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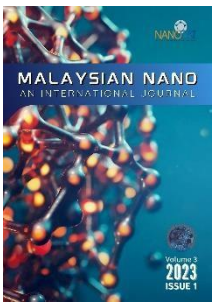

Review article

Nanocellulose-based carbon nanocomposite for the electrochemical sensing application for pharmaceuticals: A review

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Abstract

Biopolymers are naturally occurring polymers which have attained profound interest and are investigated widely due to their outstanding characteristics and several advantages such as cost efficiency, hydrophilicity, film formation capability, chemical inertness, non-toxicity, high mechanical integrity and biocompatibility. Cellulose is the most abundant biopolymer in the world. Nanocellulose (NC) is a cost-efficient, biodegradable, eco-friendly, biocompatible and abundant, renewable biomaterial obtained from cellulose by nanoscale isolation. The most interesting property of NC as the precursor material, is that it does not possess hierarchical structural defect. Recently,the incorporation of NC into electroconductive platforms i.e., nanostructured carbon nanocomposite is a hot topic for researchers. NC is not an electroconductive material but an ionic conductor. Availing this property NC can be incorporated with carbon-basedmaterials. CNTs is a carbon-based material with excellent electronic properties, high electroconductive and mechanical properties. This review focuses on the application of nanocomposites containing NC and carbon nanostructured especially CNTs and their properties and sensing applications.

Keywords: nanocellulose, carbon nanomaterials, nanocomposite, electrochemicalsensing, drugs.

1. Introduction

Recently, nanocellulose (NC) has been manufactured and utilized frequently in different areas such as supercapacitors, food packaging materials, paper electrodes, electrochemical sensors and others. NCis extracted from the native cellulose compounds by nanoscale isolation but it contains most of the inborn advantages of cellulose and is free from the inherent structural defects of cellulose. Several methods are available for the extraction of NC from cellulose sources resulting in different sizes and shapes of nanocrystalline with different mechanical features and surface chemistry, the structure of NChas shown in Fig. 1. NC has improved mechanical integrity and thermal stability which is suitable for several host matrices. Nanocellulose could increase the selectivity and sensitivity of a sensor device forthe electrochemical determination of specific desired analytes [1] [2]. NC has an excellent adsorption capability which helps to counter the leach of nanomaterials from the electrode surface.

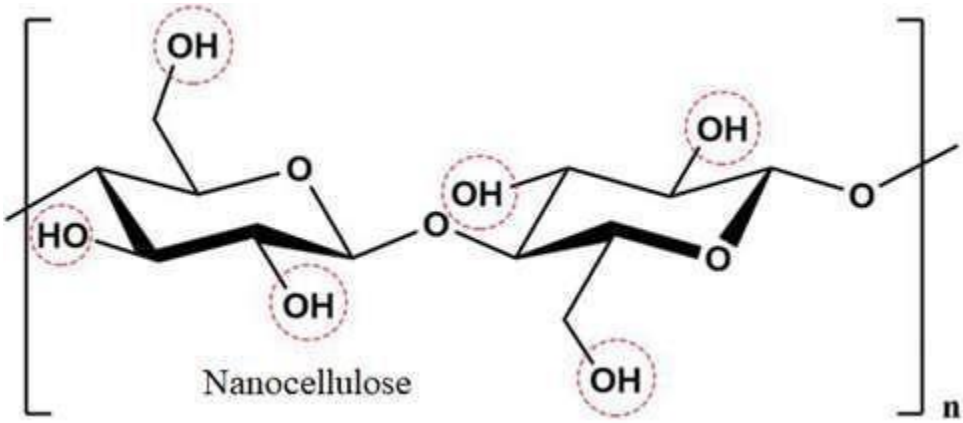

Figure 1: The chemical structure of nanocellulose

Thus, the prolonged stability of the sensor electrode can be achieved [1]. NC is a potential buildingblocks for the attachment with other carbonaceous nanomaterials such as polypyrrole [3], graphene (Gr)[4], CNT [5] and others.

Carbon nanotubes (CNTs)consist of graphene sheets coiled inside a nano-cylinder which exhibit several distinct properties such as outstanding mechanical integrity, electronic conductivity and catalytic activity.

CNTs are composed of a single or multiple layer of sp^2 hybridized carbon atoms. On this basis of composition, CNTs can be classified into two groups i.e., single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) [6].

The incorporation of carbon nanostructured materials within the nanocellulose matrices allow the nanocomposite intense mechanical and functional characteristics such as controlled electrical and enhanced thermal conductivity, rapid sensing capability, and sufficient molecular porosity.

MWCNTs is comparatively cost-efficient and have several outstanding properties such as tensile strength, good mechanical and thermal stability, flexible elasticity and high electronic conductivity andcatalytic activity [7]. Due to the poor solubility, the pristine CNT is not suitable for electrode fabrication.Hence, the CNTs functionalization has been considered as a technique for activation of electroconductivity. The functionalization technique effectively eliminates the impurities from CNTs and enhances the dispersion ability in water and improves the electrochemical performances. Moreover,this technique provides abundant anchoring sites on CNTs which facilitates the fabrication of electrodes[8]. In recent years functionalized MWCNTs has been used enormously in different fields of material science such as electrochemical sensors, biosensors [9] [10] [11], tissue engineering [12], energy storage [13] etc. In this review, we will focus on nanocellulose and its classification, synthesis methods,and application of nanocellulose modified with CNT (MWCNTs, SWCNTs) based nanocomposites fordrug molecules sensing.

