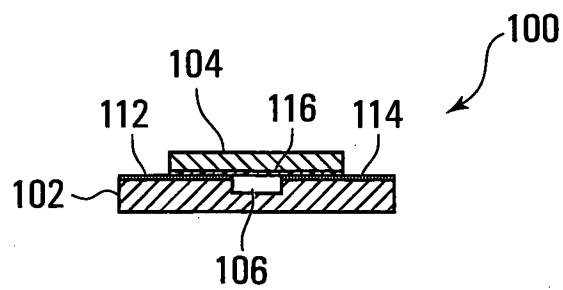
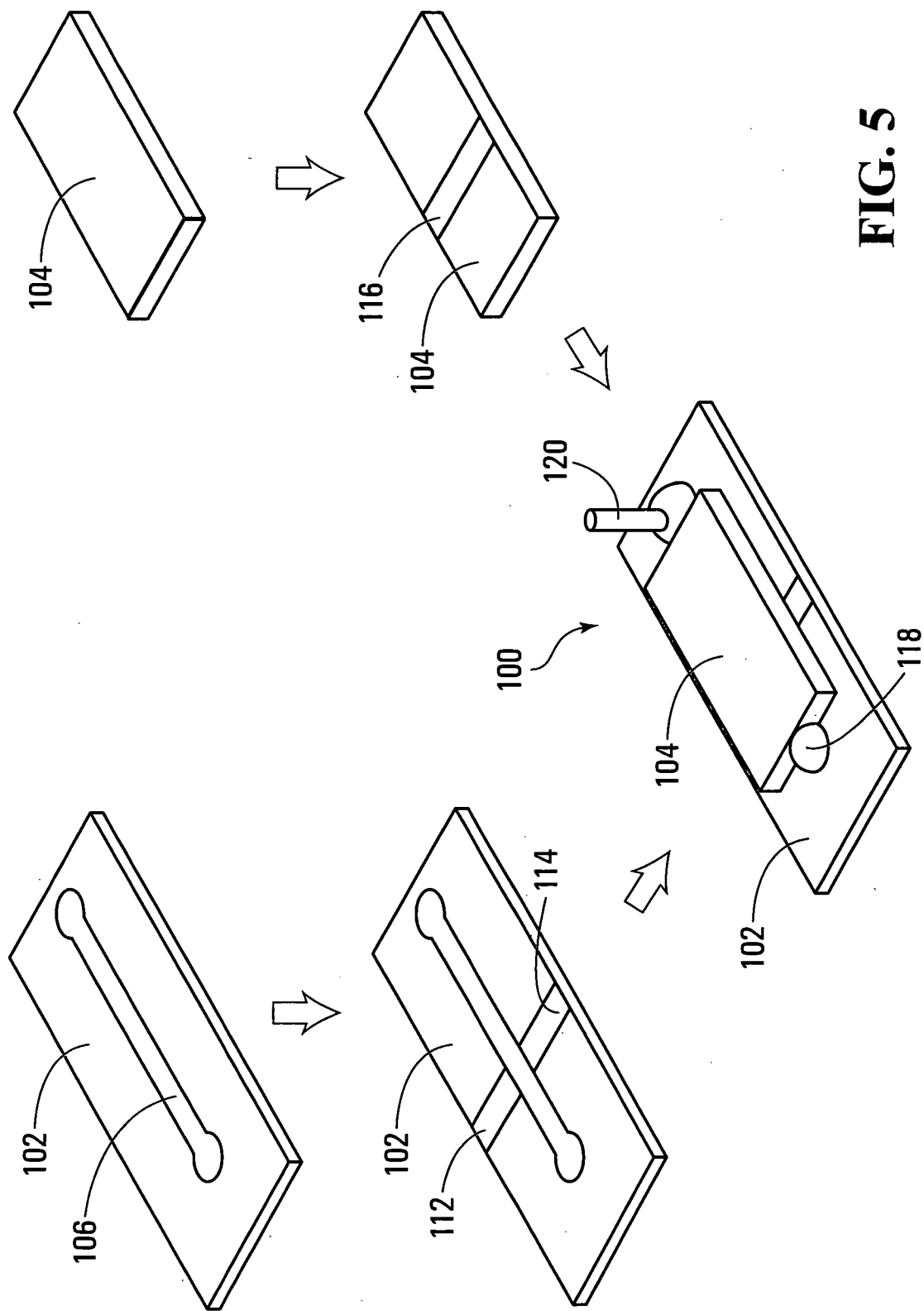


