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Mini Review

The Application of Nanomaterials as Electrode Modifiers for the Electrochemical Detection of Ascorbic Acid: Review

Saifeldin M. Siddeeg[1,2], *Norah Salem Alsaiari*[3], *Mohamed A. Tahoon*[1,*] *and Faouzi Ben Rebah*[1,4]

[1] Department of Chemistry, College of Science, King Khalid University, P.O. Box 9004, Abha 61413, Kingdom of Saudi Arabia
[2] Chemistry and Nuclear Physics Institute, Atomic Energy Commission, P.O. Box 3001, Khartoum 11111, Sudan
[3] Department of Chemistry, College of Science, Princess Nourah Bint Abdulrahman University, Riyadh, Saudi Arabia
[4] Higher Institute of Biotechnology of Sfax (ISBS), Sfax University, P.O. Box 263, Sfax 3000, Tunisia
[*]E-mail: tahooon_87@yahoo.com

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Ascorbic acid (AA) is one of the most important biomolecules that play an important role in the synthesis of collagen, norepinephrine and neuronal hormones in addition to its role in folic acid tyrosine and tryptophan metabolisms. Also, vitamin C participates in many biochemical processes; it functions as a cofactor for several enzymes, enhances the intestinal iron absorption and is involved in carnitine biosynthesis and cellular metabolism. Various analytical methods with high sensitivity and selectivity have been developed for ascorbic acid quantitative detection. Recently, a highly sensitive, effective and rapid ascorbic acid detection was achieved using nanomaterials-based sensors. Herein, we discussed the latest published articles for the ascorbic acid sensors based on nanomaterials and their fabrication.

Keywords: Ascorbic acid; Electrochemical sensors; Nanomaterials

1. INTRODUCTION

Electrochemical studies have significant importance in the last years due to their potential applications in different chemical and industrial processes [1-5]. Biosensors are analytical instruments that turn a biological reaction into an observable test. Biosensors have to be highly specialized, reusable and sensitive to specific analytes. Cammann recognized the term "biosensor" and IUPAC presented its description [6]. Since the 1960s, biosensors that began the development of glucose oxidase (GOx) have been introduced and renowned [7]. This biosensor was used for blood glucose monitoring in diabetes patients. Since then, different sensors with a variety of applications have been developed [8]. With the ability to reduce or oxidize different electro-active molecules at different potential values, electrochemical sensors can determine the redox reactions mechanism. Furthermore, electrochemical techniques for small volumes of analyte content biofluids can be used because they have a good detection limit (LOD) [9]. Due to the human interest and world market demand, several new sensors were developed using simple devices that can precisely detect the analyte concentration in samples, such as urine, blood, and serum. Therefore, many attempts were performed by researchers to determine L-ascorbic acid in different biological samples. L-ascorbic acid, also known as vitamin C, is a water-soluble vitamin essential for human nutrients. The acid cannot be synthesized inside the human body because of the absence of l-gulono-gamma-lactone oxidase enzyme required for vitamin C synthesis from D-glucose [10]. Therefore, vitamin C, which is found in foods, is essential for the synthesis of collagen, norepinephrine and neuronal hormones; also, it is required for folic acid, tyrosine, and tryptophan metabolism [11]. Vitamin C participates in many biochemical processes as a cofactor for several enzymes. It is also involved in intestinal iron absorption, carnitine biosynthesis and cellular metabolism as a reduction agent [12]. The daily requirements of vitamin C are 90 mg/day and 75 mg/day for men and women, respectively as recommended by Recommended Dietary Allowances (RDA) [13]. There are many types of food that are rich in vitamin C such as potatoes, peppers, tomatoes, brussels sprouts, cauliflower, and broccoli [14]. Eye tissues and fluids, brain, leukocytes, adrenal gland, and pituitary gland contain the high levels of vitamin C [15] while a significant amount is absorbed inside the gastrointestinal tract (GIT) then transferred to other tissues via plasma [16]. Vitamin C is a superb reducing agent known as a scavenger of free radicals in biological systems because of its high anti-oxidant activity, leading to the neutralization of free radicals by the formation of the radical ascorbate as an intermediate via hydrogen radical interaction. Ascorbate radical is a relatively non-reactive free radical with its unpaired electron within a strongly relocated π system which makes it a superior biological donor antioxidant ascorbate [17]. Cell damage resulting from reactive oxygen species and toxic free radicals can be inhibited using vitamin C [18]. Furthermore, ascorbic acid effectively interacts with oxidants in the aqueous media leading to the protection of low-density lipoprotein (LDL) and human blood lipids toward oxidative damage [19].

