

Advanced nanomaterials for Organic Compounds detection and Oxidation by Electrochemical methods with the Aid of Active Catalysts

Mamadou Kalan. DIALLO ^{a,b,*}

^a: Guinea Energy Corporation (GEC), Conakry. Guinea.

^b: Hassan II University of Casablanca. Faculty of Science and Technology, Mohammedia. Morocco.

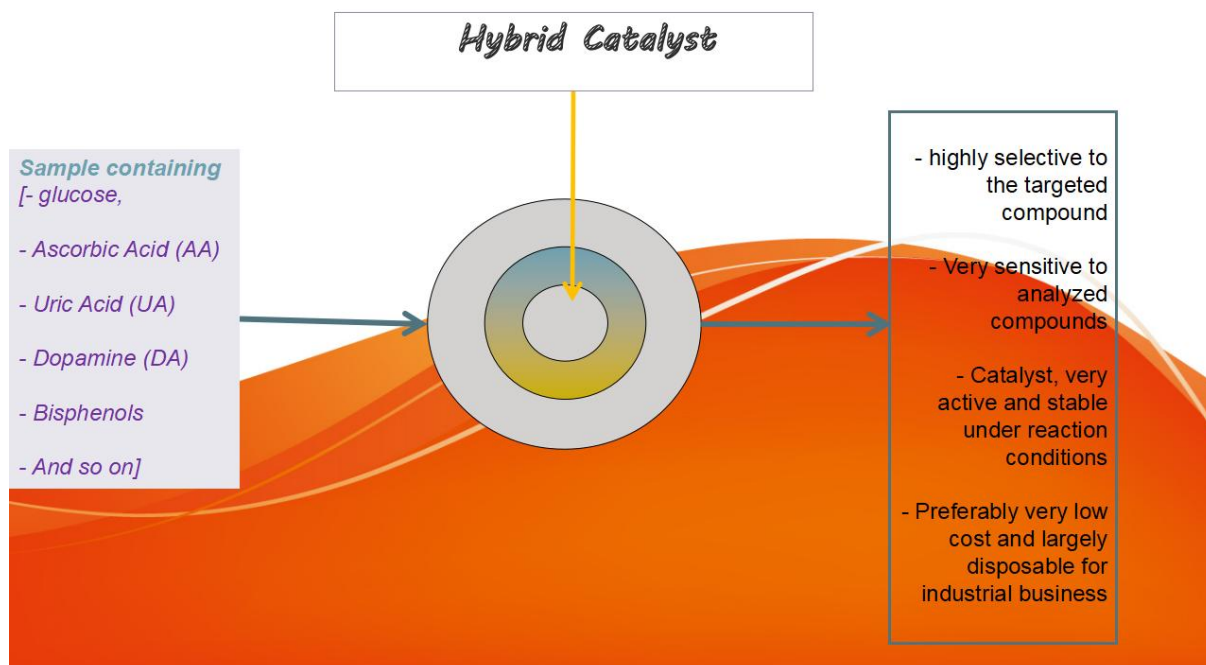
* Corresponding Author: m.k.diallo@outlook.fr

Abstract

The coexistence of biomolecules in the living being blood, and urine plays a crucial function in the biological systems. For i.e., in human organism, generally there coexist Ascorbic Acid (AA) and Dopamine (DA) in the extracellular fluid. Moreover, in the human's blood and urine, it coexists Uric Acid (UA) and (AA). Therefore, these compounds are of great interest in biomedical function which play a essential factor in the human metabolism, central nervous, renal systems, and beyond. However, abnormal levels of these molecules can lead to a drop-off in the biological system purpose. So, that, since long time ago, as described here, there are many materials in diverse forms that have been designed and developed for both biomolecules oxidation and detection. However, it still some issues to master this concern such the materials costs, the process optimization and cost, the environmental friendliness of the suggested catalysts, and so on. Therefore, there is a higher need in the development of a new and powerful platforms for the early and facile sensing of these biomolecules with cost-effective, high selectivity, as well as best sensitivity, superior stability.