2. Classification of Nanocellulose

Generally, NC can be classified into three categories, such as:

1. Cellulose nanocrystals (CNCs)
2. Cellulose nanofibers (CNFs)
3. Bacterial nanocellulose (BNC) [14]

Generally, cellulose nanocrystals (CNCs) contain high crystalline nanostructure, but cellulose nanofibers (CNFs) and bacterial nanocellulose (BNC) contain high crystalline configurations. Besides, they consist of disordered locations of amorphous regions, which provide high flexibility and more continuity [15]. Generally, CNCs are synthesized from cellulosic fibers by acid hydrolysis treatment, where the splits of the readily exposed amorphous regions occur to produce rod-shaped crystalline cellulose [16]. The acidic mixture was diluted upon reaching the appropriate level of hydrolysis, and then centrifugation and dialysis were applied to eliminate residual acids and impurities. The physicochemicalcharacteristics of CNCs firmly depend on parameters such as the nature of the acids utilized during synthesis and their concentration, temperature and time duration of hydrolysis, and cellulose source. Generally, CNCs of 5-10 diameter and 100-300 nm length could be synthesized from wood pulp [17].

Generally, the CNFs can be synthesized by several methods, such as grinding, acid hydrolysis, high pressure homogenization, ultrasonication, and others. The cellulosic fibers comprise two types of macrofibers – hemicellulose and lignin. The macrofiber contains microfibrils which consist of nanofibrilsof cellulose. The nanofibrils of cellulose consist of crystal and amorphous regions [18]. As the amorphousregion is responsible for the transverse split of the microfibrils, and turns

them into short monocrystals, thus the amorphous region is considered the structural defect of cellulose [19]. To overcome the structural defect, a recent drive to synthesize nanocellulose from cellulose by nanoscale synthesis has sparked the interest of scientists.

Bacterial nanocellulose (BNC) is an exceptional biopolymer with various potential applications. Several bacterial species, such as Acetobacter G. xylinus can synthesize BNC. The expanded surface areaand a bunch of interconnected porous systems suggest that BNC in the form of a 3D cellulosic network structure can be utilized as a carrier for catalysts [20].

3. Synthesis process of nanocellulose

Nanocellulose can be synthesized in three different ways; a) Mechanical method, b) Chemical methods, and c) Bacterial method. Among them most commonly used is a chemical method.

3.1 Mechanical method

In the mechanical method, NC can be obtained by breaking down cellulose fibers from the lignocellulosic biomass. Generally, the derived cellulose has a diameter of less than 200 nm. Before mechanical method, some steps such as grinding, decrystallization, acid hydrolysis and derivatization are performed to prepare NC. NC also can be obtained from the lignocellulose biomass with the application of high pressure. It is done by suspending the biomass with a high-speed stirrer combined with ultrasonication before high-pressure homogenization [21]. Ionic liquids can be applied to treat lignocellulose biomass before mechanical method, which penetrate through the microcrystalline cellulose and attack the hydrogen bonds between the cellulose molecules. Intra- and intermolecular bondsare demolished during high-pressure homogenization, and through this disintegration, NC is obtained from the biomass [22]. However, the main drawback of this method is the high energy consumption, anda large number of cycles are required for the defibrillation of the cellulosic biomass [23].

3.2 Chemical method

Acid hydrolysis is the most common technique to synthesize NC from lignocellulose biomass. The synthesis process of NC is shown in Figure 2. The cellulosic fibers are screened through a mill witha 20- mesh screen, transferred into 5.00 M of 250 mL sodium hydroxide (NaOH) solution, and warmedfor 3 h until the temperature reaches 80 °C. The slurry is thoroughly filtered and washed until the pH reaches 7.

Then the cellulosic fibers are air-dried properly, and dimethyl sulfoxide (DMSO) is added to an 80 °C waterbath for 3 h. Mainly, DMSO is added to swell up the matrix of cellulosic biomass so that the concentrated acids can penetrate inside the domain configuration of lignocellulosic biomass and break down the internal bonds easily. After that, the fibres are filtered and washed three times with 250 mL of water. The above pretreatments are performed prior to the acid hydrolysis method. The pretreated cellulosic fiber is dispersed in a combination solution containing hydrochloric acid and sulfuric acid in a 1:3 ratio and refluxed for 16 h. The obtained product is diluted by DI water with centrifugation at 2000 RCF. After several washing, NaOH is added to make it neutral. The solution isfurther washed three times, followed by dialyzation through the membrane tube with constant stirring for 24 h. The obtained product is dried at 70 °C for 24 h and the final product (nanocellulose powder) is obtained in pale white powder [24]. In the acidic treatment, the yield of NC depends on parameters such as reaction time, acid concentration, temperature, and amount of lignocellulosic biomass. The yield of NC declines with the increased duration of acidic treatment of cellulosic biomass. The experimental parameters are optimized to obtain the maximum yield of NC, and top reserve the NC morphology [22]. There are several advantages to synthesizing NC using the mixed acid hydrolysis method using hydrochloric acid and sulfuric acid. In the acid hydrolysis method, due to the alkaline pre-treatment of cellulosic biomass, comparatively less energy is required to defibrillate the cellulosic nanofibrils than the mechanical method. The synergistic effect of mixed acid hydrolysis and ultrasonication energy produces NC with decreased particle size, ensures high crystallinity of the NC, allow for effective removal of amorphous regions, and increases the availability of C-OH groups morethan any other method. NC obtained from the acid hydrolysis method has a greater surface area and is more stable and dispersable than NC obtained from other methods [23]. In this research, NC synthesized by the chemical method can be used as a binder and film-forming agent to prevent the leaching of nanomaterials.