The accurate determination of vitamin C is still present as one of the most interesting area of research because of its high important for both human lives and industrial applications. Electrochemical techniques [20], fluorescence [21], titrimetry [22], chemiluminescence [23], solid-phase iodine [24], spectrophotometry [25], electrophoresis [26], and High Performance Liquid Chromatography (HPLC) [27] are the common used techniques for ascorbic acid detection. Overall techniques, the electrochemical method represents by sensors is the most used method by researchers for ascorbic acid detection due to easy operation, high sensitivity, low cost, and simplicity. Conventional bare electrodes like Pt, Au, and glassy carbon were used for the ascorbic acid detection but the high voltage of vitamin over these electrodes lead to fouling in detection leading to low sensitivity and selectivity. In order to decrease vitamin C overpotential, nanomaterials were used for the modification of conventional electrodes to enhance the electrode sensitivity, selectivity, and reproducibility toward vitamin detection. So, in the current study, we review the nanomaterials based sensors used recently for ascorbic acid.

2. PRINCIPLE

Electrochemical sensors are based on the transfer of charges between two phases; the liquid or solid sample and the electrode in which chemical change happens for both the sample phase and the electrode. The sensing process depends on the reactions at the electrode or on the transported charge that can be chemically modulated to serve this sensing [28]. There are three types of sensors conductometric, amperometric, and potentiometric sensors. The electrolyte conductivity measurement is the basis of conductometric sensors that differs by the exposure of the cell to another environment in which the sensing depends on the variation of the number of charge carriers. The amperometric sensors depend on the current changes of the analyte concentration during the oxidation-reduction process. The potentiometric sensors depend on the change of potential between the indicator and reference electrodes.

3. NANOMATERIALS BASED SENSORS OF ASCORBIC ACID

During the last years, the research outcomes related to nanomaterials have been increased in different application fields due to the development in the preparation and application of these new materials [29-38]. One of the most important applications of nanomaterials is the biosensing technology in which nanomaterials enhance the sensors reproducibility, stability, selectivity, sensitivity, and performance. So, the development of biosensors based on nanomaterials for the detection of important biomolecules has attracted the interest of researchers. Many researchers have improved the ascorbic acid electrochemical sensors by using different nanomaterials like carbon nanocomposites, conducting polymers, metals oxides/hydroxides, and metals/noble metals nanoparticles. Thus, in the next sections, the ascorbic acid biosensors will be reviewed depending on their construction using these nanomaterials.

3.1. Conducting polymers

The neutral states of polymers have no conductivity but the conducting polymers have charge carriers due to doping of their conjugated structure which leads to natural conductivity outcome [39]. Conducting polymers can replace semiconductors and metals because of their unusual features like low cost, low weight, tunable electrochemical properties, transparent, and flexibility [40] that allow using of these polymers in different applications like biomedical applications and polymers [41], thin-film transistors [42], solar cells [43], energy storage [44], and light-emitting diodes [45]. Many pharmaceutical compounds, uric acid, dopamine, and ascorbic acid have been detected using conducting polymers [46-49]. High stability, good redox reversibility, and high conductivity of polypyrrole and polyaniline make these two polymers mostly used for the sensing of different compounds using different techniques like conductimetric/impedimetric, voltammetric, and amperometric techniques [50-52]. The addition of polypyrrole and polyaniline polymers to the electrode material improves its electrochemical sensitivity [53]. For example, the copolymer aniline with 3,4-dihydroxybenzoic acid-modified gold electrode in which polyaniline serves as a mediator for ascorbic acid electrocatalytic oxidation and leading to the decrease in ascorbic acid overpotential and subsequently, enhances the electrode sensitivity toward ascorbic acid detection [54]. Likewise, the reproducibility, current response, and reaction kinetics of the gold electrode were enhanced toward the detection of ascorbic acid when this electrode was modified with 4-hydroxy-6-methyl-2-mercaptopyrimidine and polypyrrole [55]. The ascorbic acid electrochemical oxidation showed a more negative potential over polypyrrole/dodecyl sulfate modified electrode than that of the unmodified electrode [56].

The nanomaterials can be used with conducting polymers in order to improve the performance of the sensor toward ascorbic acid sensitivity. These nanomaterials include carbon nanomaterials, metal oxide nanoparticles, and metals nanoparticles. For example, ascorbic acid sensitivity was improved by using silver nanoparticles, multi-walled carbon nanotubes composite with poly neutral red and poly(amido amine) dendrimer to modify paraffin wax impregnated graphite electrode to give an electrode with LOD=0.054 µM, a linear detection range= 0.16-2500 µM, and current response at +0.27 V toward ascorbic acid in commercial samples [57]. Glassy carbon electrode was modified by carbon nanotubes and poly aniline doped with silicotungstic acid to improve ascorbic acid sensitivity. The electrode was used to investigate ascorbic acid in orange juice [58]. In another study, gold nanoparticles and polypyrrole were electrodeposited over titanium oxide nanotubes in order to improve the electrode sensitivity toward ascorbic acid. The obtained electrode was used to investigate ascorbic acid in lemon juice [59]. The high electroactive surface area and high electrocatalytic activity of poly(sulfonazo III) were exploited to modify electrode for ascorbic acid detection in vitamin C tablets offering high sensitivity [60]. Also, ZnO nanoparticles and poly(5-amino-2,3-dihydrophthalazine-1,4-dione) coated electrode was used to highly sense the ascorbic acid at micromolar concentrations and with high stability [61]. The ascorbic acid sensors based on conducting polymers and their composites were presented in Table 1.