Keywords: Hybrid-catalysts, Electrochemical, Sensors, Organic/Inorganic, Compounds, Oxidation

Graphical Abstract



1. Introduction

Bioactive molecules such as glucose (G), and related compounds, dopamine (DA), Ascorbic Acid (AA), Uric Acid (UA), and so on are of great interest in human metabolism, nervous system, and as well as renal systems [1,23]. So that, measuring and controlling the levels of these biomolecules is highly regarded in order to master their linked function in the human health. As it is known that abnormal levels can cause many diseases which generate uncountable death worldwide. So, according to the WHO data's, the number of adults living with diabetes has significantly quadrupled from 1980 to nowadays yielding of 422 million as described by Dong's research team [2]. Indeed, as known there exist two principal type of diabetes. So, type (I) is characterized by the lack of insulin production by pancreatic islet cells, and the type (II) is a result of under activity of insulin produced in the pancreas by the body. However, Diabetes are known as a chronic disease, which can lead to complications and severe symptoms, yielding a significant number of deaths. Other these diseases, like Parkinsonism, chronic kidney disease (CKD) are connected to neurological disorder with evolving layers of complexness. As reported in Kalia's work [3] that parkinson's disease has been defined by the classical motor characteristic of Parkinsonism joint with low bodies and failure of dopaminergic neurons in the substantial nigra. However, it is concluded from Su's research analysis [4] that uric acid-lowering medical care appear to improve kidney outcomes and reduce the risk of cardiovascular events in adults with CKD. So, nowadays there are many diseases connected to the abnormal levels of some biomolecules in our organism or by the large consumption of industrial products. So, due to that, Researchers, engineers, Doctors, professionals, and so fourth are exploring new routes for the design of favorable catalysts for diverse disciplines. Indeed, recently a full descriptions of nickel-based hybrid catalysts were investigated in [5] research work for diverse considered reactions. Also, the same researcher analyzed the performance of Copper-based materials for reduction reactions like (CO_2 , NO_3^-) [6] to high value-added products and these catalysts might be taken as an anode catalyst we believe. Furthermore, Mouratib, R et al [7] showed a development of modified carbon paste electrode with a material that contains high amount of Al-and-Si. This modified electrode was used as active catalyst for water treatment sludge (WTS) for Bis-phenol-A sensing. According to their manuscript, the prepared catalyst delivers superior electrocatalytic activity toward the targeted reaction (oxidation of Bis-phenol-A) which yields a low limit of detection as high as $0.1 \mu\text{M}$. In addition, Salhi, O et al [8] fabricated a poly-1.8-Diaminonaphtalene and cysteine modified black carbon electrode (Poly-1.8DAN/Cys/CB) for the determination of nitrite in real samples. So, the designed electrode yields outstanding electrocatalytic activity toward nitrite ions finding. As a result, the prepared (Poly-1.8-DAN/Cys/CB) showed a very low limit of detection as high as $0.25 \mu\text{M}$. It is mentioned that this research work highlights a simple preparation method of a polymeric film which might contains considerable amount of both amine and thiol groups for nitrite sensing. Recently, Govindaraj, M et al [9] demonstrated that nowadays, there exist many several categories of non-enzymatic glucose sensor materials. These materials include, composites, non-precious metals and their hydroxides, alloys, oxides, precious metals, and their alloys, carbon-based materials, conducting polymers, metal-organic frameworks (MOFs) and so fourth. Moreover, Naikoo, G.A., et al [10] dedicated a review on the trends in bimetallic nanomaterials and methods for the fourth-generation glucose sensors (FGGS). So, it shows that bimetallic-based glucose sensors are an up-and-coming evolution in the field of glucose biosensing. Also, those biocompatible, nanocomposites exhibits considerable surface area for sensitive glucose discovery. Moreover, bimetallic nanomaterials have advisable physical phenomenon and sensing performance than the monometallic one. Indeed, bimetallic nanomaterials are used for signals amplification during glucose determination. Although, there exist such candidate nanomaterials, but their prices, the preparation methods, the availability, and as well as the environmental-friendliness must be well issued for best purposes. Therefore, in this research manuscript, we aim to address a brief review on the recent development of novel nanomaterials for both organic compounds detection and oxidation like (glucose, dopamine, so on) by electrochemical methods with the help of active catalysts. It is believed that this research review may guide for new perspectives to design powerful candidate materials for the considered issue. This may overcome many facts that limit the widely use of sensor and Biosensors for both organic and inorganic compounds detections in real samples.

