A Review of Carbon Nanotubes Field Effect-Based Biosensors

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ABSTRACT Field effect-based biosensors (BioFETs) stand out among other biosensing technologies due to their unique features such as real time screening, ultrasensitive detection, low cost, and amenability to extreme device miniaturization due to the convenient utilization of nanoscale materials. Nanodevices pave the way for the detection of tiny biomolecules and minute concentrations of analytes as they are ultrasensitive to surface charge modulation, allowing for better point-of-care screening of various life-threatening infectious diseases. Semiconducting carbon nanotubes (sc-CNTs) are exceptionally promising for FET-channel integration to replace bulky silicon technology beyond the dimensions of the short channel effects for their 1D ultrathin structure, superior electronic features, and biocompatibility. However, performance of CNTFET biosensors is influenced by the inhomogeneous interface between sc-CNTs and metallic source and drain electrodes. This article reviews recent studies on CNTFET biosensors, morphology of these devices and the cause-and-effect of the interface issues between sc-CNTs and metallic electrodes. Finally, future outlook on suggested technology to improve the performance of such CNTFET devices is presented.

INDEX TERMS BioFETs, biomolecules, biosensors, carbon nanotubes, contact resistance, metal electrodes.

I. INTRODUCTION

The outbreak of a series of highly infectious life-threatening diseases, including the most recent 2019 novel Coronavirus disease (COVID-19), Ebola virus, Middle East respiratory syndrome coronavirus (MERS-CoV), Poliovirus, and Zika virus in different parts of the world, is an indication that the current laboratory screening and diagnostic techniques are slower in keeping up the pace with the outbreak of these diseases. Viruses depend on latent parasitic mechanism of reproduction; once infecting, they dictate the host (i.e. a cell; human cell, or even a bacteria) to make clones or replica of progeny genomes in a steady state. Diseases develop specific to the various types of viruses [1]. Hence, the need for early diagnosis of diseases by highly sensitive and selective biosensors which can effectively detect small quantities of analytes in real-time and rapid response is crucial to the wellbeing of mankind.

A biosensor is defined by Thévenot et al. (2001) as a “self-contained integrated device, which is capable of providing specific quantitative or semi-quantitative analytical information using a biological recognition element (biochemical receptor) which is retained in direct spatial contact with an electrochemical transduction element” [2].

In general, biosensors can be classified into two categories based on the recognition element (receptors) and the transducer as shown in Fig. 1. The biological recognition system translates information from the biochemical domain, usually an analyte concentration, into a chemical or physical output signal with a defined sensitivity. The main purpose of the recognition system is to provide the sensor with a high degree of selectivity for the analyte to be measured [2]. The other important part of the sensor is the transducer. The transducer serves to transfer the signal from the output domain of the recognition system, usually, to the electrical domain. The transducer part of a sensor is also called a detector, sensor or electrode, but the term transducer is preferred to avoid confusion [3].
Electrochemical transduction based biodevices are the most widely exploited so far because of their simple principle of measurement and lower cost. Electrochemical transducers include: voltammetric, amperometric, potentiometric, conductimetric, impedimetric, and semiconductor field-effect. The latter measures the modulation of conductance, changes in the current, potential accumulation or charge accumulation, electrical impedance, and the current or the potential across a semiconductor channel in response to a binding process at the gate surface [4].

Field effect-based biosensors (BioFETs) have been used for numerous bio-applications to detect different biomolecular targets. These targets are typically important biomarkers for clinical diagnosis of diseases, such as cardiac diseases, kidney injury, diabetes, cancers, inflammatory, and infectious diseases. Other potential applications include viruses or bacteria detection for infectious disease diagnoses such as AIDS and hepatitis B, as well as other important bio-analysts, such as metabolites. With these developments, field-effect transistor (FET) based biosensors have been identified as a good candidate for next generation point-of-care testing (POCT) [5].

FET devices typically consist of a source, a drain, and a gate terminal where modulation of the current ($I_D$) in the semiconducting channel (i.e. conventionally Silicon as in MOSFET devices) is due to the effect of the electric field generated by the voltage at the gate ($V_G$) and the voltage applied between the source and drain ($V_{SD}$) terminals. Gate voltage can either be channeled through bottom-gating (back-gate FET) or top-gating (top-gate FET) [6]. Contemporary FET-based biosensors incorporate nanomaterials as channel sensing membrane, with some of them such as carbon nanotube (CNT), graphene, MoS$_2$, and SiNWs receiving more attention.

Other FET-based biosensors feature the implementation of organic semiconductor (OSC) materials as sensing membranes. This subtype of organic transistors is called electrolyte-gated organic field effect transistors (EGOFET) in which the organic semiconducting channel is capacitively coupled with the gate electrode by means of an electrolytic solution [7]. Examples of OSC recently reported in the literature include poly(N-alkyldiketopyrrolopyrroledithiopheno[3,2-b] thiophene) [8], poly-3-hexylthiophene (P3HT) [9], poly(2,5-bis(3-tetracetylthiophen-2-yl)thieno[3,2-b]thiophene) [10], pentacene [11].