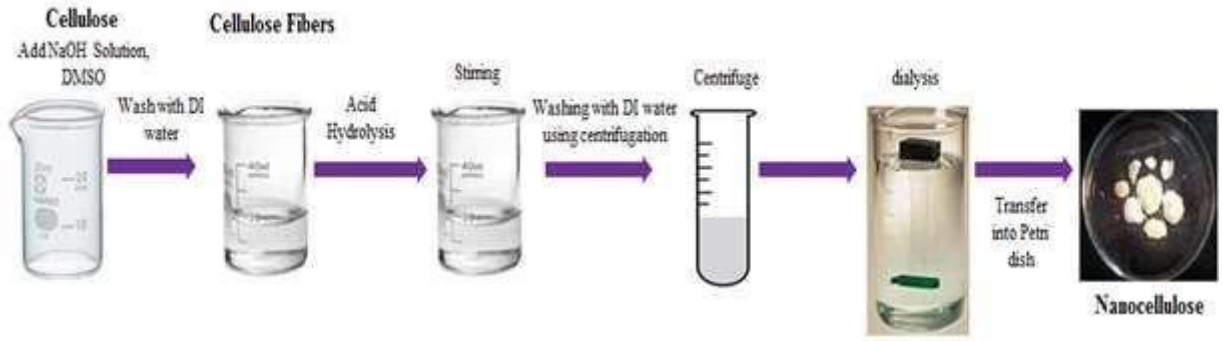

Figure 2: The steps of nanocellulose synthesis process

3.3 Bacterial method

The NC synthesised via the bacterial method has the same chemical configuration as NC synthesized via chemical and mechanical techniques from lignocellulosic biomass. Moreover, an ultrafine network of nanofibers is formed due to the culture medium, which exhibits remarkable features such as high purity, uniform morphology, good mechanical characteristics and flexibility, good absorption capability, and others. Bacterial nanocellulose (BNC) can be synthesized by aerobic cultivation of bacteria such as *Gluconacetobacter xylinus,* which grows in glucose-enriched medium [25].

4. Carbon nanotubes (CNTs)

Meanwhile, CNTs consist of graphene sheets coiled inside a nano-cylinder, which exhibit several distinct properties such as outstanding mechanical integrity, electronic conductivity, and catalytic activity. CNTs are composed of a single or multiple layer of sp^2 hybridized carbon atoms. On this basis of composition, CNTs can be classified into two groups i.e., single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) [8]. MWCNTs are comparatively cost- efficient and have several outstanding properties such as tensile strength, good mechanical and thermal stability, flexible, high electronic conductivity, and excellent catalytic activity [26]. Due to its poor solubility, pristine CNT is not suitable for electrode fabrication. Hence, CNTs functionalization has been considered a technique for activation. The functionalization technique effectively eliminates the impurities from CNTs, and enhances their dispersion in water, and improves their electrochemical performances. Moreover, this technique provides abundant anchoring sites on CNTs, which facilitates the fabrication of electrodes [1]. In recent years, functionalized MWCNTs have been used enormouslyin different fields of material science, such as electrochemical sensors [26, 27], biosensors [28, 29], tissue engineering [30], energy storage [13], and others.

5. Functionalization of CNTs

There are several methods to functionalize CNTs according to its unique characteristics. However, it is quite challenging to find the best technique to improve the distinct properties, such as mechanical, electrical, optical and others. One of the challenges is achieving the optimal functionalization of CNTs for a specific application. Functionalization is based on the covalent bonds among the functional groups on the outer wallof the CNTs. Functionalization of CNTs can be done by the acid hydrolysis method, which requires the presence of one or two strong acids such as hydrochloric acid, sulfuric acid, nitric acid, etc. Acid hydrolysis is one of the suitable methods for CNTs functionalization, where CNTs is used as a nanocomposite and applied for sensor fabrication [31].

6. Nanocellulose-based carbon nanocomposite

Nanocellulose-based nanocarbon composites are cellulose/carbon nanoparticles hybrid materials (such as nanofibrils, nanocrystals, carbon nanoparticles, fullerenes, graphene, nanotubes, and nanodiamonds). The functionality of nanocarbons is obtained by combining nanocellulose with nanocarbon materials using a non-metal matrix or carrier, non-melting, nontoxic, dimension-stable, eco-friendly, and low-cost, has so many applications including, biomedicine, biotechnology, and industry. Furthermore, because of its structural resemblance, tunable chemical properties, chemistry, viscosity- elastic properties, and non-animal origin to biological molecules, it is considered as an exciting materialfor biomedical applications [32].