FIG. 1
Prior Art

**FIG. 2****FIG. 3****FIG. 4**



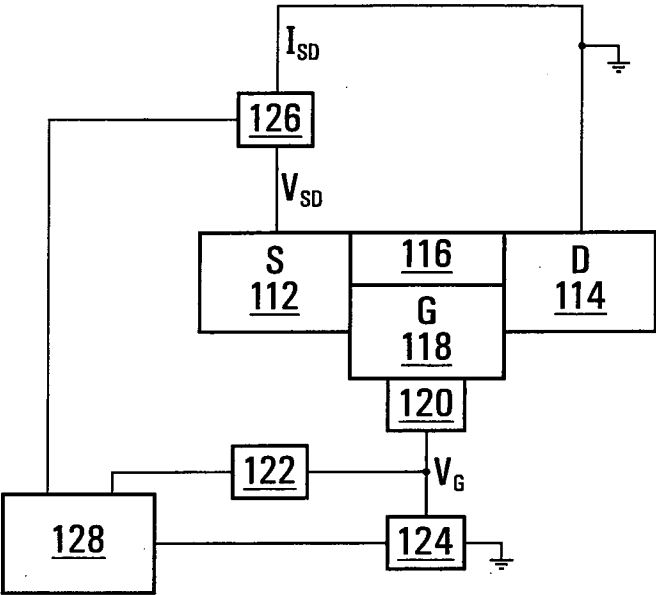
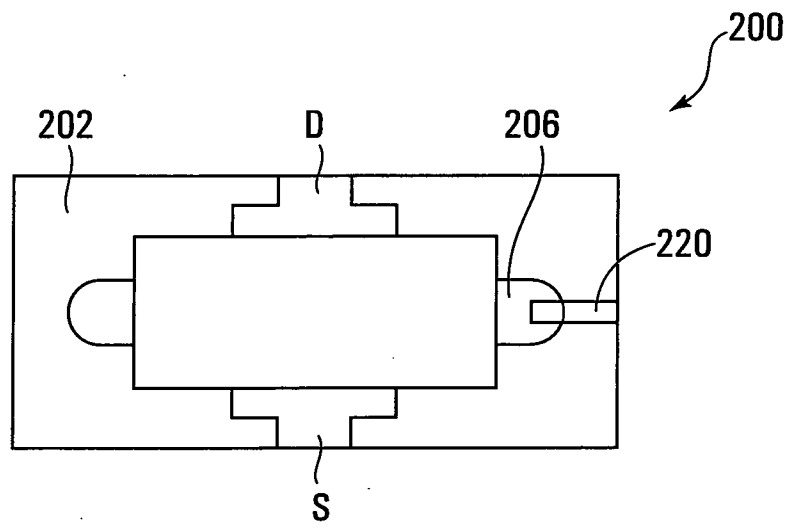
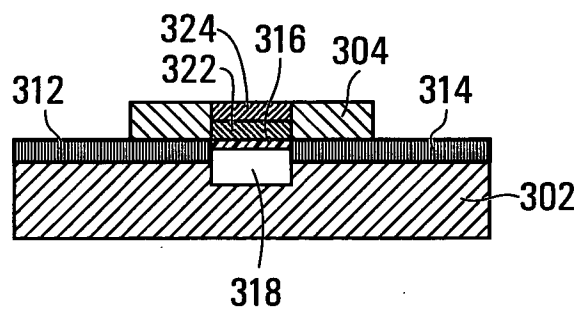