2. Hybrid nanomaterials for sensing/oxidation of Glucose from samples

Due to the advanced features of monocyclic 5-aminoisophthalic acid (5-AIP), it was chosen to construct both mono and bimetallic metal organic frameworks through a facile one-step solvothermal method as depicted in Fig.1. So, these metallic MOFs nanomaterials were analyzed as a potential candidate for both oxygen evolution reaction (OER), and glucose oxidation reaction (GOR) catalysts as described by Baby's research team [11].

Indeed, as a result of the experimental analysis, it is found that the bimetallic CoNi(1:3) MOF delivers the best performance among the considered catalysts, whether in the OER test or GOR analysis. So, the CoNi(1:3) MOF

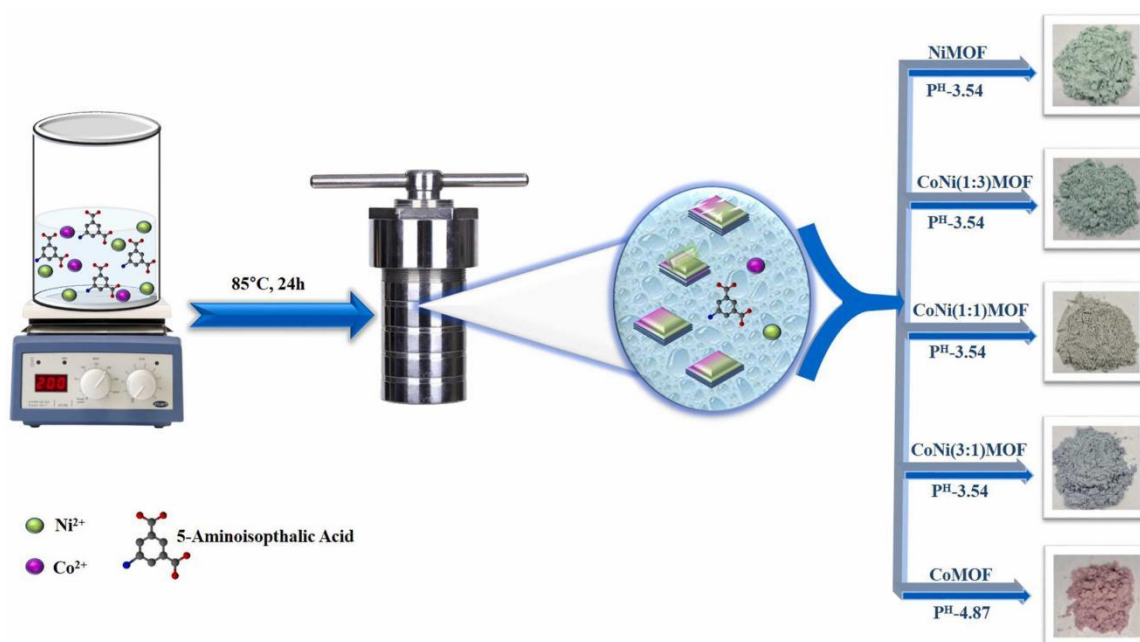


Fig.1 The synthesis route of monometallic and bimetallic metal organic frameworks (MOFs) catalysts for both oxygen evolution reaction (OER) and glucose oxidation reaction (GOR), reproduced with permission from [11] Copyright @ 2023, Elsevier B.V.