7. The benefits of the incorporation of nanocellulose and carbon-based composites

Compared to the synthetic polymer-based nanocomposite, NC possesses several inborn expediences, such as biodegradability, biocompatibility, mechanical robustness, less toxicity, and multifunctional properties [33]. When carbon nanomaterials are introduced in the NC matrix, some excellent properties can be achieved from this combination, such as excellent dispersion and highly organized configuration, high electrical conductivity, excellent catalytic activity, magnetic properties, stretch-ability, optical activity and others [34]. In addition, incorporating carbon nanomaterials across the NC matrices offers the bio nanocomposites outstanding mechanical support and functional characteristics [33]. Therefore, the biointerface between the carbon nanomaterials and NC (bionanocomposite) is highly desired and involves unique morphologies. These combined structural arrangements not only preserve the actual properties of the carbon nanostructures but with extra biofunctionality. Due to the highly conductive nature of the bionanocomposites, they can be used as electronic devices. For example, multiple attemptshave been reported to incorporate highly conductive graphene materials with NC to fabricate a conductive bionanocomposite [9, 33].

8. Application of (*f*-MWCNTs/NC) nanocomposites for electrochemical sensing

Shalauddin *et al.* developed a nanocomposite of NC and functionalized multiwall carbon nanotubes(*f*-MWCNTs/NC), followed by the preparation of nanocomposite via drop-casting onto the surface of glassy carbon electrode for the electrochemical detection of painkiller drugs diclofenac sodium. The acid hydrolysis method synthesised the NC and functionalized multiwall carbon nanotubes.The electrochemical performances were investigated using cyclic voltammetry, electrochemical impedance spectroscopy, and differential pulse voltammetry. The synthesized nanomaterials and nanocomposites were investigated for their structure and surface morphology using Fourier-transformed infrared spectroscopy (FTIR), Raman spectroscopy, powder X-ray diffraction (XRD), atomic force microscopy (AFM), transmission electron microscopy (TEM), and field

emission scanning electron microscopy (FESEM). The morphological assessment confirmed the spherical shapes of NC and functionalized multiwall carbon nanotubes, which are homogeneously distributed with a smooth and extended surface area with a porous appearance and a high surface-to-volume ratio of the functionalized multiwall carbon nanotubes and NC. Furthermore, the outstanding electroconductivity of *f*- MWCNTs/NC/GCE facilitates the rapid electron transfer process. Here, NC not only prevents the leaching of nanomaterials but also promotes ionic conductivity and the more binding site with the presence of hydroxyl groups that could efficiently adsorb an increased number of moieties. The *f*- MWCNTs/NC/GCE nanocomposite was utilized for the sensitive detection of DCF in biological fluids and pharmaceutical preparations. The electrode response was linear towards the diclofenac sodium (DCF)concentration of 50 µM, with a broad linear range from 0.05 to 1.00 µM and 2.00 to 250 µM, and a low limit of detection of 0.012 µM. The fabricated sensor showed good sensitivity and selectivity toward the electrochemical determination of DCF. The sensor's selectivity was also investigated successfully in the presence of various interfering substances [1].

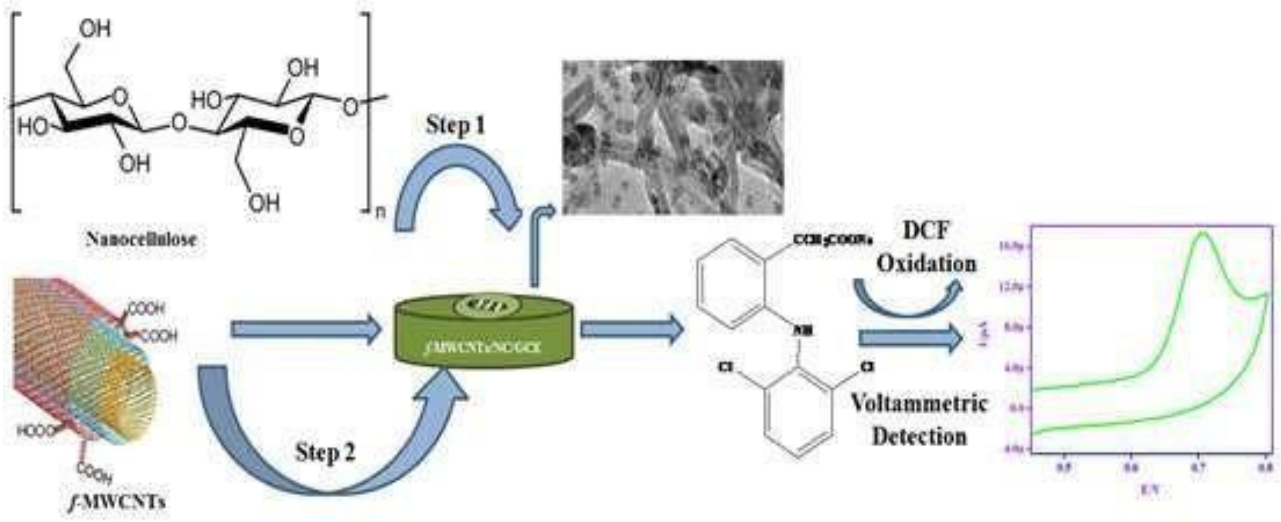

Figure 3: Fabrication of *f*-MWCNT/NC/ GC modified electrode for the detection of DCF [1]