FIG. 6

**FIG. 7****FIG. 8**

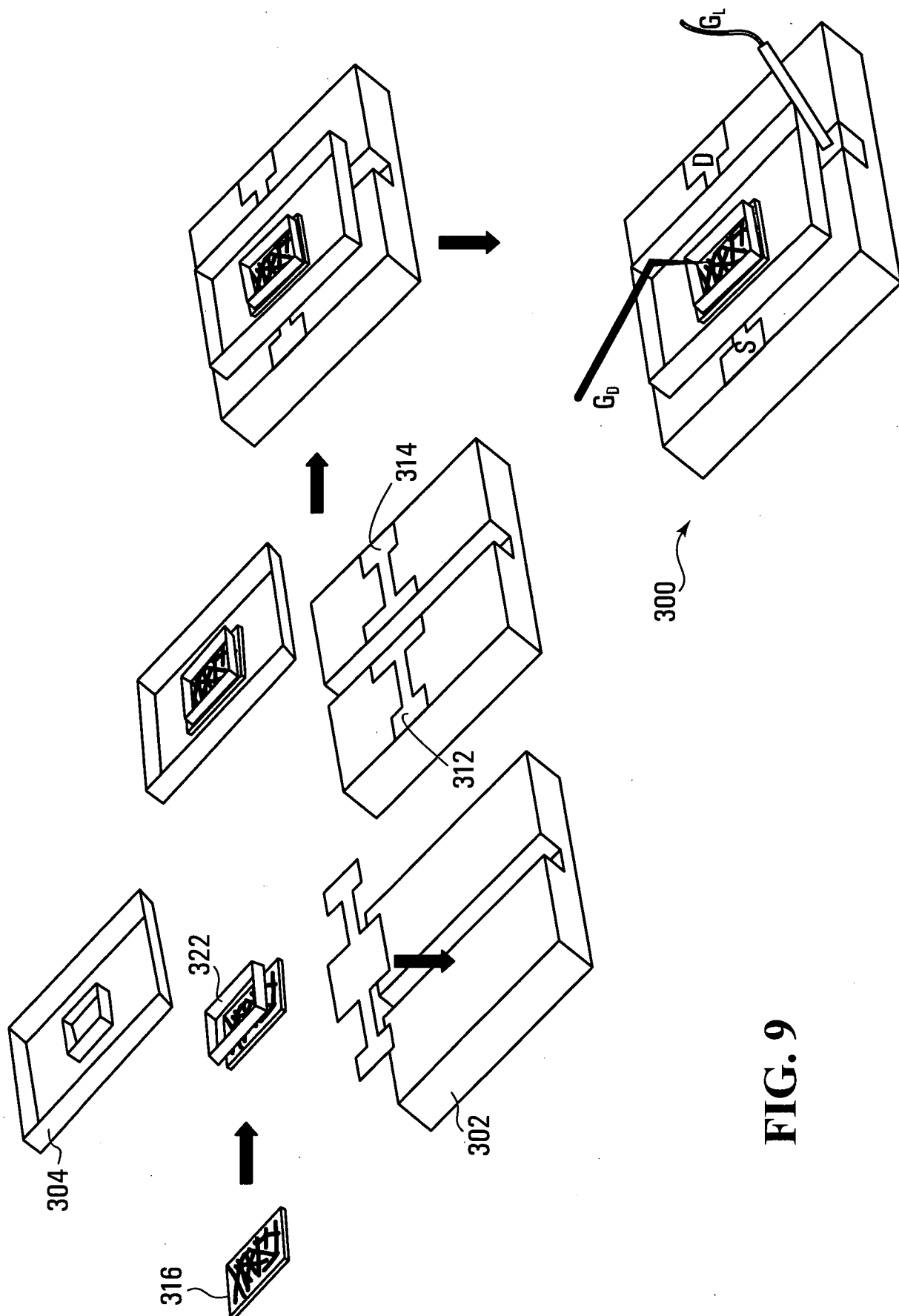
