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ECG Electrode (Sensor) printing with Ion Sensitive Membrane on Flexible substrate

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Abstract: *Globally 30% deaths occur due to Cardio Vascular Diseases(CVD). ECG is one of the way to monitor this circulatory disease. Ag/Agcl Gel based ECG electrodes have their inherent problems, for prolonged continuous monitoring of ECG, connecting to a Cellular Mobile Phone, carried for early warnings. ECG electrodes with solid electrolyte connectable to mobile phones for Digital Signal Processing(DSP) is the alternative, in such continuous monitoring requirement. There are three fundamental ways of measuring ECG. These are electron conduction current, displacement current and Ionic conduction current measurements.*

ECG Bioelectric signals are due to electric conductivity or polarization/depolarization of the heart muscles, involving ions as charge carriers. So, electron conduction or displacement current measurements have side effects, reflected in the form of additional noise and signal vander when compared to direct ion current measurements. Unlike in current ECG electrodes, where electron current is picked up for ECG, along with various related electron current noise potentials, ideally speaking, ion current need to be picked up for very good noise and vander free Electro Cardio Gram(ECG). In Picking up such signals, there is involvement of interaction with Ionic charge carriers of the body and to transduce Ionic currents in to electron currents required for the signal carrying wires and interface electronics.

Nernst potential of the dry electrode, for specific Ion like K+ and Na+ play crucial role, in these ECG electrodes. Over Potential or Over Voltage in these electrodes is due to excess polarization of the electrolyte, which is not desired. Perfectly, polarizable electrodes, pass a current between the electrode and electrolyte solution, by changing the charge distribution within the electrolyte near the electrode. So, nano-polarizable electrode structures, with good characteristics are to be preferred, for very good ECG measurements.

Ion sensitive Thin Film Transistors (TFT) with Ion selective membranes at the skin interface is one of the alternative to sense Ion currents emanating from the human body. In order to rap around the body, a flexible substrate is more suitable for forming printable ECG electrodes with Ion sensitive membrane interface. Fully printable SWCNT (Single Wall Carbon Nano Tube) based Thin Film Transistors(TFT) with Top Gate structure are realizable on flexible substrates. The Ion transport or charge carrier transport, in general is through the intrinsic Frenkel or Shottky defects and 1-dimensional, 2-dimensional and 3-dimensional defects of the crystals or nanocrystals of the membrane. These Ion selective membranes are also printable, thus leaving the scope of ECG electrode to printable electronics, for online ECG applications.

In the publication, based on Ion conduction path analysis of the electrode, a methodology is evolved for printable ECG electrodes on Flexible substrates, considering the existing research results in, Printable Thin Film Transistor field and Printable Ion selective membranes and reference electrodes. After analyzing methods of individual subsystems, best combination is chosen, for evolving the integrated methodology for Printable ECG electrodes, to form Ion conductive path for 10^{-8}M concentration and 7 to 8 KΩ/Sq resistance per membrane layer, to suite the two stage skin electrical model.

Keywords: *ECG, Printable TFT, SWCNT, Membrane, Ion Conduction*

1. Introduction

Bio-electric signals/Bio-potentials such as ECG are due to electric conductivity of the heart muscles producing mechanical pumping action, involving ions as charge carriers. Picking up these signals involve interaction with these ionic charge carriers and transduction of ionic currents into electronic currents required for the carrying wires and the interface electronics. A good interaction between electrons in the Bio-potential electrodes and ions from the body has effect on the performance of these sensors, specifically at low frequency signals. Unlike in the current ECG electrodes, where electron current is picked up for ECG, along with various related electron current noise potentials, ideally speaking, ion current need to be picked up for very good noise and vander free Electro Cardio Gram(ECG). More over, it is difficult to use current ECG electrodes for continuous on-line monitoring application and also flexible substrate for printing the electrode, suits the firm contact requirement of the human body shape, to avoid arte facts in ECG.