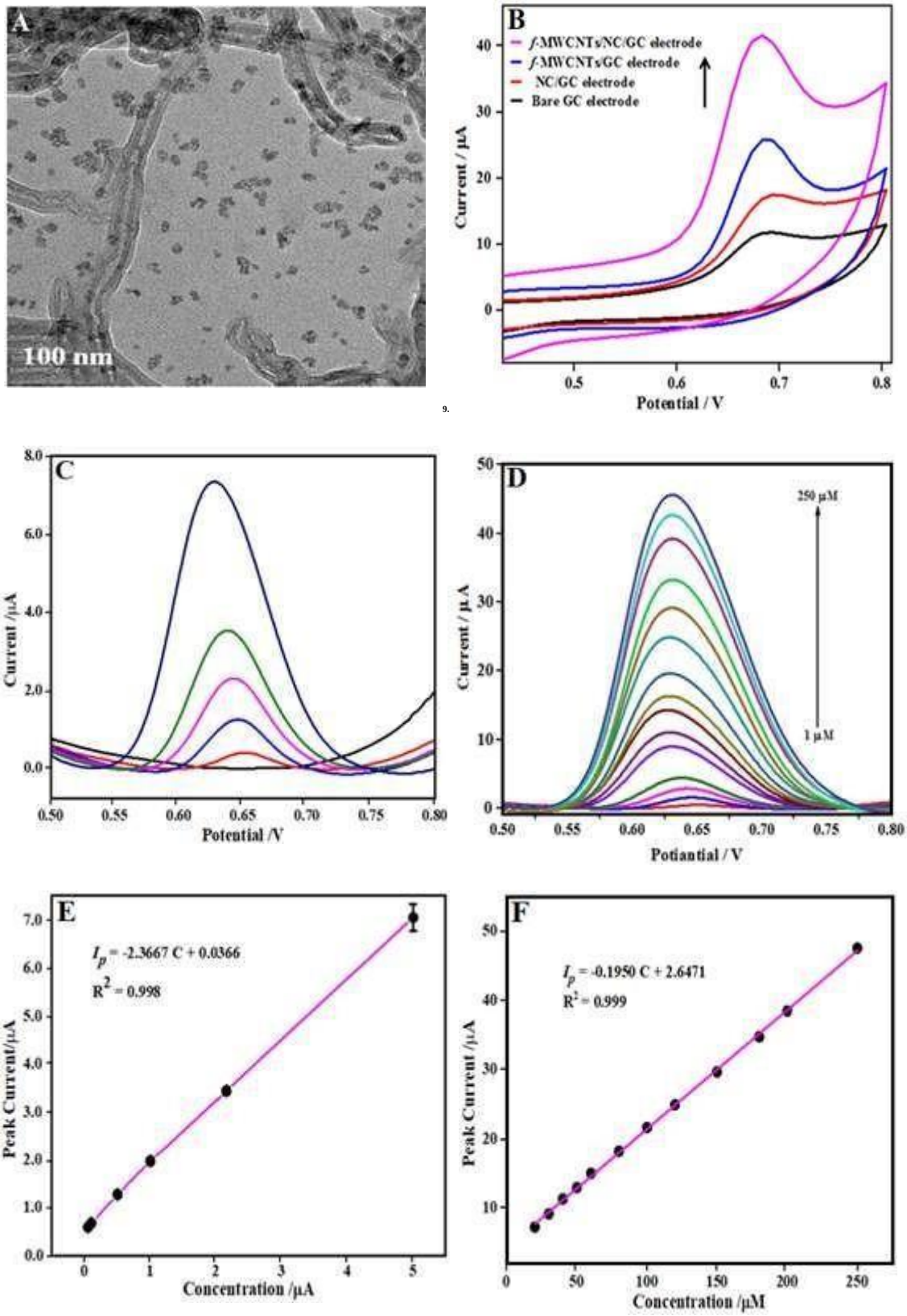

Figure 4: (A) TEM image of NC / *f*-MWCNTs composite, (B) CV of the bare GC electrode (black solid curve),

NC/ GC electrode (red solid curve), *f*-MWCNTs/GC electrode (blue-solid curve) and *f*-MWCNTs/NC/GC electrode (pink- solid curve), in 50 µM DCF with 0.2 M PBS (pH 4.0) at 100 mV s^{-1}. (C) DPV response at *f*-MWCNTs /NC/GC electrode in 0.2 M PBS (pH 4.0) with different concentration for DCF between 0.05 µM – 5 µM. (E) Calibration curve (i.e. peak currents *vs.* DCF concentration) from graph C. (D) DPV response at *f*-MWCNTs /NC/GC electrode in PBS (pH 4.0) containing different concentration for DCF between 0.05 µM – 250 µM. (F) Calibration curve (i.e. peak current *vs.* DCF concentration) from graph D, containing different concentration for DCF between 6 µM – 250 µM [1]