Table 1. Ascorbic acid electrochemical sensors based on conducting polymers and their composites

Sensor	**LOD (µM)**	**Sensitivity (µA mM^{-1} cm^{-2})**	**Ref.**
Polypyrrole hydrogel/GCE	1.283	253.52	[62]
Porous g-C_3N_4 PCN/PEDOT/GCE	9.3	450.70	[63]
Poly-Trypan Blue/GCE	0.1	39	[64]
Polyimide doped AuNP	18.49	824.78	[65]
PANI-halloysite nanotubes	0.21	826.53	[66]
PGM silica/CPE	0.97	95.95	[67]
PM silica/CPE	6.76	15.13	[67]
CA-MWCNT-PEDOT	4.2	1699.36	[68]
PANI/MnO_2-Sb_2O_3	$1.05X10^{-3}$	5673.8	[69]
ZnO-Cu_xO/PPy	25	-	[70]
PBG/GCE	1.67	28	[71]
Polyoxometalate/Au –Pd alloy	0.43	-	[72]
PPy/Au/MEA	0.01	2500	[73]

MWCNT/PNB/GCE	2.4	860	[74]
Au-PANI nanosheets	8.2	25.69	[75]
MSA-PANI/MWCNT/GCE	0.6	363	[76]
BPEI-EGDE/[$Fe(CN)_6$] ^{4}/GCE	1	56.3	[77]
BPEI-EGDE/GCE	10	28.2	[77]
SiW_{12}-CNT-PANI/GCE	0.51	22.42	[78]
Methionine/CPE	3.5	8486.8	[78]
$CuGeO_3$/PANI nanowires	0.26	-	[79]
Poly-Xa/MWCNT/GCE	0.19	300	[80]
OPPy-PdNP	1	570	[81]
PANI/PAA/MWCNT	0.25	9458.6	[82]
PANI/SPE	30	17.7	[83]
p-AMTa/GCE	0.25	-	[84]
AuNP/PPy/TiO_2	0.1	63.912	[59]
PANI-OAP/GCE	1.4	-	[85]
poly(Ani-co-m-FcAni)/GCE	2	-	[86]
Pd/PoPD nanocomposite	-	0.75	[87]
PXSP/GCE	4	286.62	[88]
PBCACPM/GCE	0.4	318.47	[89]
Quantum-sized gold nanoparticles/GCE	0.068	155.6	[90]
PEDOT film/Au	2.5	870	[91]
Au/PANI/Ti	-	-	[92]
Poly(sulfonazo III) film/GCE	0.17	191.08	[60]
Poly-ACBK/GCE	10	192.96	[93]
Poly(luminol)/ZnONP	1	4568.6	[61]
PCDDA/GCE	1.43	3503.2	[94]
Au NP/DMT/GCE	0.2	2587	[95]
p-AMT/GCE	$92X10^{-5}$	1415	[96]
Poly (direct blue71)/GCE	1	-	[97]
Poly-CCA/GCE	0.5	-	[98]
Cetylpyridine bromide/chitosan/GCE	-	222.05	[99]
Nano PANI/SPCE	8.3	10.75	[47]
f-MWCNT-PNR	-	-	[100]
Au/HDT/AuNP	1	-	[101]
CoHCF	33.3	-	[102]
PANOAA/Pt	-	-	[103]
PANI/Pt wire electrode	50	124	[104]
Poly N, N-dimethylaniline/GCE	-	0.178	[105]
PANI/GCE	0.4	-	[106]
PPy/hexacyanoferrate/GCE	500	-	[107]

3.2. Carbon nanomaterials

The amazing properties, accessible synthesis, and exceptional structures of carbon nanomaterials (nitrogen-doped carbon materials, carbon nanoparticles, graphene derivatives, carbon nanofibers, carbon nanohorns, carbon nanodots, single/multi-wall carbon nanotubes, etc.) make them as the most used nanomaterials in the electrochemical applications [108, 109]. Ascorbic acid was electrochemically detected using different allotropes of carbon nanomaterials as discussed in the next sub-sections.