Half cell potential or the Nernst potential of the electrode plays crucial role. Over potential or over voltage, in the electrode is due to excess polarization of the electrolyte, which is not desired in the ECG electrode. Perfectly, polarisable electrodes pass a current between the electrode and the electrolyte solution by changing the charge distribution within the electrolyte near the electrode. So, nano polarisable electrode structures with good characteristics are to be preferred, for very good ECG measurements. Ion sensitive FETs with Ion selective membrane at the skin interface is one of the alternative to sense Ion currents emanating from the human body.

2. Skin Electrical Model

Quality of Bio-Potential measurement is highly dependent on electrode skin contact impedance, which oppose time-varying current. Electrode contact is divided into two components, the metal electrolyte interface and the electrical model of the skin. The metal-electrolyte (gel) interface has been understood more by researchers than the more complex electrical characterisation of the skin. Skin consists of epithelial tissue. The three layers of skin are epidermis, dermis and subcutaneous. The epidermis has innermost sub-layer 'stratum basale' and outermost 'stratum corneum, as compacted, flattened, non-nucleated and dehydrated cells. There is continuous process of these dead cells transported from inner sub-layers, across the 10 μm thickness of highly in-homogenous stratum corneum. The second layer 'dermis' formed with network connective tissue has collagen fibres providing elastic properties, blood vessels, hair follicles, sweat glands and oil glands, as depicted in the skin structure shown below in the Fig 1.

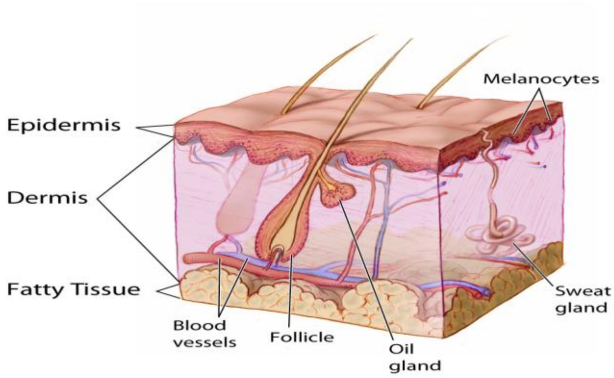


Fig 1

Stratum corneum is more dielectric in nature, whereas dermis and subcutaneous is conductive in nature. Subcutaneous layer has connective tissue, which allows skin to move freely. Hair follicles and sweat ducts pass through epidermis. An increased hair follicle density decreases the resistance of skin. Hair follicle density can vary between 40 and 70 $cm^{-2,}$ thus electrical properties of skin can vary significantly between different subjects and body sites. Resistance of the skin also depends on activity of sweat glands and location dependent. This skin resistance is time dependant and varies within limits, based on the activity of sweat glands.

Capacitive coupling:

ECG is a changing electric field, which cause displacement currents to flow through it's measurement system to earth. These displacement currents have to be coupled via. matching capacitive impedances. So, in the case of Bio-electric measurements such as ECG, the displacement currents have to be coupled optimumly.
Undesired signals in the existing electron conduction based ECG Bio-Potential recording:

- Electrolyte-skin noise
- Metal-electrolyte noise
- Thermal noise

3. ECG Electrodes Based On Electron Conduction Current:

3.1 ECG Electrodes:

Bio-electric signals/Bio-potentials such as ECG are due to electric conductivity of the body parts, involving ions as charge carriers. Picking up these signals involve interaction with these ionic charge carriers and transduction of ionic currents into electric currents required by carrying wires and interface electronics. A good interaction between electrons in the Bio-potential electrodes and ions in the body has effect on the performance of these sensors, specifically at low frequency signals.

Half cell potential or the Nernst potential of the electrode plays crucial role. Over potential or over voltage, in the electrode is due to non uniform charge distribution, which is not desirable in the ECG electrode. So, nano electrode structures with good characteristics are to be preferred, for very good ECG measurements. So, Ion sensitive FETs with Ion selective membrane at the skin interface is one of the alternative to sense Ion currents emanating from the human body, to eliminate electron conduction based conventional electrodes.

Types of Electrodes:

Mainly, there are two types of electrodes, Polarisable or Non Polarisable and also known as dry or wet electrodes.