9. Application of NNC-PPY/SWCNT nanocomposite for the simultaneous determination of paracetamol and ciprofloxacin

Shalauddin and his group fabricated another electrode nanocellulose (NNC), polypyrrole (PPY) and single-wall carbon nanotubes (SWCNTs), (NNC-PPY/SWCNTs) where there used conductive polymer PPY with NNC and synthesized followed by chemical polymerization and addition of SWCNTs by ultrasonication method for the simultaneous determination of paracetamol (PCM) and ciprofloxacin (CPR) in the pharmaceutical preparations, biological fluids and water samples [32]. Here, NNC has been synthesized by the same acid hydrolysis method. The layers of PPY were distributed homogeneously on NNC fibers through chemical polymerization. The presence of OH- groups in NNC and NH- groups of PPY are interconnected through a strong hydrogen bond, demonstrating that the incorporation of PPY with the NNC ultimately offers a 3D network with a mechanically stretchable structure [35]. In addition, when SWCNT is incorporated with NNC-PPY, it possesses a large surface area with a porous appearance [36]. The attachment of NNC and PPY with SWCNT generates more ionic pathways, accelerating ion transportation and ultimately enhancing the electroconductivity and electrocatalytic performance of NNC- PPY/SWCNT. The NNC-PPY/SWCNT nanocomposite morphological characteristics were investigated by SEM, TEM, FTIR, and Raman spectroscopy. The electrochemical performance of NNC-PPY nanocomposite was investigated by CV, EIS, and DPV methods. The proposed NNC-PPY modified GCE sensor showed a sharp electrochemical response towards the redox of PCM and oxidation of CPR with a dynamic linear range of 0.01 to 100 µM for PCM and 0.05 to 80 µM for CPR. The calculated LOD was 0.0017 µM for PCM and 0.0011 µM for CPR. The selectivity of the NNC-PPY/SWCNT modified GCE sensor was assessed through the interference test in the presence of some common interfering molecules. From the interference test, it was shown that, except for dopamine (DA) and ascorbic acid (AA), the interfering molecules did not interfere with the redox peak current of PCM and the oxidation peak of CPR. It could be clearly demonstrated that NNC-PPY/SWCNT-modified

GCE sensor has high selectivity towards the PCM and CPR. Moreover, the fabricated electrode is also efficient for simultaneously determining PCM and CPR from a wastewater sample. Therefore, the real sample analysis was performed in pharmaceutical dosage forms and biological fluids to evaluate the practical application of the proposed electrode with satisfactory recovery. The results demonstrated that the NNC-PPY/SWCNT-modified GCE is a potential candidate for the simultaneous determination of PCM and CPR in pharmaceutical dosage forms and biological fluids [32].

Figure 5: Fabrication of NNC-PPY-modified GC electrode for the detection of PCT and CPR [32]