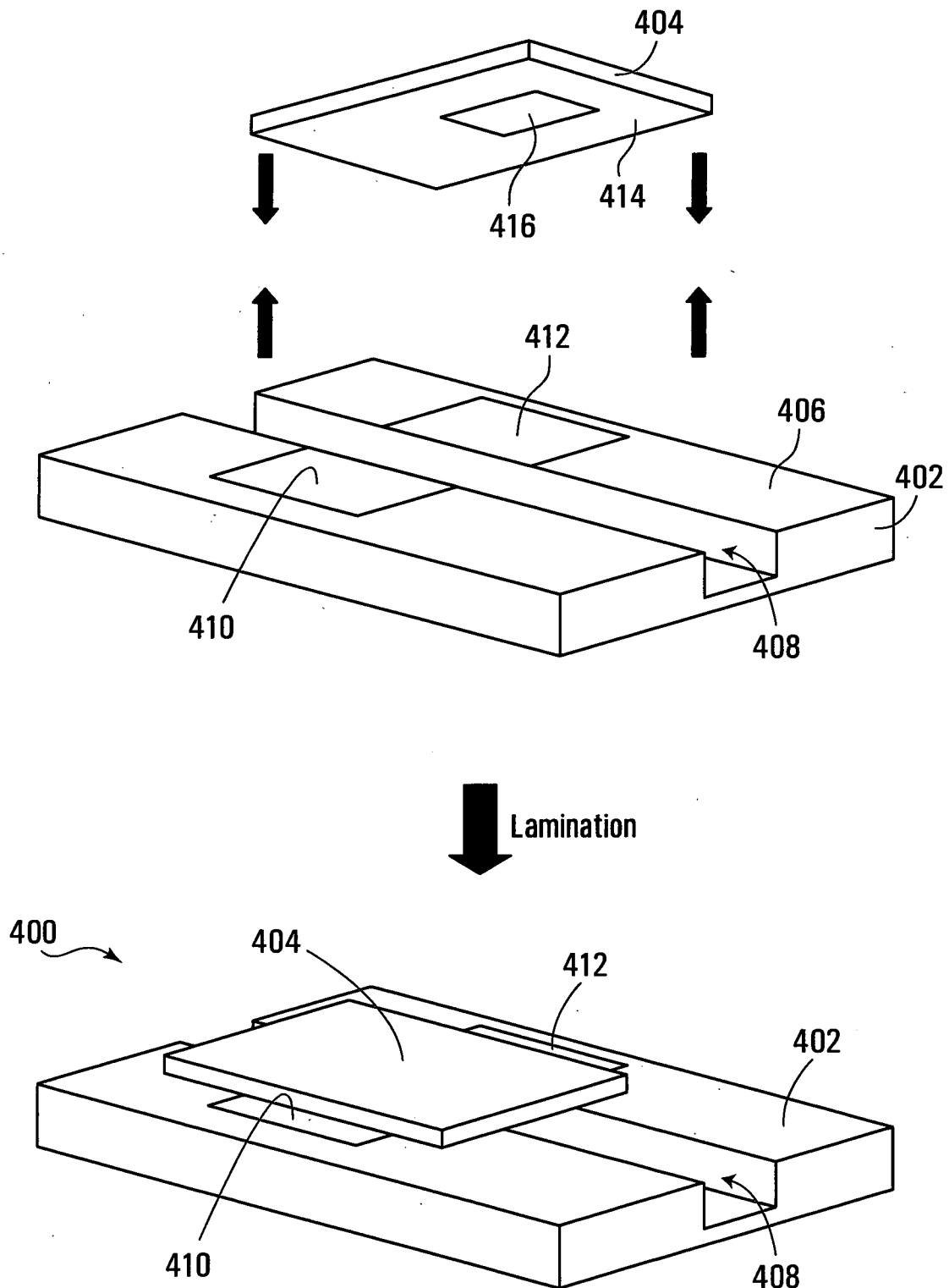
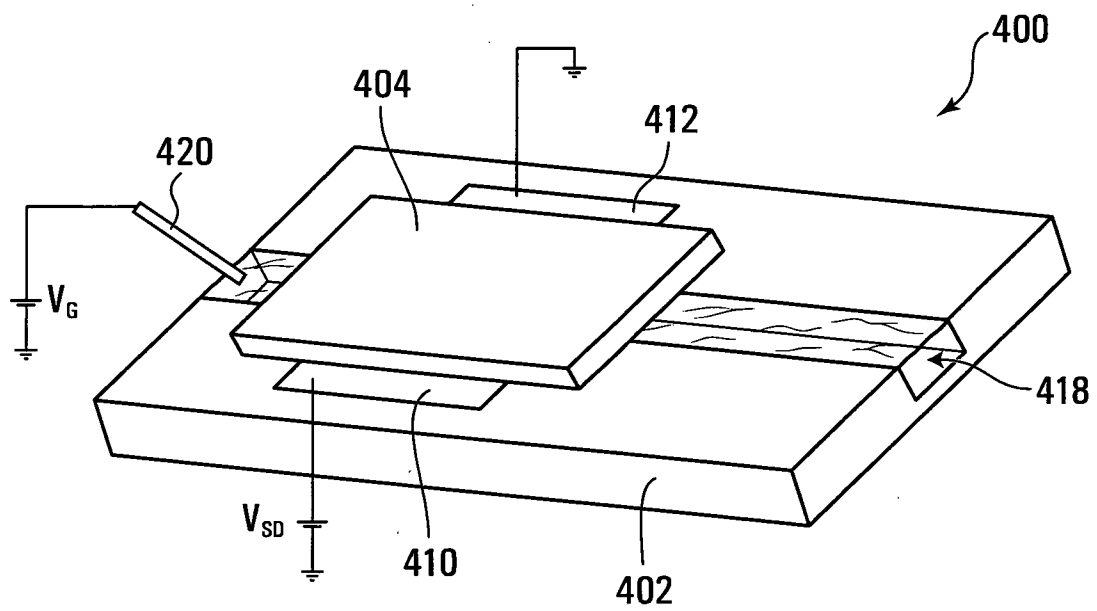