For the last two decades, researchers have been exerting efforts, tailoring methods and developing strategies for the integration of biomolecules and nanomaterials in an endeavor to produce ultrasensitive biosensors. The integration of charged molecules (biomolecules) with nanoobjects, and inorganic/organic nanohybrids with semiconductor filed effect-based biosensors (BioFETs) certainly promises high potential for label-free and real time biosensing [12]. Various bio-probes can be anchored to the sensor’s channel for the detection of biological analytes achieving high sensitivity and selectivity. For this, channel material is a critical factor for FET-based biosensor performance [13].

Since the first demonstration of individual molecule semiconducting single-wall carbon nanotube field effect transistor device by Tans et al. in 1998 [14], and in the same year, the fabrication of a single-wall and multi-wall carbon nanotube field transistor by Martel et al. [15], carbon nanotubes received more attention as channel nanomaterial due to their excellent mechanical, thermal and electrical features. Amongst a multitude of applications (i.e. in electromagnetic devices, integrated circuits, electric motors...etc.), CNTs are also used in biosensing applications such as a sensing channel in FET biosensors (BioFETs) for the transduction of biochemical reactions for the detection of various analytes and biomolecules.

II. CNT AS BioFET SENSING CHANNEL

As scaling of silicon technology is becoming increasingly challenging, intense research on nanomaterials to complement or replace silicon has become essential [16]. In comparison to silicon-based FETs, CNTFETs have quasi-ballistic transport at low voltage, higher transconductance, higher drive current, higher average carrier velocity, lower heat dissipation and higher switching speed, and can accommodate higher $k$ gate dielectric [17]. The one-dimensional (1D) structure and ultra-thin body which minimizes the short channel effects while simultaneously achieving high carrier transport are features claiming CNT as the potential channel material for future high-performance scaled technology [18]. In addition, CNTs are extremely sensitive to surface modification, even by a single biomolecule, since they consist only of surface; there are no bulk C-atoms [19]. Compared to devices made of microscale materials or bulk materials, reduced dimensionality and larger surface/volume ratio of nanomaterials is believed to be the factor that leads to better
sensitivity towards any biomolecular reactions that takes place on its surface [20], [21].

CNTs are majorly divided into two groups: single-walled carbon nanotubes (SWCNT) and multi-walled carbon nanotubes (MWCNT). The intrinsic properties of CNTs are unique, depending on the chiral indices of the graphene sheet. This dependence is nonmonotonic, and the CNTs can have distinct band gap energies which principally determine the behavior and type of the CNTs as either metallic or semiconducting. Semiconducting CNT is sensitive to its environment and varies significantly with surface adsorption of various chemicals and biomolecules and hence able to detect various biological species such as DNA, proteins, antibodies, living cells and single molecules [22], [23]. Typical CNTFET with top gate and back gate structure is illustrated in Fig. 2.

A. CNT CHANNEL MORPHOLOGY

The optimum structural device parameters such as CNT diameter ($D_{CNT}$), number of aligned nanotubes ($N$) with uniform internanotube spacing ($S$), and oxide thickness ($T_{ox}$), channel length, in addition to the selection of operating supply voltage, are all important in the design of CNTFET devices.

Channel morphology of CNTFETs depends on the fabrication of the device; a single-tube channel implementing a single CNT bridging S-D electrode, or multi-tube channel implementing aligned CNTs bridging the S-D electrodes, or a thin-film channel implementing intercrossing CNT chains bridging S-D electrodes. A single nanotube FET is more effective in biosensing, but it is difficult to control reproducibility of the device due to variations in diameter, chirality, and semiconducting properties from one CNT to another. Intercrossing CNT network FET-based biosensors are less effective, however, due to the manifold entangled path that the current flows through to reach from source to drain. However, variability in this type depends on the density of the nanotubes suspended in between the source and drain electrodes [19]. One third of single-walled carbon nanotubes is metallic and two thirds are semiconducting, which makes the chance of forming a continuous bridge of interconnected semiconducting CNTs between the source and the drain electrodes very high [25].

A few methods of CNTs synthesis exist today such as laser ablation (LA) of carbon rods, direct current arc-discharge (AD) between electrodes, or by chemical vapor deposition (CVD) [26], [27]. CVD is the most popular and widely used because of its low set-up cost, high production yield, and ease of scale-up [28], [29]. While the first two techniques are appropriate for large-scale production of CNTs, they cannot be used for self-assembly on surfaces [26]. CVD appears to be more appropriate for direct deposition on specific structures and substrates, though there arise the issues of unselective growth of metallic CNTs along with semiconducting ones, and the more structural defects compared to CNTs produced by AD and LA [26], [27]. Sonication is one technique for sorting CNTs to segregate metallic from semiconducting counterparts, but this method can degrade the quality of the CNTs, since sonication-induced shortening of the CNTs affects their thermo-oxidative and rheological and behavior [30].

Orientaiton of CNTs growth is another important point in CNTFET design and fabrication. Growing CNTs on a SiO$_2$ substrate results in forest-like crisscrossed random network of CNTs, which is less desirable because of the higher resistance of the entangled path that the current has to go through, while growing them on a quartz yields a well aligned CNTs [31]. However, these aligned CNTs have to be deployed on a dielectric substrate such as Si/SiO$_2$ substrate for FET application. There are methods of depositing or transferring CNTs from a platform to another. Polymer transfer method can be used to transfer aligned pre-grown CNTs-on-quartz onto a silicon dioxide substrate, but this comes at the expense of CNTs being contaminated by the polymer which causes traps and thus causes hysteresis [32]. Dielectrophoresis (DEP) is another method used for deposition and alignment of pre-sorted CNTs from a solution into the substrate of choice by bridging CNTs across the electrodes in accordance to the polarizability of an applied alternating current [33]. Deposition of aligned CNTs on a Si/SiO$_2$ substrate using an improved Langmuir Blodgett method of gradually increased surface pressure by cycles of compression/expansion of the CNT film on the water surface was also reported [34].