Performances of OER, and GOR were attributed three effects, i) its loosely stacked, ii) layered sheet-like structure with nanocutting channels along the edges, and iii) the presence of a synergistic effect between the distinct metal ions within the MOF. In fact, **Fig.2** gives a summarized analysis of CoNi(1:3) MOF performance for glucose oxidation with two ranges of concentration, first range ($2\mu\text{M}$ to $500\mu\text{M}$), and second range is to (1mM to 15mM), and as well as a respectively linear relationship between concentrations and current densities is done. Moreover, the CoNi(1:3) MOF provides higher performance by delivering at least overpotential of 348 mV at 20 mA/cm^2 , and as well as higher stability for OER. Also, it exhibited superior glucose detection limit of ($1.03\mu\text{M}$) by synergy. So, authors expect that this new nanomaterials would be a great advantages in the OER and GOR perspectives. Nikov's team [12] has been long-time ago, suggested the glucose oxidation over alumina supported Palladium ($\text{Pd}/\text{Al}_2\text{O}_3$). So, a catalyst deactivation has been featured to happen by the Pd-O formation in the metal lattice attended by the adsorption of the reaction products. Many properties have been considered such as the effects of material mass, glucose concentration, particle diameter, and as well as the multiple use of the catalyst on the targeted product conversion. Furthermore, Wang's research group [13] studied the tuning structure effects of Platinum (Pt) shells on Palladium (Pd) nanocubes as a neutral glucose oxidation catalysts and sensors. Indeed, the stellated $\text{Pd}@\text{Pt}$ NCs ($\text{Pd}@\text{Pt}$ SNC) with (211) facets showed an electron transfer number of 12.69 and a higher specific activity than other concave $\text{Pd}@\text{Pt}$ NCs. Also, it is described that Pd NCs with Pt-coated corners ($\text{Pd}@\text{Pt}$ CNCC) with index facets of (210) and (950) exhibited a sensitivity of $11.06\ \mu\text{A}\cdot\text{mM}^{-1}\cdot\text{cm}^2$ and a linear range of (2.4-10.6) mM. As the fabricated electrode gives higher recoveries (98.8 - 99.85)% and low relative standards ($\leq 1.06\%$) in the serum sample, it is suggested that this electrode ($\text{Pd}@\text{Pt}$ CNCC) might be useful as a non-enzymatic glucose sensor. Although these activity, the electrode may have high costs, as it is constructed with noble metals. So, to decrease the loading of noble metals on glucose oxidation catalyst synthesis, Li's group [14] proposed the use of MnO_2 -C nanocomposite to anchor the gold nanoparticles (Au NPs). In fact, it is observed that Au NPs are uniformly distributed on the support (MnO_2 -C). Higher catalytic activity has been notified for the constructed catalyst (Au/MnO_2 -C)NPs than other considered catalysts. This was supported by the effect of MnO_2 toward GOR. In particular, it is thought that MnO_2 -C hybrid composite is an auspicious approach for decreasing noble metal catalyst charging. Moreover, a very cheap catalyst than (Au/MnO_2 -C)NPs catalyst is reported from Hong's research team [15]. Where, a (110) faceted rhombic dodecahedral (RD), (111)-faceted octahedral (OCT), and as well as (100)-faceted nanocube (NC) Cu_2O crystals have been demonstrated as D-glucose oxidation catalyst. Specially, due to the facile surface oxidation of CuO , the RD catalyst showed an early rest potential of -0.228 V vs ($\text{Ag}/\text{AgCl}, 3\text{mol/l KCl}$) and it has higher specific activity compared to other catalyst forms. This demonstrated that the catalyst structure plays a crucial role in the catalysis reaction mechanisms.