FIG. 9

**FIG. 10**

**FIG. 11**

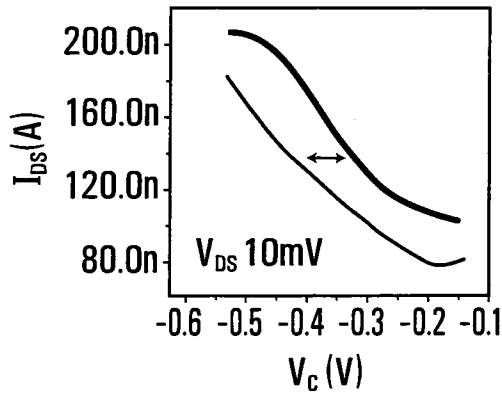


FIG. 12

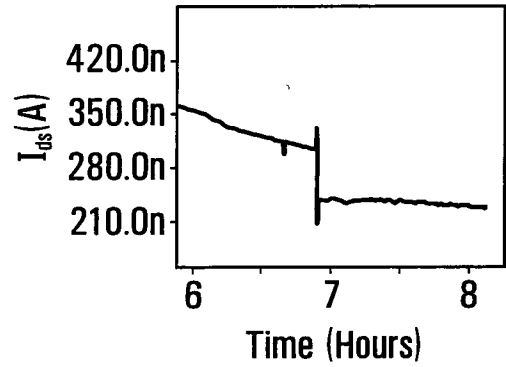


FIG. 13

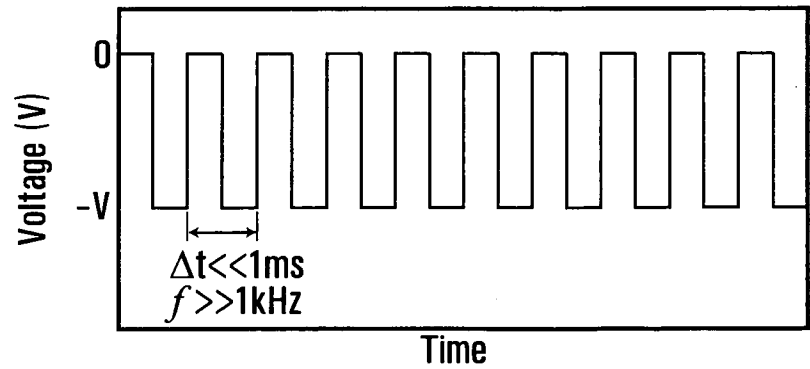
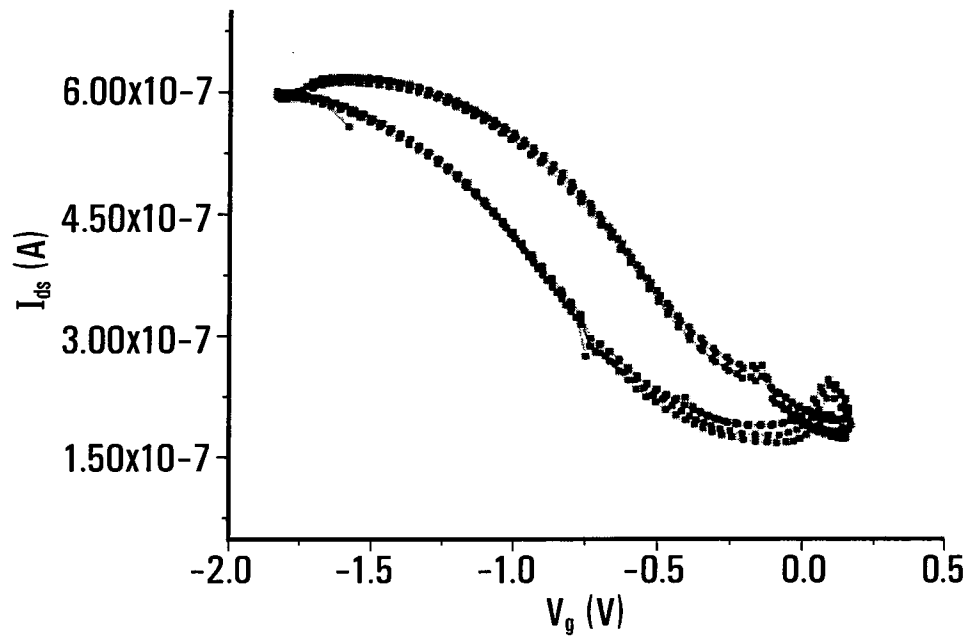
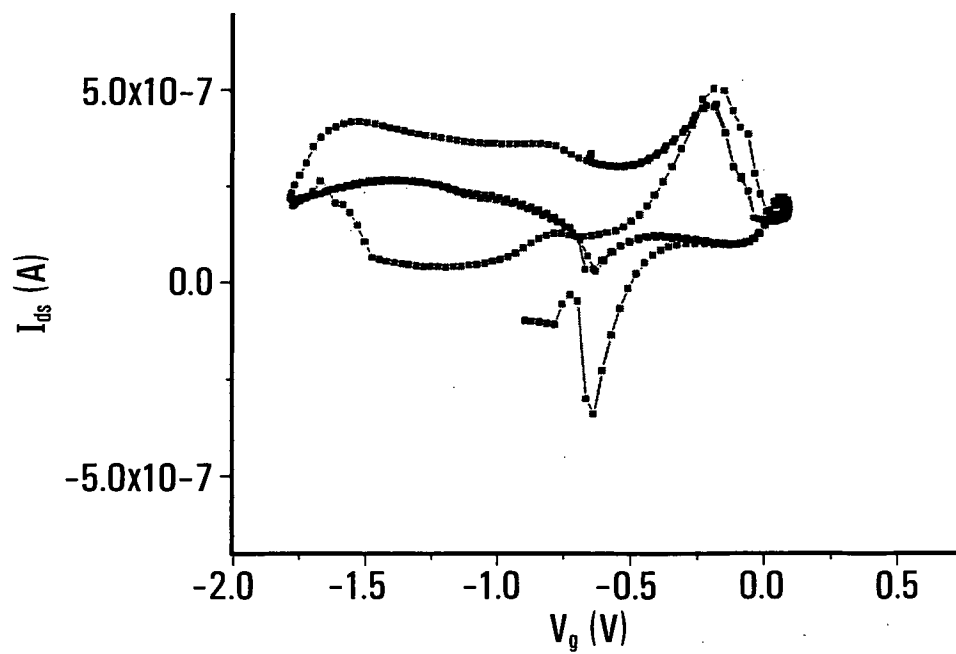