Gate formation is another intriguing aspect of channel morphology. For CNT-BioFETs, when assays involve a liquid solution with some electrolyte on the top phase of the device, a reference electrode (i.e. Ag/AgCl) is submerged in the electrolyte solution to achieve top gating. When electrolytes are present in the solution, top-gating is also referred to as electrolyte-gating or liquid-gating [6]. Otherwise, the source
and drain electrodes are fabricated on top of the SiO\(_2\) and the Si substrate used as a bottom gate/bottom gate.

In device miniaturization following Moore’s Law, high \(k\) materials have been implemented as dielectric materials to replace bulky silicon dioxide which is normally applied on top of the silicon substrate or by oxidizing the silicon substrate itself. Examples of materials used, among others, are hafnium silicate (HfO\(_2\)Si), zirconium silicate (ZrSiO\(_4\)), hafnium dioxide (HfO\(_2\)), and zirconium dioxide (ZrO\(_2\)) [35, yttrium oxide (Y2O\(_3\)) and lanthanum oxide (La2O\(_3\)) [36]. Gate insulators with high-\(k\) dielectric provide high electrostatic capacitance that exceeds the quantum capacitance of CNTs, which is unattainable by scaling low-\(k\) dielectric films (i.e. SiO\(_2\)) without resulting in large leakage current. Moreover, high-\(k\) materials can achieve quite same capacitance with small thickness compared to bulkier low-\(k\) materials. Indeed, the integration of high-\(k\) dielectric materials with CNTs is the way forward for device miniaturization as they exhibit superior performance in terms of subthreshold swings, high transconductance and mobility at the sub-microscale [23].

### III. RECENT STUDIES ON CNT-BioFETs

In recent years, carbon nanomaterials have been the subject of intense research and seen wide applications in electrochemical sensors and the favorable incorporation of CNTs in field effect transistor-based biosensors is no exception. These nanodevices are biocompatible and serve a dual purpose; as a carrier that supports the immobilization of bio-probes on their surface in what is known as biofunctionalization, and as a transducer that translates the detection of various biotargets into electrical conductivity [37].

In 2018, Lee and coworkers incorporated a binary hybrid of gold and iron oxide nanoparticles to decorate carbon nanotubes (bNP-CNTs), which is utilized as a biosensing channel to read an electrical resistance depending on the target DNA [38]. The bNP-CNTs was magnetically aligned on Pt-IDE for the detection of influenza virus and norovirus achieving limits of detection 8.4 pM and 8.8 pM, respectively. Compared to their previous study in 2014, the conductivity of Au/MNP-CNT is better than that of Au-NP decorated graphene (Au-GRP), where in the latter case, the average resistance of Au-GRP was relatively high, of around 5 k\(\Omega\) [39].

Melzer et al. (2013) fabricated an electrolyte-gated thin film carbon nanotube based filed effect transistor on a flexible polyimide substrate as a selective platform [40]. The channel was formed of a random CNT-network of approximately 15 \(\mu\)m tube density and modified with polymeric ion-selective membranes for the detection of second messengers of cell-cell communication ions (K\(^+\) and Ca\(^{2+}\)). Change of membrane potential was the sensing mechanism that relies on the transduction of the ion activity at the membrane/electrolyte interface whereby the change in the effective gate-potential affects the charge transport in the semiconducting channel. Detection limit was reportedly in the orders of \(\mu\)M. In another study, Magliolo et al. (2015) developed a thin film transistor (TFT) based immunosensor to detect C-reactive protein (CRP) (i.e. a biomarker of infectious and inflammatory protein diseases, including cardiovascular diseases) [41]. The device was structured with an n-type Si back gate, while the channel was formed of a composite of a single carbon nanotube and a copolymer of fluorene-thiophene (F8T2). The channel was functionalized with monoclonal anti-CRP antibody as a bioreceptor layer to achieve selectivity. This TFT had a dynamic limit of detection reported from 0.4 nM to 2.2 \(\mu\)M.

An interesting paper of a quantum simulation study proposing a novel label-free dielectric-modulated CNTFET that employs a zigzag CNT channel with a coaxial gate morphology was presented by Tammersit and Djefal (2019) [42]. This is a new structure whereby the dielectric coaxial gate is enveloping the nanorod CNT leaving a nanogap in between which is to be filled with ssDNA probes, and thus upon hybridization with target DNA, the high density of the hybridized DNA molecules induces a sensitivity behavior realized as a threshold voltage shift difference between pre- and post the hybridization activity. This new structure is interesting but requires in vivo verification.