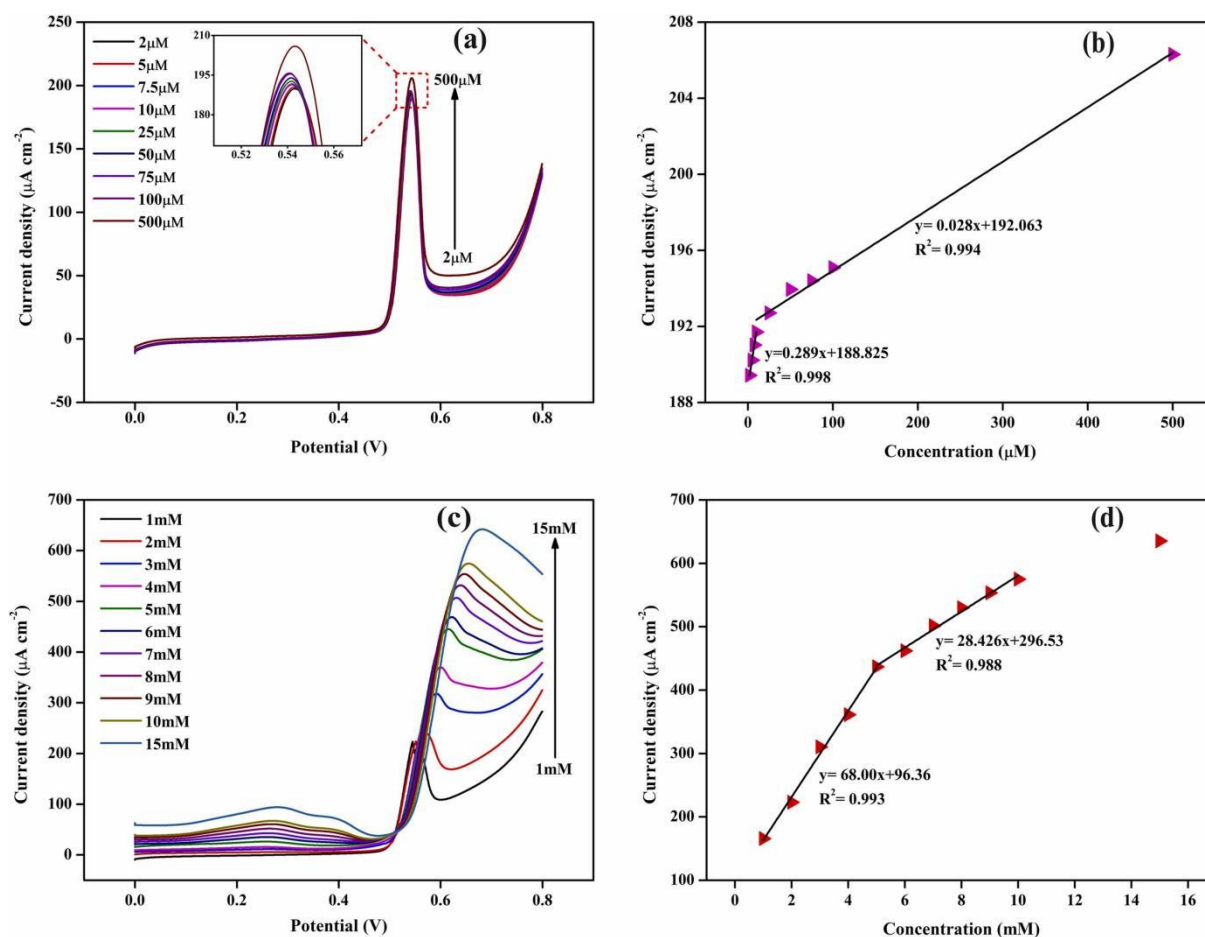


Fig.2 The evaluation of bimetallic metal organic frameworks ($CoNi(1:3)MOFs$) catalysts for glucose oxidation reaction (GOR) (a) LSV response of this catalyst towards low glucose concentration at $20mV/s$, (b) pointed out the relative relationship between the response current and glucose concentration, (c) LSV response of this catalyst towards high glucose concentration at $20mV/s$, and (d) indicates the relative relationship between the response current and glucose concentration, reproduced with permission from [11] Copyright @ 2023, Elsevier B.V