Non-Polarisable electrodes:

Electrochemical Process between the Gel and the Biological tissue, yields a conductive path between the patients skin and the electrode, for electrical current flow along the path

Advantages and Disadvantages of non-polarisable electrodes:

- Easily fixed, preventing motion Artifacts
- Relatively clean and reliable ECG signals.
- Normally require skin preparation in advance, such as using alcohol to clean the contact area. Some times the adhesive part, cause skin irritation and contact skin dermatitis, if used continuously for long time.

4. Impedance of electron conduction based ECG Electrode

Almost all acquired bio-signals including ECG are of small amplitudes (uV to mV level) and low frequencies 0.1 Hz to 1 kHz. There are two Skin-Electrode interface models i) Single Time Constant ii) Double-Time Constant model. Neuman's model, which uses two stages of the single time constant is called Double Time Constant model and is relatively accurate. The first stage represents skin, while the second represents the electrode. Pictorial depiction of the electrical equivalent of skin impedance is as shown in the Fig 2 below:

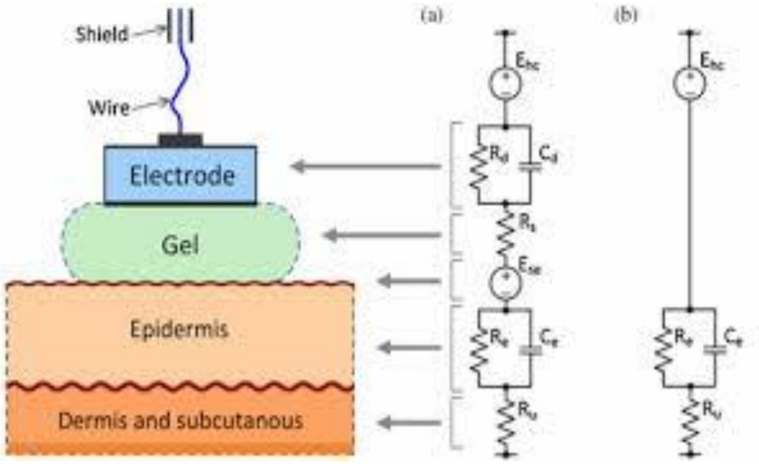


Fig 2

5. Flexible Ion Sensitive Ecg Electrode Printing:

There are three subsystems in an ECG electrode, Thin Film Transistor (TFT) device printed on Flexible substrate, Ion Sensitive membrane and reference electrode. Based on low cost, in this method a Carbon Nano Tube(CNT) based semiconductor device is chosen. Research has been taking place in the area of Ion Sensitive Membranes(ISM) doped with ionophores [1]. K^+ Ionophore doped polymer material is chosen for the methodology, assessing it's advantages in low cost and relative ease of formation of Ion Sensitive Membrane layer.

Low cost, high throughput and ability for mass production advantages of industrial ink-jet printing technique on flexible substrate for fabrication of Carbon Nano Tube (CNT), Thin Film Transistor (TFT) has gained sufficient ground for using in applications over the last few years [1]. Carbon Nano Tube (CNT) has been regarded as one of the promising materials for TFT applications, due to its excellent electrical characteristics as a semiconductor. One of the popular method among researchers for realizing CNT solution/ink based TFT printing or deposition has been droplet-evaporation [2][3]. As FET channel narrows, there is drastic improvement in channel resistance, when compared to contact resistance [2,4,5,6,7], which has been proved over a period for TFTs based on, organics[4], microcrystalline silicon[5], carbon nanotubes[2], silicon nanowires[6] and graphene[7] and extendable to inkjet based CNTs . Ideally, a narrow channel and reduced overlap area between the Source-Darin and Gate Layer structure is advantageous in reducing the parasitic capacitance.