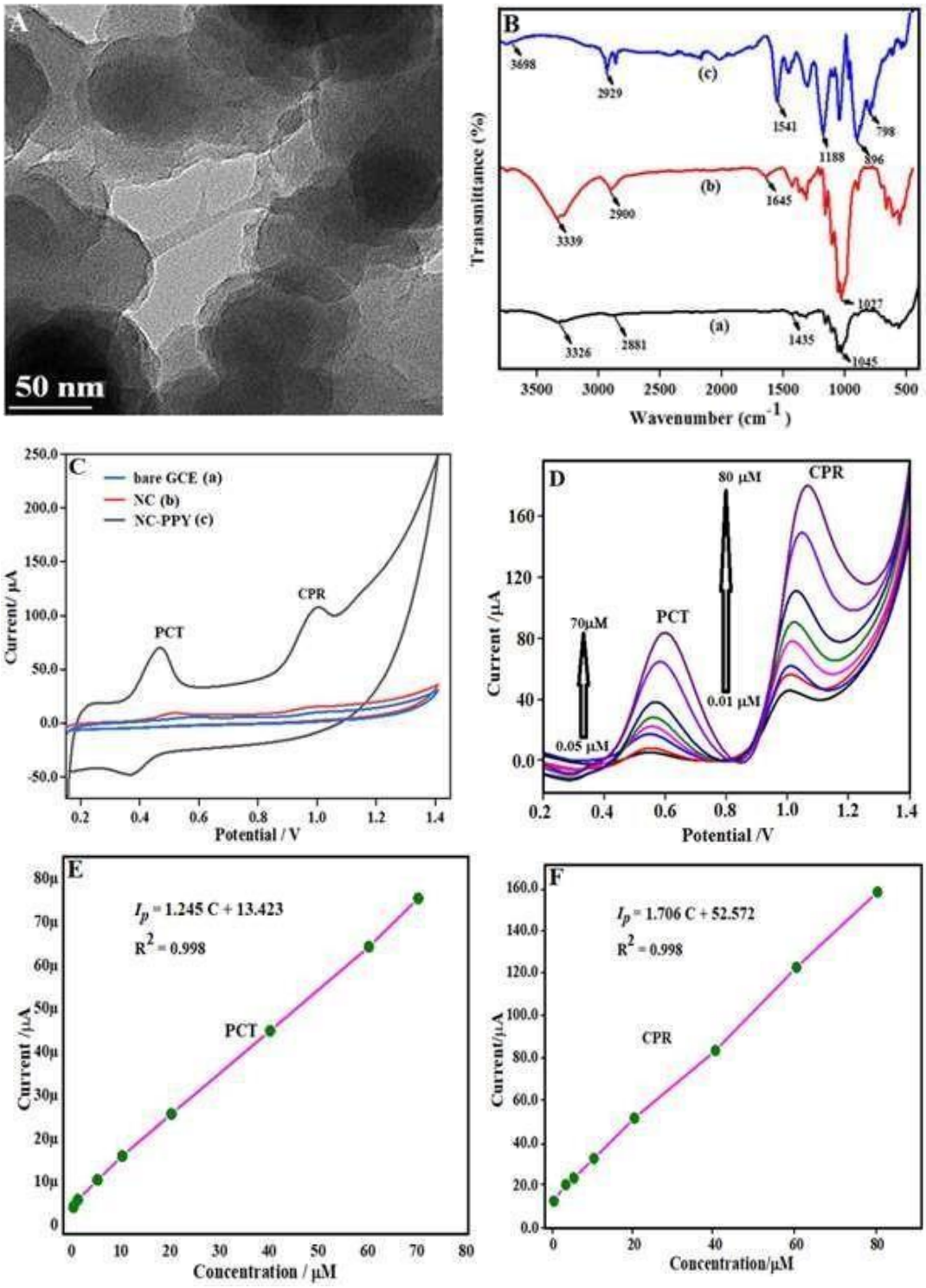

Figure 6: (A) TEM images of NC-PPY (higher resolution), (B) FTIR spectra of cellulose(a), NC (b), NC- PPY(c), (C) CVs of 45 µM PCT and 30 µM CPR in 0.1 M PBS (pH 7.0), potential range 0.2 V to 1.4 V and scan rate at 0.1 Vs^{-1} on (a) bare GCE, (b) NC/GCE and (c) NC-PPY/GCE. (D) SWV at NC-PPY/GCE at different concentrations of PCT (0.05–70 µM) and CPR (0.01–80 µM) in 0.1 M PBS (pH 7.0) at 0.1 V s^{-1}, (E) Calibration curve of PCT concentration *vs.* current

(0.05 –70 µM), (F) Calibration curve of CPR concentration *vs.* current (0.01 – 80 µM) [32]

10. Conclusions and future work

NC is considered as a rising star functional material in this present time. The eco-friendliness, abundance, biocompatibility, and biodegradability of NC make it an alluring candidate for multi-purposed materials. The combination of NC and carbonaceous nanomaterials could contribute to the electrochemical sensing of desired analytes. The performance of the nanomaterials depends on the particle's size, shape, crystallinity, and morphology. The efficacy of the nanocomposite depends on several factors such as film formation, proper distribution of nanoparticles within the NC matrices, expanded electroactive surface area, available binding sites for the chemical modifications, enhanced electrocatalytic activity, successful interactions between the carbon nanostructured material and biopolymers (such as hydrophobic-hydrophobic interactions, polar-polar interactions, hydrogen bonds, π-πinteractions, van der Waals forces, ionic interactions, covalent interactions and others). NC is an ideal dispersing agent for carbon nanotubes and graphene components. These types of nanocomposites are effective for sensing applications of drug molecules and heavy metal ions. However, metal oxide, metal ions, metal nanoparticles, semiconductor nanoparticles or different types of MOFs can be incorporated with NC-based carbon nanocomposites for enhancing electrochemical performances and prominent electrocatalytic effect, which could be further investigated for energy storage, carbon reduction, biosensor, supercapacitor, water splitting and battery applications.

Conflicts of interest

The authors declare no conflict of interest.

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References

1. M Shalauddin et al. Hybrid nanocellulose/f-MWCNTs nanocomposite for the electrochemical sensing of diclofenac sodium in pharmaceutical drugs and biological fluids. *Electrochim. Acta.* 304, 323-333. (2019)

2. A Hashem et al., Applications of nanocellulose as biosensing platforms for the detection of functional biomacromolecules: A Review. Malaysian NANO-An International Journal. 2,1; 15-45. (2022)

3. C Sasso et al., Highly conducting polypyrrole/cellulose nanocomposite films with enhanced mechanical properties. *Macromol Mater Eng*. 295,10; 934-941. (2010)

4. JM Malho et al., Facile method for stiff, tough, and strong nanocomposites by direct exfoliation of multilayered graphene into native nanocellulose matrix. *Biomacromolecules*. 13,4;1093-1099. (2012)

5. H Koga et al., Transparent, conductive, and printable composites consisting of TEMPO- oxidized nanocellulose and carbon nanotube. *Biomacromolecules*. 14,4; 1160-1165. (2013)

6. M Shalauddin et al., Bio-synthesized copper nanoparticle decorated multiwall carbon nanotube- nanocellulose nanocomposite: An electrochemical sensor for the simultaneous detection of acetaminophen and diclofenac sodium. *Surf. Interfaces*. 34, 102385. (2022)

7. M Shalauddin et al., Immobilized copper ions on MWCNTS-Chitosan thin film: Enhanced amperometric sensor for electrochemical determination of diclofenac sodium in aqueous solution. *Int. J. Hydrog Energy*. 42,31; 19951-19960. (2017)

8. S Akhter et al., Enhanced amperometric detection of paracetamol by immobilized cobalt ionon functionalized MWCNTs-Chitosan thin film. *Anal. Biochem*. 551, 29-36. (2018)

9. S Akhter et al. Hybrid nanocomposite of functionalized multiwall carbon nanotube, nitrogen doped graphene and chitosan with electrodeposited copper for the detection of anticancer drug nilutamide in tablet and biological samples. *Mater. Chem. Phys*. 253,123393. (2020)