Majd and Abdollah (2018) developed an aptasensor for label-free detection of ovarian cancer antigen (CA125) [43]. The FET-type sensor was fabricated on a flexible poly-methyl methacrylate (PMMA) with a carboxylated multi-walled carbon nanotube on a multilayer of reduced graphene oxide (rGO) integrated into a liquid-ion gated FET system via surface engineering technique. Gold electrodes were implemented as source and drain and platinum wire as a gate electrode. The carboxylated acid groups of the MWCNTs facilitated the conjugation of the CA125 aptamer onto the MWCNTs via the amide bond formation with the amine groups of the ssDNA. The binding of the antigens with the aptamer probes creates a field-induced response in an indication of the recognition of a target CA125 biomarker with reportedly low concentration of 5.0 \(\times\) 10\(^{-10}\) U/mL [43].

Liu and colleagues (2015) drop-casted a suspension of SWCNT in between gold electrodes of 1 mm gap to fabricate chemiresistive detectors for amine vapors [44]. The CNT was functionalized with cobalt porphyrin to enhance detection of various biogenic amines. Reported limit of detection is below 0.5 ppm of ammonia gas (NH\(_3\)).

Rajesh et al. (2016) reported the fabrication of a single-walled carbon nanotube-based field effect transistor biosensor by employing gold microelectrodes with 3 \(\mu\)m gap on a SiO\(_2\)/Si substrate for the detection of C-reactive protein (CRP) [45]. The SWCNT was functionalized with polyamidoamine (PAMAM) dendrimer with 128 carboxyl groups as anchors. A decrease in source-drain current due to the carbodimide coupling reaction of the protein antibodies and the CRP analyte was the metric of sensitivity. Reported detection limit is \(\sim\) 85 pM.

Barik et al. (2015) fabricated a junctionless dual-gated CNT-based field effect sensor on indium tin oxide (ITO)
Table 1 summarizes recent breakthroughs and state-of-the-art researches that achieved limits of detection ranging from orders of $\mu$M to orders of pM. Except for a few attempts, most of the previous BioFET studies used metallic source and drain electrodes in contact with the semiconducting channel that renders the device to exhibit a Schottky barrier (SB) effect, which is the primary obstacle for the free transport of electrical charges [24], [48], [49]. The contact between metallic electrodes and semiconducting channel amounts to a considerably large resistance arising mainly from two aspects; physicochemical due to the mismatch of the fermi levels resulting from the difference in the work functions [50], [51], wettability [52], [53], and structural inhomogeneity of the two materials in contact [54], and geometrical influenced by some dimensional measures such as contact length [32], [55], contact geometry [54], and even CNT pitch size [24], [56]. The following section discusses the cause and effect of the contact issue between sc-CNTs and metallic electrodes with extrapolation from previous studies and analysis of its effect on certain electronic performance parameters.

### IV. CNT-BioFET Sensing Mechanism

The most widely used biochemical interactions for field effect transistor-based biosensors are DNA probe-target oligos hybridization, antibody-antigen binding, and enzyme-substrate complexing [61]. Field effect biosensors are biochemically and charge sensitive devices, and thus capable of detecting any kind of charge or potential change at or near the conducting channel induced by biochemical reaction or molecular interaction (i.e. adsorption or binding of molecules) [12]. The feature of CNTs being tiny molecules at the scale of nanometer is rendering these devices capable of responding to the presence and the biological reactions of minute biomolecules immobilized on their surface. The biochemical coating glass substrate for the detection of acetylcholine (i.e. a neurotransmitter) achieving a limit of detection 0.37 $\mu$M [46]. The CNT was uniformly doped with polyethylene imine (PEI) to make the CNT act as an n-type source, drain and channel regions. In this study, Abdul Barik and colleagues reported that junctionless CNTFET can improve some FET devices drawbacks, such as lowering high threshold voltage and increasing low on-off current ratio. Basically, this study explains that junctionless FET devices have no pn, n+n and p+p junction between source/drain and the channel leading in turn to low internal contact resistance. A nanocomposite sensing membrane of CH/NiO was deposited on the top gate insulator reportedly to enhance biocompatibility and electrocatalysis of the device.

Chen et al. (2016) developed a liquid-gated field effect transistor-based biosensor based on horizontally aligned single-wall carbon nanotubes for label-free assay of Interleukin-6 (i.e. anti-inflammatory myokine and pro-inflammatory cytokine) [47]. Aligned CNTs were grown on a quartz substrate by means of chemical vapor deposition (CVD) and Au S/D electrodes were defined by means of evaporation with dimensions of 2 mm wide, 100 nm thick and spaced by 200 $\mu$m. A change in the drain current induced by change in the conductance of the transducer due to the binding of the analyte with the immobilized IL-6R constitutes the basis of the sensing mechanism. This immunosensor reportedly had a limit of detection 1.37 pg/mL.

Sharma and Dutta (2018) developed a CNT based enzyme filed effect transistor biosensor on ITO coated glass substrate passivated by a thin insulating layer of intrinsic ZnO for acetylcholine (Ach) detection [23]. Quasi-ohmic contact between the Al source and drain and the CNT channel was observed as the work function of Al is less than CNT. To make ohmic contact, a depinning technique was applied whereby the CNT was doped heavily with K$^+$ ions, thus to bring the fermi level up closer to the conduction band, thereby matching its Fermi level with that of Al. Limit of detection achieved was 0.6 $\mu$M.

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reaction of the probe biological species binding to the target species induces an electrostatic disturbance to the charge distribution on the surface of the carbon nanotubes. This translates into field effect behavior characterized by a change in the threshold voltage of the of the CNT. Conductivity can be monitored for the change in resistance before and after biorecognition reaction depending on the concentration of the analyte [38].