FIG. 14

**FIG. 15****FIG. 16**

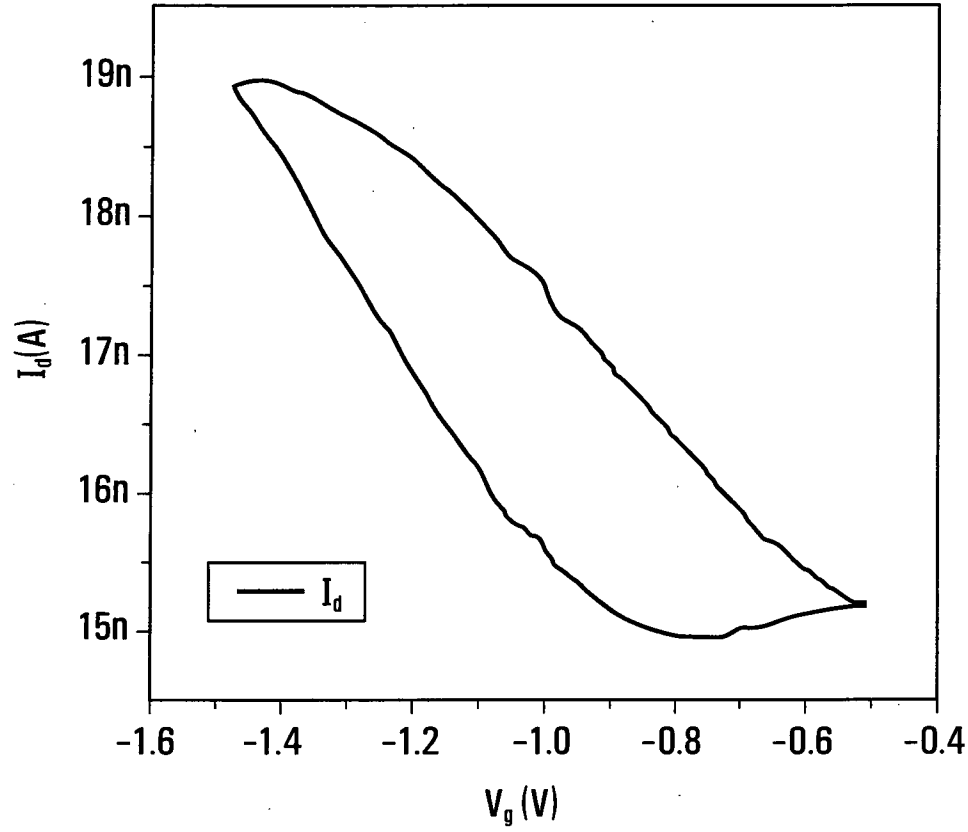


FIG. 17