10. M Shalauddin et al., Carboxylated nanocellulose dispersed nitrogen doped graphene nanosheets and sodium dodecyl sulfate modified electrochemical sensor for the simultaneous determination of paracetamol and naproxen sodium. Measurement. 194,110961. (2022)

11. S Akhter et al. Tri-metallic Co-Ni-Cu based metal organic framework nanostructures for thedetection of an anticancer drug nilutamide. *Sens. Actuator A Phys*. 325, 112711. (2021)

12. NK Shrestha, X Du, and J Wang. Assessing climate change impacts on fresh water resourcesof the Athabasca River Basin, Canada. *Sci. Total Environ*. 601, 425-440. (2017)

13. F Markoulidis et al. Composite electrodes of activated carbon and multiwall carbon nanotubes decorated with silver nanoparticles for high power energy storage. *J. Compos. Sci*. 3,4; 97. (2019)

14. H Kargarzadeh et al. Handbook of nanocellulose and cellulose nanocomposites. *Wiley Online Library*. (2017)

15. X Xiong et al. Sources and distribution of microplastics in China's largest inland lake– Qinghai Lake. *Environ. Pollut.* 235, 899-906. (2018)

16. K Hettrich et al. Novel aspects of nanocellulose. *Cellulose*. 21,4; 2479-2488. (2014)

17. Y Wang et al. Study on nanocellulose by high pressure homogenization in homogeneous isolation. *Fibers Polym*. 16,3;572-578. (2015)

18. JH Kim et al. Review of nanocellulose for sustainable future materials. *Int. J. Pr. Eng. Man.-GT*. 2, 2; 197-213. (2015)

19. M Rosa et al. Cellulose nanowhiskers from coconut husk fibers: Effect of preparation conditions on their thermal and morphological behavior. *Carbohydr. Polym*. 81,1;83-92. (2010)

20. F Wesarg et al. In situ synthesis of photocatalytically active hybrids consisting of bacterial nanocellulose and anatase nanoparticles. Langmuir. 28, 37; 13518-13525. (2012)

21. H Wang et al. A comparison study on the preparation of nanocellulose fibrils from fibers and parenchymal cells in bamboo (Phyllostachys pubescens). *Ind. Crops Prod*. 71, 80-88. (2015)

22. S Mondal. Preparation, properties and applications of nanocellulosic materials. *Carbohydr. Polym*.163, 301-316. (2017)

23. F Niu et al. The characteristic and dispersion stability of nanocellulose produced by mixed acid hydrolysis and ultrasonic assistance. *Carbohydr. Polym*. 165, 197-204. (2017)

24. J Zhang et al. Facile synthesis of spherical cellulose nanoparticles. *Carbohydr. Polym.* 69,3; 607-611. (2007)

25. S Taokaew et al. Biosynthesis and characterization of nanocellulose-gelatin films. *Materials*. 6,3; 782-794. (2013)

26. M Shalauddin et al. Immobilized copper ions on MWCNTS-Chitosan thin film: Enhanced amperometric sensor for electrochemical determination of diclofenac sodium in aqueous solution. *Int. J. Hydrog. Energy*. 42,31; 19951-19960. (2017).

27. H Bagheri et al. Composite of Cu metal nanoparticles-multiwall carbon nanotubes-reduced graphene oxide as a novel and high performance platform of the electrochemical sensor for simultaneous determination of nitrite and nitrate. *J. Hazard. mater*. 324,762-772. (2017)

28. J Li and EC Lee. Functionalized multi-wall carbon nanotubes as an efficient additive for electrochemical DNA sensor. *Sens. Actuators B Chem*. 239, 652-659. (2017)

29. V Gautam, KP Singh and VL Yadav. Polyaniline/multiwall carbon nanotubes/starch nanocomposite material and hemoglobin modified carbon paste electrode for hydrogen peroxideand glucose biosensing. *Int. J. Biol. Macromol*. 111, 1124-1132. (2018)

30. BK Shrestha et al. Bio-inspired hybrid scaffold of zinc oxide-functionalized multi-wall carbon nanotubes reinforced polyurethane nanofibers for bone tissue engineering. *Mater. Des.* 133, 69-81. (2017)

31. S Mallakpour and S Soltanian. Surface functionalization of carbon nanotubes: fabrication and applications. *RSC Adv*. 6,111; 109916-109935. (2016)

32. M Shalauddin et al. A metal free nanosensor based on nanocellulose-polypyrrole matrix and single- walled carbon nanotube: experimental study and electroanalytical application for determination of paracetamol and ciprofloxacin. *Environ. Nanotechnol. Monit. Manag.* 18, 100691. (2002)

33. C Wan et al. Recent progress in flexible nanocellulosic structures for wearable piezoresistive strain sensors. *J. Mater. Chem. C*. 9,34; 11001-11029. (2021)

34. Z Weng et al. Graphene–cellulose paper flexible supercapacitors. *Adv. Energy Mater*. 201,5; 917-922. (2011)

35. S Akhter et al. A highly selective bifunctional nanosensor based on nanocellulose and 3D polypyrrole decorated with silver-gold bimetallic alloy to simultaneously detect methotrexate and ciprofloxacin. *Sens. Actuators B Chem*. 373, 132743. (2022)

36. G Nystrom et al. A nanocellulose polypyrrole composite based on microfibrillated cellulose from wood. *J. Phys. Chem. B*. 114,12; 4178-4182. (2010)