The relation between the contact resistance R_C and the contact length L_C is given [8] by

$R_C . W = 2 * R_{scheet} L_T \coth (L_C/L_T)$ ⟶ (1)

Where R_{sheet} is the sheet resistance of the semiconductor layer and L_T is the transfer length. The transfer length L_T is the characteristic length over which 63% of the charge-carrier exchange between the contacts and the semiconductor occurs. In a printed semiconductor, due to additional spread and charge transfer between the contacts and the channel in a non-perpendicular direction an additional contribution to the contact length L_C may need to be taken. This additional contribution can be taken care of by the extended contact length L_{EXT}, rewriting the above equation as

$$R_C * W = 2 * R_{sheet} L_T \coth \left[L_C + L_{EXT} / L_T \right] \longrightarrow (2)$$

The cut off frequency f_T of a field-effect transistor in terms of effective mobility μ_{eff} is

$$f_T = \mu_{eff} (V_{gs} - V_{th}) / 2\pi L(L+2L_c) \longrightarrow (3)$$

The gate-source voltage $V_{GS,}$ the threshold voltage V_{th}, the channel length L, and the contact length L_C . The contact length L_C is the distance by which the gate electrode overlaps the source and drain contacts. L_C appears in the above equation, to reflect the parasitic capacitance that is charged and discharged during each switching event. In order to achieve, high cut off frequency, there is a requirement to minimize contact length.
When the channel length is reduced, so that channel resistance $R_{Channel}$ becomes less than contact resistance R_C , it reduces effective mobility μ_{eff} of the transistor to drop substantially below the intrinsic mobility μ_o of the semiconductor [1][8]

$$\mu_{eff} = \mu_o * \left[1 - \left(\frac{\mu oCiWRC(V_{GS} - V_{th}L)}{\mu_o CiWR_C(V_{GS} -)} \right)^{2} \right] \longrightarrow (4)$$

With the gate-dielectric capacitance per unit area C_i and the channel width W. It is the precise reason why the effective mobility μ_{eff} in Thin Film Transistors (TFTs) with reduced dimensions has been usually below 1 $cm^2 (V s)^{-1}$, [9,10,11,12,13] even if the semiconductor is known to have a much larger intrinsic mobility μ_o . But a record cutoff frequency of 28Mhz measured at a relatively high voltage of 25V has been reported for TFTs based on Carbon Nano Tubes having channel lengths of the order of 2µm [14]

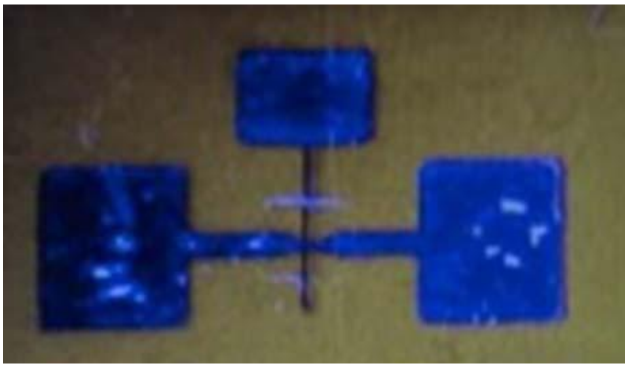
Transmission Line Analysis (TLA) for the CNT TFT Channel is very much applicable with the help of equations (1), (2), for various contact lengths Lc, fixing required contact resistances much less than 7 KΩ, for known W of the transistors printed with the methodology given below. After printing, from the measured values of $R_{C,}$ and characteristic effective charge transfer length L_T the values of L_{EXT} are computed. The values resulted give a fitness curve for the specific methodology, for extrapolation, for designing printable ECG electrode(sensor) with Ion Sensitive Membrane on Flexible Substrate .

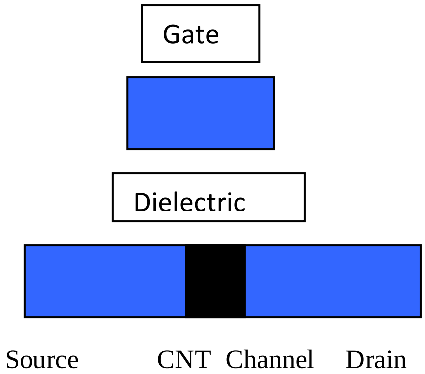
6. Fabrication Methodology:

6.1.1 CNT-TFT

Choose any material printer to fabricate the Carbon Nano Tube (CNT) – Thin Film Transistor (TFT), with ink droplet volume size of 10 pL and droplet size estimation of 25 µL in diameter. Use Kapton as flexible substrate for printing top gate FET. Calculate the physical area of the Source (S) and Drain (D) layers and print Source (S) and Drain (D) layers with silver nano particle ink(Source: Aldrich). Then sinter Source (S) and Drain (D) region already printed for about 20 minutes at 120^oC. Towards the channel, while printing the Source and Drain region, only few drops in a controlled way to be dropped for better end of the tips on either side of the channel, as shorter channel length TFT has better performance [1][2][8]. The structure shown is advantageous in reducing the parasitic capacitance, due to elimination of overlapping area between the S-D regions and the Gate Layers.

Now, the S and D regions are ready with very narrow channel gap. The channel is printed with a semiconducting Carbon Nano Tube (CNT) ink. The Carbon Nano Tube (CNT) ink is prepared by mixing 99% pure semiconducting Single Walled Carbon Nano Tube (CNT) powder (Source: Aldrich) with 1- Cyclohexyle -2-Pyrrololidine (CHP), followed by 4 hours of sonification [14] . Over the period, it is found by researchers, a concentration of 0.01 mg/ml of CNT powder gives good deposition properties without clogging the nozzles. Subsequent to printing, carry out Laser sintering process at 120^0C for 7 minutes. Then print dielectric layer with dielectric inks (Source:Aldrich) and sinter at 120^0C for 7 minutes. The Gate layer is finally printed, with Silver based inks, followed by sintering at 150^0C for 15 minutes. An optical image of the completed structure with Source and Drain would look like the figure 3 below.





Source Channel Drain W X L - 5µ X 300 µ
Fig. 3

6.1.2 Ion Sensitive Membrane (Ism) Printing:

Ion Sensitive Membranes (ISM) typically contains a lipophilic ion receptor, an ionophore and in a specified optimized molar ratio and Ion exchanger, both incorporated into a polymeric membrane such as plasticized PVC. As per the established research literature protocol [15], the K^+ Ion Sensitive Membrane(ISM) contain 1.4 wt% of Valinomycin, 0.3 wt % of Potassium Tetrakis (4-chlorohenyl) borate KT_p ClPB, 65.5 wt% of 2-nitrophenyl octyl ether (o- NPOE), 32.8 wt% of Poly Vinyl Chloride (PVC high molecular weight) and Tetrahydrofuran (THF) (Source: Aldrich). The membranes are made dissolving the above stochiometric mixture into 1.5 – 4 mL of Tetrahydrofuran (THF). Then the solution is Poured in to a Petri dish and the Tetrahydrofuran is allowed to evaporate over 24 hours. Cut the membrane in to small pieces less than 1 sq cm and condition them soaking overnight in Potassium solution of 10^{-3} M concentration.

6.1.3 Reference Electrode:

In top gate Ion Sensitive FET(Field Effect Transistor) based, Ion current measurements a reference electrode is required. The reference electrode with AgCl nano ink of concentration, which give 10^{-3}M equivalent current is printed, in the vicinity of the Thin Film Transistor (TFT) already printed on the Flexible substrate (Kapton). The printed reference electrode is Laser sintered at 120^{o}C for 20 minutes.

7. Conclusion

The methodology allows low cost and continuous usage application of ECG electrode (sensor) for online monitoring, at low power/voltage operation of the TFT and Source/Drain region. The K+ ionophore based membrane works in sweat conditions. There is a possibility of reference electrode, which is not sticked to the body, printing on the other side of the Flexible substrate, for longevity. In the present methodology, it is little away from the printed electrode (sensor) device sticked to the body. The work can be extended for improving the sensitivity of the Ion Current sensing, for measuring various Bio-potential signals and digitizing the signals for wireless transmissions using Near Field Communications (NFC), as the printable transistor frequency cut off [18][19], current sensing, digital[16][17] blocks improve, in the Flexible electronics research[20] arena, as researchers, already succeeded printing improved parameters on glass substrate.

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