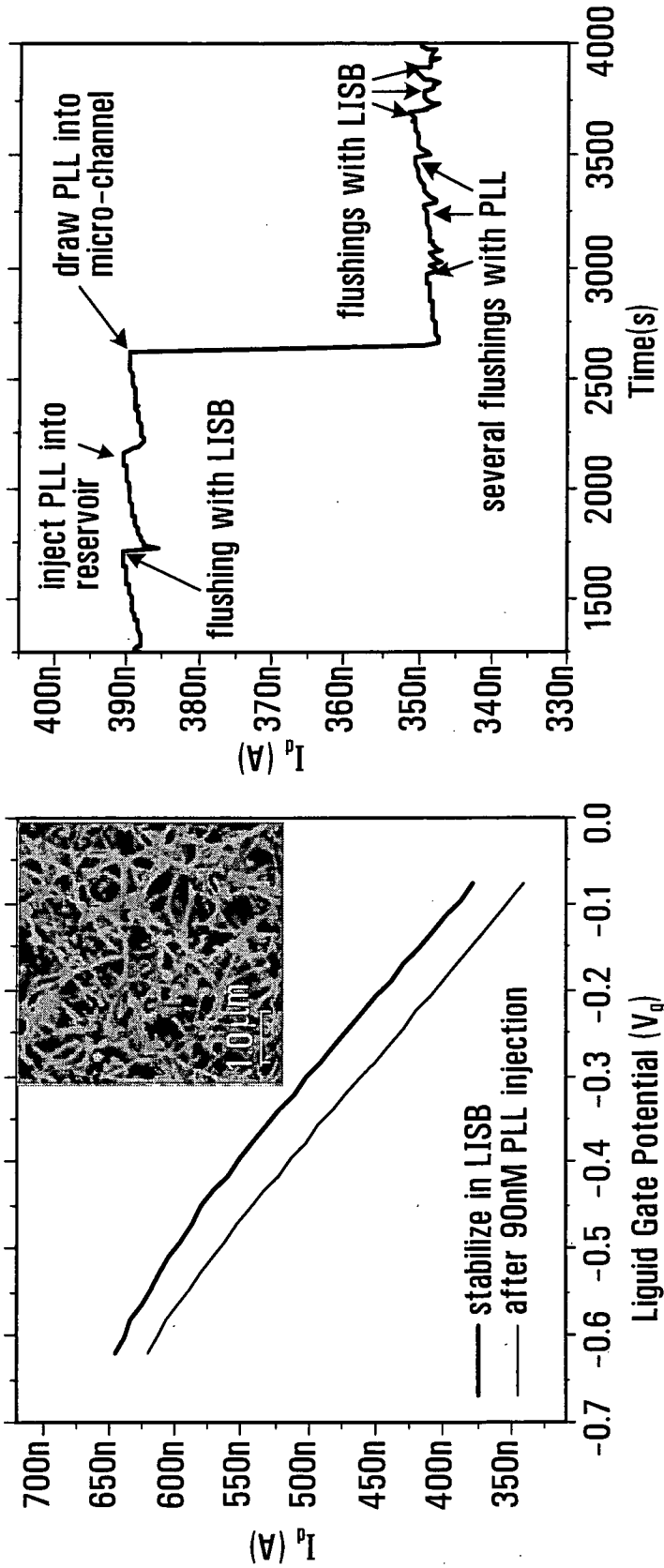
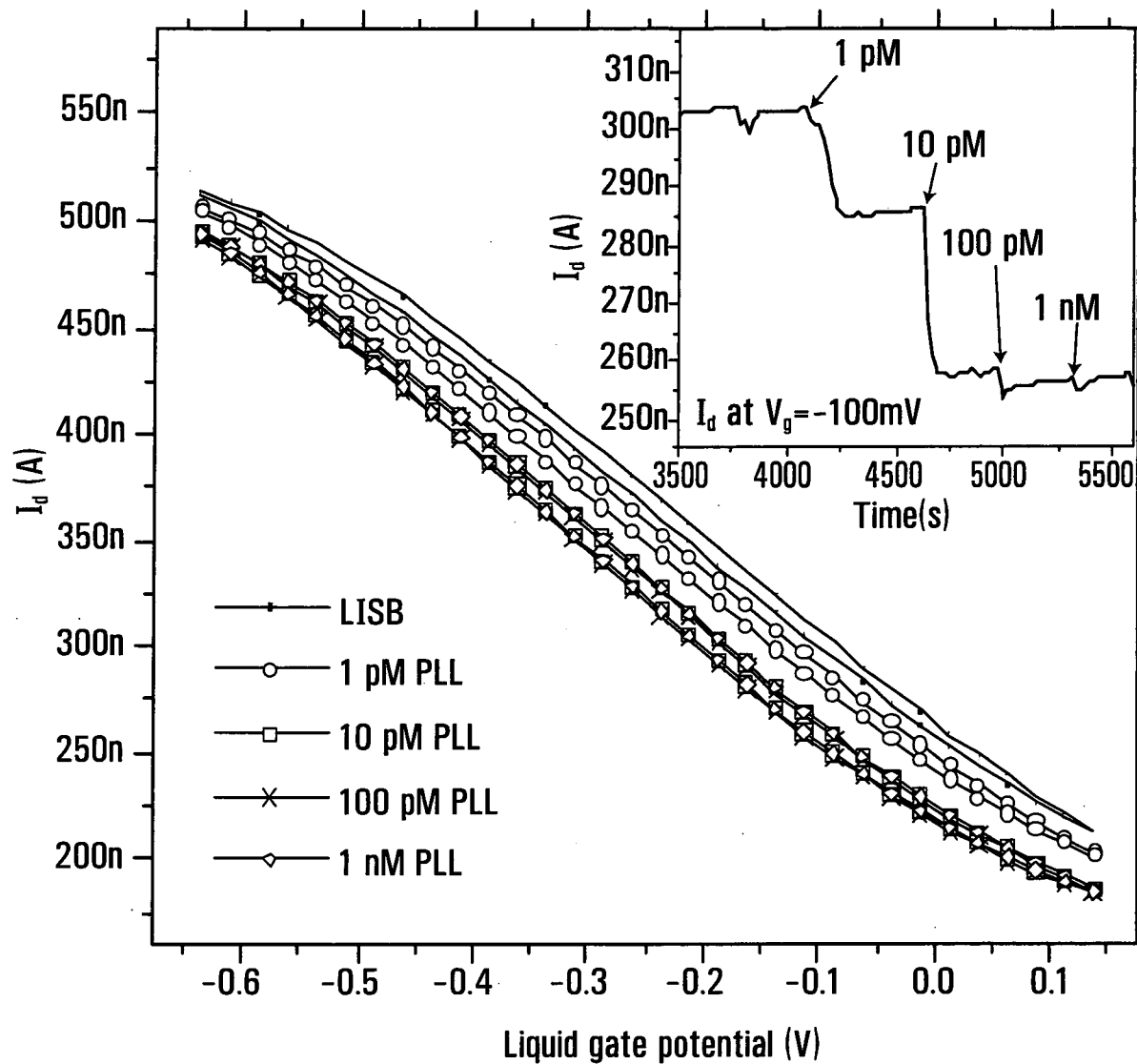