In traditional field effect transistors, applying a gate voltage to a semiconducting channel, depending on the biasing, results in either accumulation or depletion of the majority charge carriers leading to enhancement or depletion effects, hence increasing or decreasing the channel conductance [62]. On the other hand, the electric field generated by the affinity binding or adsorption of charged biomolecules causes the modulation of the charge carriers of the sc-CNT channel, which is observed as a change in conductance or resistance of the nanotube channel [6].

The mechanism of charge transfer between NO$_2$ and the as-considered hole doped (p-type) sc-SWCNT described by Kong and coworkers (2000) using density functional theory can be ascribed to an oxidation of the nanotube whereby the binding of the NO$_2$ with the nanotube is the effect of energy adsorption ($E_a \sim 0.9$ eV) and the transfer of electrons from the sc-SWCNT to a NO$_2$ molecule; thus increasing hole carriers and enhancing conductivity [63]. Biomolecules, in this case, provide gating effect similar to that of applying a voltage potential at the gate terminal. In some chemiresistors; a variation of BioFETs, the gate electrode is excluded in order to simplify the device structure [6], [62].

**FIGURE 3.** I-V characteristics during protein adsorption on SWCNT-FET ambipolar devices (a) Strong gating effect (b) Strong Schottky barrier effect [64].

Charge modulation by binding or adsorption of biomolecules on the active surface of the CNT channel may result in one or more of the following electrical properties: (1) electrostatic gating or surface charge-induced gating, (2) charge transfer between the biomolecule and the nanotube (nanotube doping), (3) or a combination between electrostatic gating and Schottky barrier effect [6], [64], [65]. In one study, the effect of protein adsorption on SWCNT devices exhibiting ambipolar conduction was analyzed by the $I-V$ characteristic curves and it was found, for the majority of the experiments, that the mechanism of the protein biosensing is governed by a combination of electrostatic gating and Schottky barrier effects as illustrated in Fig. 3.  

Tae Hyun Kim (2017) reported that the selective binding of human serum albumin with the monoclonal anti-human serum albumin (m-AHSA) antibody was realized by the induction of an electrostatic gating effect and the modulation of the charge state of the channel conductance of the sc-CNT nanodevice [66]. Nevertheless, intrinsic negative charges in nucleotides are directly transformed into potential variations at the gate-solution interface where the electrical double layer works as a capacitor [20]. Distinctly from the above, the enzymatic based CNT-BioFETs work differently; a reference electrode (i.e. typically Ag/AgCl or Pt) submerged in electrolytic solution acts as a gate electrode and the drain current is modulated in response to the pH value change reflecting a shift in the threshold voltage due to the screening of the enzyme and substrate complexing interaction in the solution [67]. Thus, a key issue is the fabrication of an organic functional nanolayer on a gate electrode that effectively captures the molecules or detects the recognition events at the gate/solution nanointerface.

FET-based biosensors provide a significant number of potential advantages over normal electrochemical sensors such as small size and light weight, label-free assay, quick response, on-chip integration of biosensor arrays, low output impedance, low-cost mass production and portable and compact microanalysis. An intrinsic miniaturization of FET devices and their compatibility with microfabrication processes make them very attractive for integration into microfluidics and micro-analytical devices [12], [20], [68].

However, screening of the analyte charge by the electrolyte ions (Debye screening) is a challenge in BioFETs. Large molecules or macromolecules (i.e. proteins) are difficult to detect since the size of a typical antibody receptor molecules is $10–15$ nm, which is beyond the Debye length of the effective distance for charge detection, that is around $1$ nm [69]. There have been attempts to address this issue by using short nanobody receptors or single-chain variable antibody fragments with achieved detection limit down to sub-picomolar regime [6], [70]. Ionic concentration of the solution is another important factor in controlling the Debye length to ensure that specific binding of macromolecules contribute to sensor response. Increasing ionic strength of the buffer solution 100 folds enhanced specific screening of streptavidin binding to biotin characterized by an increase in the drain current as discussed in [71].

Despite the numerous advantages as aforementioned, CNTFETs have several limitations such as poor reproducibility due to properties variability of CNTs [40], [72], the hysteretic effect observed upon sweeping the gate voltage back and forth [45], the unselective growth of metallic and semiconducting nanotubes with the limited separation techniques [69], and the nonuniform and increased scattering events in CNT-network-based FET devices due to variations in bundle networks and thus the bulk resistance [52]. However, amongst other issues, the contact issue at the interface between the CNT and the metal electrodes is the most challenging and persisting issue reported by most of the studies.
in this type of biosensors. For this reason, the next section is allocated for discussing in detail the background of this issue and the numerous techniques implemented to resolve or mitigate its effect.

V. CNT/METAL INTERFACE ISSUES IN BioFETs

All semiconductor devices have contacts with other components, which may be of the same or different material, in the larger electronic systems. And since all contacts result in relative resistance, it is of paramount importance to analyze the distinctive features of these contacts. In general, they are typically metal-semiconductor contacts, but semiconductor-semiconductor contacts also exist, where both semiconductors can be single crystal, polycrystalline, or amorphous [50].