Also, recently, a binary oxides $Mo(1)-MnO_x$ catalyst has been proposed from Guo's group [16] as a potential catalyst for the glucose oxidation reaction from water containing higher glucose levels. Superior catalytic activity was remarked by using this special catalyst. A maximum 79% yield rate of formic was notified from glucose oxidation. Indeed, many other carbohydrates including (xylose, fructose, maltose, sucrose, cellobiose) with higher concentration levels furnish formic acid with a yield rate superior to 45%. Therefore, it shows the power of this special catalyst ($Mo(1)-MnO_x$) for the transformation of lignocellulosic biomass to formic acid. Recently, it is demonstrated from Oña's work [17] that higher catalytic activity could be achieved by using of small gold nanoparticles (Au NPs) loaded on mesoporous subunit carbon (SC) when studying both glucose and xylose oxidation reactions. Indeed, when this catalyst (SC/Au NPs) is used, it is noticed that the electrocatalytic oxidation of glucose and xylose happen at -0.05 , $+0.3$, and $+0.4$ V vs Ag/AgCl respectively. So, analysis revealed that there is a strong influence of Au cluster sizes on the catalytic performances of SC/Au NPs toward sugar electrocatalytic hydrogenation (ECH) and oxidation (ECO). More recently, Maghsoudi's team [18] supported both Ag and Mn over zinc oxide (ZnO) through hydrothermal route, to develop active catalysts. These materials (Ag/ZnO; Mn/ZnO) were applied as an anode catalyst to oxidize molecular glucose in alkaline media. Both physical and electrochemical methods were adopted in the study. So, experimental analysis revealed that the larger effective surface area of M/ZnO (9.25 cm^2) and Ag/ZnO (6.9 cm^2) attended by faster glucose diffusion ratio due to the solid impact of Mn and Ag charging over ZnO nanostructures reinforced the glucose oxidation reaction for both developed catalyst as compared to the unmodified ZnO. Moreover, Ma's group [19] designed and synthesized a silver Nanowire Aerogel catalyst for the sensitivity determination of glucose. The prepared non-enzymatic catalyst (AgNW-gel) shows a unique porous structure, lower resistivity, rapid electron transport and transfer, and as well as considerable active surface area, which supply a large number of active sites. So, the (AgNW-gel) exhibits superior sensitivity as high as 1940 $\mu A \cdot mM^{-1} \cdot cm^2$, and a limit of detection of 0.8 mM/cm^2 . Hence, it is suggested that the (AgNW-gel) could also be embedded into compact blood glucose meters which might be useful for real-time monitoring of blood glucose or for medical human activity in remote regions.

Jiang's group [20] described a powerful sensitive nonenzymatic glucose sensor based on CuO NPs modified carbon nanotubes (MWCNTs) catalyst. Many physicochemical methods were employed to evaluate the performance of the developed anode material. So, at +0.4 V applied potential, the (CuO NPs/MWCNTs) shows higher sensitivity as high as $2596 \mu\text{A}\cdot\text{mM}^{-1}\cdot\text{cm}^{-2}$ to glucose. Also, a linear range hold over a concentration up to 1.2 mM with a detection limit as low as $0.2 \mu\text{M}$ (signal/noise = 3). Particularly, the sensor was highly resistant against the poisoning ion chloride, the impact of the oxidation of common interfering species. As supposed, the (CuO NPs/MWCNTs) sensor was far beyond to the unmodified MWCNTs. Indeed, justifying the role played by each component of the hybrid catalyst. Therefore, it summarized that this kind of sensor is bright for the future elaboration of nonenzymatic glucose sensors. Furthermore, in 2012, Luo's research team [21] proposed a sensor based copper nanoparticles (Cu NPs) modified graphene sheets (GSs) as a nonenzymatic glucose sensor. Indeed, it is thought that due to the synergistic effect between Cu NPs and GSs, a high oxidation current and as well as a negative shift in the peak potential were achieved. So, at 0.5 V, the Cu NPs/GSs exhibited a wide linear range up to 4.5 mM glucose with a LOD of $0.5 \mu\text{M}$. Due to the synergistic effects, the Cu NPs/GSs delivers high stability, selectivity, sensibility, and as well as fast amperometric detection of glucose, which suggest that Cu NPs/GSs might be useful for the development of nonenzymatic glucose sensors. More recently, a modified electrode is reported from Yuan's group [22] as a nonenzymatic glucose sensor and beyond. The (NiO NPs/GO/GC) catalyst was characterized by many technics including physical and electrochemical methods. So, when compared to (NiO NPs/GC) electrode, the proposed (NiO NPs/GO/GC) catalyst showed poor resistivity, powerful capacitive properties behavior (16-folds) and as well as better glucose oxidation reaction. The glucose linear range was from $3.13 \mu\text{M}$ to 3.05 mM with a detection limit of $1.0 \mu\text{M}$. Also, common interfering substances were no longer have a meaningful impact on the detection of glucose. Therefore, as described here, there are many materials in diverse forms nowadays which are available in the mark for both glucose oxidation and detection. But, always it still some issues to master this concern such the materials costs, the process optimization and cost, the environmental friendliness of the suggested catalysts, and so on. In the next section, it will be focused on the catalytic oxidation or detection of dopamine levels in the human serum and urine.