FIG. 18

FIG. 19

**FIG. 20**

INTERNATIONAL SEARCH REPORT

International application No.

PCT/SG2009/000350

A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl.

H01L 29/66 (2006.01)

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)

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)

EPODOC & WPI: Limit H01L or B81B & keywords: Wafer, substrate, base, matrix, laminate, mould, cavity, recess, hollow, chamber, groove, channel, trench, fluid, liquid, electrolyte, gel, biomolecule, solvent, analyte, flow, FET, transistor, memory, electrode, contact, terminal, nanocrystal, nanoparticle, nanodot, nanowire, nanorod, nanocomposit, nanostructure, nanotube, nanofiber, nanofilament, nanowhisker, nanofiber, nanolayer, CNT, nanofibre, modulate, change, alter, vary, resistance, conduct, current, voltage, capacitance, charge, electrostatic-gating, potential)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Zhou et al., "Supported lipid bilayer/carbon nanotube hybrids", Nature nanotechnology, March, 2007, vol. 2, pp. 185-190 Pages 185-186, 189-190, Fig. 1	1-7, 9-18, 20-25
X	US 2007/0218610 A1 (Lim et al.) 20 September 2007 Paragraphs 0075, 0077-0078, 0084-0089, 0094, 0098, Figs. 3, 8A, 9	1, 2, 6-13, 15, 17, 19-25
X	Cui et al., "Nanowire nanosensors for highly sensitive and selective detection of biological and chemical species", Science, Vol. 293, pp. 1289-1292, 17 August, 2001	1-25



Further documents are listed in the continuation of Box C



See patent family annex

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
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"E" earlier application or patent but published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search
02 November 2009

Date of mailing of the international search report
17 NOV 2009

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/SG2009/000350

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report				Patent Family Member			
US	2007218610	CN	1461350	EP	1392860	KR	20020082357
		KR	20020090734	KR	20030048178	US	7235389
		US	2003102510	US	2008093229	WO	02086162
Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.							
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