The first acceptable theory of metal-semiconductor contacts was developed by Schottky in the 1930s. Referred to as Schottky barrier devices in his honor, metal-semiconductor devices are usually used as rectifiers whose current-voltage characteristics is distinctly non-linear. In contrast, a linear or quasi-linear current-voltage characteristic is ideally observable in ohmic contacts. However, practically, not all ohmic contacts have linear current-voltage characteristics. Ohmic contacts must be able to provide the current necessary for the device with only small voltage drop across the contact compared to the voltage drop across the active regions of the device.

![FIGURE 4. Metal-semiconductor contacts according to the simple Schottky model [50].](image)

According to the Schottky theory, semiconductor electron affinity and the metal work function are the factors that determine the barrier height regardless of the semiconductor doping density. Hence, hypothetically, metals of appropriate work function can be implemented to vary the barrier height in order to fabricate any of the three barrier types shown in Fig. 4. It is also evident from Fig. 4 that ohmic contacts correspond to the accumulation type of contact whereby electrons flow freely into and out of the semiconductor with minimal barrier.

In CNT-BioFETs, the small contact area between carbon nanotubes and metal electrodes makes electrical coupling between them extremely difficult. The contact between semiconducting CNT and metal electrode is generally modeled as a Schottky barrier (SB), arising from the mismatch between the Fermi levels of the sc-CNT and the metal electrode [51], [52], [56]. Upon outfitting the sc-CNTs on the metal electrodes, the Fermi energy level of sc-CNTs tend to force align with that of the metal causing bending of the conduction and valence bands of the nanotubes and resulting in a Schottky barrier type of contact shown in Fig. 4-c [25]. This barrier impedes charge transfer between the nanotubes and the metal source and drain electrodes giving rise to what is known as contact resistance ($R_c$).

Contact resistance is a critical factor limiting the performance of CNT-BioFETs [55], [73], [74]. It is the resistance generated at the interfacial contact between the metal electrodes source and drain and the semiconducting CNT carbonaceous material [75], [76]. As depicted in Fig. 5, there are two regions influenced by the interfacial effect of the metal-semiconductor contact.

![FIGURE 5. Internal device interfaces involved in the CNTFETs contact resistance. (Only one contact side is shown) [73].](image)

The first region is the interface between the metal and the nanotube, while the other region is between the coated and the uncoated parts of the nanotube. Different factors can influence each of the contact regions. The first region, between the metal-semiconductor, is affected by the material used for the fabrication, and the intercalation of any interfacial layer between the metal and the nanotube. The second region between the coated and uncoated nanotube can be influenced by the change of the electronic structure of the metal-coated tube portion since the interaction with the metal induces a potential step (barrier). An important contribution to the height of the potential step is the Schottky barrier height $\phi_B$ [73]. The contact resistance $(2R_c)$ collectively from both electrodes (i.e. source and drain) dominates the performance of scaled devices as the channel transport becomes ballistic. Early SWNT transistors were plagued by poor electrical properties of the metal contacts [77]. It is determined by three factors: 1) Schottky barrier height (i.e. difference in work functions); 2) interface quality (i.e. metal-CNT adhesion); and 3) physical contact length ($L_c$) [32]. In conjunction, Ning Yu et al. (2017) investigated the effect of varying the contact length, that is the area of contact between the carbon nanotube and the metal electrode, and their results concluded that larger contact length yield lower contact resistance [55]. In another study, Jinwook Baek and colleagues...
TABLE 2. Contact resistance of CNT-FETs implementing metallic electrodes.

| S-D Contacts | Channel Material | Resistance | ON current | Ioff/Ion Ratio | Transconductance | Reference |
|--------------|------------------|------------|------------|---------------|------------------|-----------|
| Pd           | CNT              | -          | 2 μA       | ~10^4         | -                | [14]      |
| Pd           | CNT              | 40 kΩ      | 10 μA      | 10^3          | 20 μS            | [31]      |
| Cz/Au        | SWCNT & WCNT     | 22.8 MΩ    | -          | -             | -                | [32]      |
| Ti/Pd/Au     | CNT              | 40 ± 20 kΩ | -          | 10^3          | 30 ± 10 μS       | [49]      |
| Pd/Pt/Au     | CNT              | 22 ± 2 kΩ/tube | 120 μA/μm | ~10^1        | 40 μS/μm         | [54]      |
| Au           | CNT              | 25 kΩ      | 97 μA      | 16 × 10^5     | 186 μS           | [56]      |
| Mo           | CNT              | 36 kΩ      | 15 μA      | 10^4          | -                | [63]      |
| Pt           | CNT              | 1 MΩ       | 4 nA       | -             | 10 nS            | [65]      |
| Au           | CNT              | 0.156 MΩ   | ~5 μA      | <             | 3.5 μS           | [72]      |
| Pd           | CNT              | 70 kΩ/tube | 1.33 mA/μm | >10^4       | -                | [85]      |
| Ti           | CNT              | 380 kΩ     | 1.91 μA    | 1.2 × 10^3    | 0.22 μS          | [86]      |

(2017) revealed that contact resistance depends significantly on carriers doping density, as well as contact length [76]. In this study, the contact resistance reportedly dropped from 2 MΩ to below 800 kΩ for semiconducting carbon nanotubes as the contact length increased beyond 10 μm.

Doping of the semiconducting channel to modulate its electrical properties is applicable. It significantly alters the Fermi level of the CNT and thus its work function [78]. In semiconductor microfabrication, it is important to characterize the doping contrast and doping profiles by means of secondary electron (SE) imaging, that is the basis of scanning electron microscopy (SEM), to evaluate and analyze the electrical properties of doped junctions (i.e. homo-or-heterojunctions) [79], [80].