3. Hybrid nanomaterials for sensing/oxidation of dopamine

It is known that the development of advanced nanomaterials as sensor for sensing the biomolecules is on the cutting edges of physiological research, in order to master its levels in the human serum and urine. So, many catalysts (sensors) have been proposed to meet this requirement but some issues like (1) the electrode fouling upon oxidation and (2) the interference among the biomolecules due their overlapping of the oxidation potentials which leads to poor selectivity and reproducibility. So, in the same way, recently, Yang, Y., et al [23] reported a (SWCNTs-array-GCE) selective sensor, which presents high catalytic performances for both Dopamine (DA), and Uric Acid (UA). Also, as can be seen from **fig.3**, it exhibits some actives for Ascorbic Acid determination. Hence, it summarized that this sensor might be useful this particular issue as it possesses higher selective with a distinct oxidation potentials for each compounds and it also revealed plausible recovery. Recently, Liu, X., and his co-workers [24] made a review about Biosensors and Sensors for the determination of dopamine from samples.

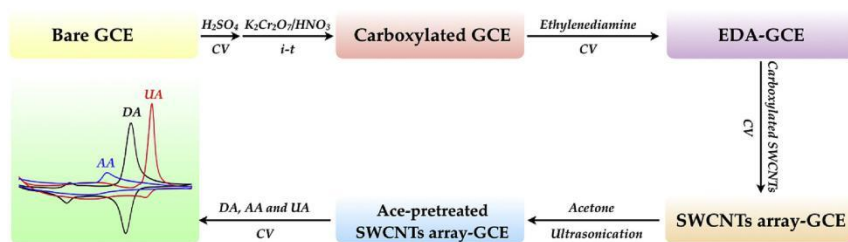


Fig.3 The synthesis route of **SWCNTs array-GCE** catalyst for all three organic compounds sensing, namely Dopamine (DA), Uric Acid (UA), and as well as Ascorbic Acid (AA), reproduced with permission from [23] Copyright @ 2023, Elsevier B.V.

So, it explores many methods including the use of enzymes, molecular imprinted polymer (MIPs), antibodies, advanced nanomaterials, and as well as Aptamers. However, the successful sensing must overcome interferences from molecules with a similar redox potential. So, as a conclusion, it is suggested that combining (MIPs) or Aptamers with Electrochemistry promises to achieve fast detection and enhanced selectivity. Suzuki's team [25] developed and characterized a Boron-doped Diamond (BDD) microelectrodes for the in vivo sensing of dopamine. So, a clear signal current response leading medical forebrain bundle (MFB) excitation could be achieved with superior sensitivity. Also, excellent stability reached, indicating that the BDD microelectrode are very promising for the next-generation in vivo electroanalysis. Dopamine constitute a neurotransmitter which plays a key role in the life Science. So, Tang's research team [26] designed and synthesized self-assembled