The transport of charge carriers at or through the CNT/metal junction is governed by the work function of the metal electrode. This is the minimum energy required for a charge to be extracted from a medium to a vacuum or, in this case, to be injected to the other medium in contact. The charge conduction at the CNT level is characterized by ballistic transport, but then the resistance to the flow of charges at the heterogenous junction of the CNT/metal contact is characterized by the difference in work functions between the two materials relative to the Schottky barrier height [48], [53], across which a tunneling transport of charges is realized [64], [81]. Peculiarly, the work function of the source and drain electrodes determines the transfer characteristic of the charge carriers of the CNTFET device. An n-type characteristic, p-type characteristic, or ambipolar characteristic may be obtained depending on the source-drain electrodes’ work function property; low, high, or hybrid of low and high work functions at either electrode, respectively [82].

Modelling and characterization of the contact resistance of CNT-BioFET devices is crucial to the improvement of their sensitivity. There have been several methods used to model \( R_c \) or extract its value and the most common of which is the transfer length method (TLM). Also called transmission line method, TLM is a technique used in semiconductor engineering to phenomenologically model the contact resistance between a metal and a semiconductor. The technique involves making a series of metal-semiconductor contacts along one single long nanotube to avoid properties variations from one CNT to another, or an evenly spin coated mat of a thin film CNT, separated by various distances [76].

4-pints probes may be applied by assigning a pair of current probes and a pair of voltage probes across each pair of contacts. Using this method, the resistance between the electrodes is measured either by applying a voltage across the contacts and measuring the resulting current through the various lengths (\( L_{Ch} \)), or passing a current through one electrode to the other and measuring the voltage drop across the transmission length, and accordingly taking the readout of the entire setup resistance as a function of length (i.e. channel length; \( L_{Ch} \)). Thus, the resistance measured is a linear combination (sum) of the contact resistance of the first contact and the second contact, and the sheet resistance of the CNT channel in-between the contacts (i.e. \( R_{Tot} = 2R_C + R_{Ch} \)) [83]. It is important to note that the metal resistance is negligibly much smaller than the contact resistance to be considered. The sheet or channel resistance is dependent on the scattering events in the channel. Shorter channels contribute fewer scattering events, and thus, lower \( R_{Ch} \). In ohmic contacts with ballistic charge transport, the total resistance may approach the quantum limit of the nanotube; \( R_Q = 6.5 \text{ kΩ} \) [32], [83]. For the measured total resistances of the variant lengths, the contact resistance is extrapolated by dragging back to \( L_{Ch} = 0 \) in the transfer length plot, as illustrated in Fig. 6 [76], [84]. Results obtained from TLM can be further verified using Y-function method (YFM), which is a numerical parameters extraction method that combines both the drain current and the transconductance transfer characteristics [49], [83].

Table 2 shows the resulting \( R_c \) and other electrical parameters. Devices with greater \( R_c \) had poorer performance in
TABLE 3. Recent techniques to reduce contact resistance.

| Technique                          | Purpose                                      | Impact                                                                 | $R_{ct}$/Resistivity | Ref. |
|-----------------------------------|----------------------------------------------|------------------------------------------------------------------------|----------------------|------|
| Ion-irradiation                   | Induce defects on CNT                        | Slight improvement for thick contact of Ag with CNT                   | 102 – 91 $\Omega\mu$m | [52] |
| Electron-beam-induced deposition (EBID) | Enhance side bonded contact                  | Increased the van der Waals force of the contact interface between Au/CNT | 7.5 $\Omega$        | [55] |
| End-bonded contact                | SWNT channel abruptly ends at the metal electrodes | Solid-state reaction between deposited molybdenum electrodes with the SWNT formed carbide | 36 $\Omega$         | [77] |
| Self-assembled monolayers (SAMs)  | Feature different dipole moments which impacts directly on their work function ($\phi_m$) | Better alignment of $\phi_m$ with the organic semiconductor energy levels achieved ensuring a more efficient charge injection. | -                    | [87] |
| He+ bombardment followed by annealing | Induce exotic defects in graphene            | Enhanced contacts by forming end-contacts at intrinsic defect locations | 118.28 $\Omega\mu$m lowest for Pd | [92] |
| e-beam lithography followed by O$_2$ plasma etching | Forming hole lattices (antidot arrays) under metal electrode | Introduced edge contact of graphene with Pd/Au | $\sim$ 200 $\Omega\mu$m | [93] |
| Ozone exposure followed by annealing | Improve contact resistance of e-beam evaporated Ti/Au contacts | Reduced specific contact resistance three orders of magnitude | $7 \times 10^7 \Omega$-$cm^2$ | [94] |

Karnaushenko and coworkers (2015) applied thermal annealing process after deposition of the SD electrodes [89]. By this, metallic Ni silicide grows into the nanowires delivering abrupt metal to Si nano-sized Schottky junctions. As an effect of thinning the barrier, enhanced transmissibility through the contact led to higher current densities and contact resistance was therefore reportedly reduced by two orders of magnitude. However, thermal annealing could compromise reproducibility and complicate the process [90].