Au@Ag nanorods dimers based on Aptamers for ultrasensitive sensing dopamine. So, it notified that the electronic field significantly enhanced by the incorporation of silver shell coating on the surface of Au NR dimer. The surface-enhanced Raman scattering (SERS)-encoded this sensor system, hence exhibiting a LOD of dopamine as low as 0.006 pM and a wide linear range of [0.01 to 10] pM of dopamine detection. According to Sajid's group [27] the electrochemical analysis is the best viable way of dopamine sensing. However, it is very ambitious due to analogous redox potentials of the co-existing and high concentration interfering species in real biological samples. So, the team fully shown the description of different materials, and related characterization by using them in the detection of dopamine. Both novel dimensions, and as well as active challenges are regarded within the study. Moreover, a single layer Nano-MoS₂ modified gold electrode (nano-MoS₂/GE) has been fabricated by Hun's research group [28] for dopamine sensing. So, under optimal conditions, the developed electrode displayed considerable sensitivity, simplicity and a wide linearity range which starting up a novel promising signal-on the photoelectrochemical (PEC) platform for the determination of small organic molecule in Bioanalysis. Hence, this elaborated sensor holds a detection limit of 2.3×10^{-12} M. Recently, Elhag, S., et al [29] used a hydrothermal chemical deposition (HCD) way to grow an ultra-thin (Co₃O₄) nanowires over gold coated glass substrates for potentiometric sensing of dopamine. Considerable methods were used in order to characterize the sensor properties, including X-Ray Diffraction, X-Ray Photoelectrons Spectroscopy (XPS), Scanning Electron Microscopy, and as well as Ultraviolet-Visible (UV) absorption technics. So, the sensor shows a linear range with a slope of 52 mV/decade when dopamine concentration is ranging from [10⁻⁹ to 10⁻²] M. Indeed, the improved electrode features is attributed to the imperfection on the metal oxide (Co₃O₄) that is settled by the used surfactant along with the full surface area-to-volume ratio. A modification of gold electrode by the use of single-walled carbon nanotubes (SWCNTs) has been adopted by Kurniawan's group [30] as sensor for the detection of dopamine. Indeed, the developed sensor exhibited 0.79 μM, and 3.414 μA.mm⁻².μM⁻¹ of detection limit and sensitivity respectively when experienced in dopamine solution at pH 4.0 under the use of cyclic voltammetry technic. Also, the sensor does not exhibits any interference signal for ascorbic acid (AA) under the dopamine detection. Moreover, Kim's team [31] reported a glassy carbon modified graphene electrode through drop-casting way in order to construct high sensitive sensor for the determination of dopamine in the existent of ascorbic acid. So, the performance of this electrode was confirmed in an enough assets of (AA) 1mM. The linear range was between 4 μM to 100 μM of dopamine concentration. The developed electrode revealed a detection limit of 2.64 μM. In particular, Wu's group [32] reported a special type of porphyrin-functionalized graphene for high selective and sensitive sensing of dopamine (DA). Indeed, the limit of detection of this electrode was as low as 0.01 μM. And specially, a possible peak potential separation was observed at 188 mV, 144 mV and 332 mV for AA-DA, DA-UA and UA-AA respectively, which allows selectively sensing DA. Therefore, with respect to the good sensitivity and selectivity of this new-kinds of electrode, this sensor was applied to determine (DA) in real hydrochloride injection sample, human urine and as well as serum samples with a satisfactory outcome.

4. Conclusion

As known many molecules coexist in the living being blood, and urine. Those molecules play a crucial role in the biological systems. As an example, in human being, generally there coexist Ascorbic Acid (AA) and Dopamine (DA) in the extracellular liquid. Moreover, in the human's blood and urine it coexists Uric Acid (UA) and (AA). So, it is known that these kinds of compounds are of great interest in biomedical systems which play a key factor the human metabolism, central nervous, renal systems, and beyond. However, abnormal levels of these molecules can lead to a drop-off in the biological system function. For abnormal level of (DA) can lead to several neurological disorders, for i.e. Parkinsonism, Schizophrenia. Also, high levels of (UA) can cause the gout and hyperuricemia. But its low level are related to the disease as Scurvy. In the case of glucose, superior levels can yields to the disease named as diabetes, which cause nowadays much loss in human life. Although, there are many materials (electrodes) available in the mark for the detection of biomolecules in the real samples. But, there is a higher need in the development of a new and powerful platform for the early and facile sensing of these biomolecules with cost-effective, high selectivity, as well as best sensitivity, superior stability.

5. Conflict of Interest

The Author declares that this advances on nanomaterials-based nanocomposite (Article) was led in the lack of any profitable or sellable relationships that could be interpreted as a possible conflict of interest.

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