Bezugly et al. (2017) fabricated two separate biosensors for the detection of avian influenza virus (AIV) subtype H5N1 DNA sequences, one with sc-SWCNT and the other with N-MWCNT bridging a Cr/Au electrode [91]. The initial resistance of the N-MWCNT-based sensor was 40.1 M$\Omega$, while for sc-SWCNT-based sensor it was 22.8 M$\Omega$. The lowest reliably detected concentration of DNA T was 2 pM for sc-SWCNT and 20 pM for N-MWCNT sensor. The better sensitivity of the sc-SWCNT is believed to be attributed to the lower contact resistance.

Haslam et al. (2018) fabricated Graphene-based FET sensors on Si/SiO$_2$ substrate using photolithography with sputtered gold electrodes for the sensitive detection of Human Chorionic Gonadotropin (hCG), a glycoprotein risk biomarker of certain cancers [85]. Haslam and coworkers used chromium (Cr) as an adhesive layer between graphene and the Au metallic contacts to reduce contact resistance. However, their device suffered non-linearity probably due to Schottky contacts or charge traps. Further optimization of contact resistance was recommended in this study.

However, most FET-based biosensors implement noble metal electrodes as their source and drain. A Schottky type contact is typically established between metal electrodes and semiconducting channel which is undesirable in such biosensing applications. The difference in work functions terms of on current and transconductance. Improved contact resistance over the various studies notably promoted better CNTFET device performance.

FIGURE 6. Extrapolation of contact resistance from the plot of the transfer length method. Adopted from [84].
between the metal electrodes and the semiconducting channel results in high resistance. High resistance has the effect of limiting the on-current, and hence resulting in reduced device performance in terms of limit of detection and sensitivity as aforementioned. This contact resistance may range from kilo-ohms to the excess of mega-ohms [14], [31], [73], [86], [88].

There have been numerous attempts to address the persisting issue of the contact resistance by implementing various techniques. Table 3 summarizes several of these techniques implemented by recent studies to reduce contact resistance, the specific purpose sought, the impact achieved and the resulting contact resistance/resistivity.

VI. FUTURE OUTLOOK
To date, most of the studies reported for the detection of biomolecules implement noble metals as electrode material [95]–[100]. These noble metal electrodes generally require a metal adhesion layer such as Ti, Cr, or Ta. However, these dual metal layers are degradable via galvanic corrosion when placed in contact with the electrolyte. This adds on to the existing metal and nonmetal (semiconductor) inhomogeneous contact and the difference in work functions, which results in Schottky type of contact. Prospectively, by eliminating the Schottky barrier effect between the source contact and source material, the transistor will be capable of delivering more on-current [24]. A good alternative to noble metallic source and drain electrodes is ones made of carbon.

Theoretical studies have predicted that carbon-based conductive materials are advantageous over metal contacts for electrically contacting carbon nanostructures [72], [76], [101]. This is due to the possibility of direct bond formation between the carbon material and the nanostructure, with a character very similar to the internal bonding in the latter, thus ensuring a well-matched bonding network and a good continuity of electronic structure. As another advantage, the relatively small work-function difference among carbon nanostructures helps avoiding changes in the electrically addressed material due to contact doping [101]. In addition to their low cost, carbon-based electrodes involve reduced number of fabrication steps. The use of an adhesion layer involved in noble metal electrodes and the lift-off process is avoided, thus increasing the fabrication yield [102].

Moreover, the use of bilayers of Au/Ti or Au/Cr in conjunction with noble metal electrodes, when in contact with electrolytic environment, form a galvanic couple that may result in corrosion effect, thus affecting their repeatability, reliability, and longevity applications in electrochemical cells. Carbon, on the other hand, comes with a wider stability window than any noble metal, and is chemically very stable. Carbon also exhibits good biocompatibility, low tendency for fouling, and low nonspecific adsorption of biomolecules [102], [103].

Hence, the motivation of using carbonaceous electrodes in contact with sc-SWCNT can be summarized in three main reasons: (1) almost identical work functions, (2) homogeneity of materials, (3) low cost of fabrication and materials. The work function of pyrolytic carbon is 4.61 eV, similar to that for glassy carbon and graphite [104], [105], while the calculated work function of sc-CNT is reported by [106] as 4.73 eV, and by [107] to be within 4.70 ± 0.03 eV. As for the second reason, using carbon-based electrodes is predicted to yield a natural bonding between the homogeneous structures of the CNT channel and both of its end contacts at the source and drain. By this, exfoliation and traps normally generated due to the heterogeneity of the metallic electrodes and the semiconductor channel is to be avoided, thus less leakage current and hysteresis are predicted.

VII. CONCLUSION
The discovery of carbon nanotubes followed by the recent large-scale production of these nanodevices paved the way into wider integration between nano-and-biotechnology. The use of carbon nanotubes in field effect transistors has led to the scaling down of biosensors which allowed the detection of biomolecules in low concentration of analytes. However, the inhomogeneous interface between semiconductor CNTs and metal electrodes results in relative high contact resistance, which negatively influence the device performance in terms of current and transconductance. Though much effort has been done in addressing this issue, most of the methods implemented today to reduce contact resistance either come with shortcomings or are not cost-effective. Further study may investigate the homogeneity of pyrolytic carbon as electrodes in contact with carbon nanotubes for better alignment of their crystallographic structures and work functions as both materials are of the same carbon allotrope family.

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