3.2.1. Graphene

The graphene-based materials were widely used in different applications like solar cells [110], lithium-ion batteries [111], supercapacitors [112], fuel cells [113], and biosensors [114] because of their exceptional properties including excellent electrical and thermal conductivity, chemical stability, high mechanical strength, and large surface area. The GCE was modified using NiO NPs anchored graphene composite in order to improve the electrode sensitivity toward ascorbic acid by benefiting the NiO electrocatalytic activity and the graphene high electrical conductivity. The performance of the prepared sensor was tested for vitamin C tablets [115]. Also, ascorbic acid was detected in a serum sample using gold nanoplates-decorated graphene in which the sensor showed high sensitivity, LOD, anti-interference and low detection potential [116]. Pt nanoparticles were used to coat graphene-GCE modified electrode in order to enhance its sensitivity toward uric acid, dopamine, and ascorbic acid in which graphene allowed the increase the electrode electrical conductivity and specific surface area [117]. The pristine graphene prepared via the organic salt-assisted exfoliation method was used for the detection of ascorbic acid. This material showed excellent characteristics compared to chemically converted graphene [118]. In order to be used in the detection of uric acid, dopamine, and ascorbic acid, GCE was modified using electrochemically reduced GO. The resulted material offered LOD=250 μM, detection range 2 mM - 500 μM, and higher electrocatalytic activity than that of the bare electrode [119]. Interestingly, the wide linearity, the long-term stability, the reproducibility, and the sensitivity of the prepared graphene-doped carbon paste electrode toward ascorbic acid were better than the unmodified carbon paste electrode [120]. Additionally, graphene nano-sheets modified pyrolyzed photoresist films based electrode showed high sensitivity and accuracy toward ascorbic acid detection in vitamin C commercial tablets [121]. Examples of carbon nanomaterials based sensors for ascorbic acid detection were presented in Table 2.

3.2.2. Carbon nanotubes and hybrid nanomaterials

Due to their interesting characteristics (variable surface chemistry, mechanical resistance, high electrical conductivity, etc.) carbon nanotubes were widely used in electrode materials [122]. The ascorbic acid sensitivity was extremely improved up to 16.29 mM when the MWCNTs modified Cu substrate was loaded with Cu_2O and Ag_2O bimetallic nanoparticles using the electrodeposition process [123].

Similarly, the ascorbic acid detection was improved by using the modified carbon paste electrode with Pd nanoparticles, ionic liquid (IL), and MWCNTs. A paraffin oil was mixed with ionic liquid and MWCNTs in order to deposit Pd nanoparticles over carbon paste electrode via chronoamperometry method. Due to the high conductivity and electroactive surface area of MWCNT, the obtained electrode provided a high ascorbic acid sensitivity and 112 μM linearity in serum and urine samples [124]. The carbon paste electrode was modified using Ag nanoparticles and CNTs allowing high sensitive detection of ascorbic acid with LOD=12 μM and linearity 0.3-2000 μM [125]. The ascorbic acid was also determined using MWCNTs modified electrode (LOD=7.1 μM, linearity of 100 –600 μM, and sensitivity1287.3 μA mM^{-1} cm^{-2}) [126]. In another study, the SWCNTs prepared via CO_2 laser ablation method at room temperature were used to modify GCE in order to enhance its sensitivity toward ascorbic acid in pharmaceuticals and serum. Interestingly, the modified electrode showed excellent properties [127].

3.2.3. Other Carbon nanomaterials

The easy mass production and simple synthetic methods of new class carbon allotropes nanomaterials (carbon dots, nitrogen-doped carbon nanomaterials, carbon nanofibers, etc.) allowed their use in ascorbic acid biosensor. Fullerenes are known to be 3D- nanomaterials that have the ability to gain electrons. Fullerenes can accept multiple electrons reversibly and generate stable intermediate multi-anions. The partially reduced fullerenes are conductive enough to be used to detect various organic compounds and biomolecules as modified electrodes [128]. Though, there was less consideration of fullerenes compounds in the electrochemical sensors. Lately, ascorbic acid in plasma and urine samples was detected over Pt nanosheet and fullerene (C60) modified electrodes in which C60 was used to increase the surface area of GCE electrode for Pt potentiostatic deposition providing a modified electrode with LOD = 0.43 μM, and linearity 10 - 1800 μM toward ascorbic acid detection with a better electrocatalytic response compared to unmodified electrode [129]. Carbon-supported $NiCoO_2$ nanoparticles modified electrode was also used to detect ascorbic acid in 5 mM KOH solution and 85 mM phosphate buffer. Interestingly, the presence of metal ions made the electrode surface positively charged increasing its ability to attract negatively charged vitamin on its surfaces providing high electrocatalytic detection of the ascorbic acid with excellent sensitivity [130]. Additionally, carbon nitride [131-133], carbon fibers [134, 135], and nitrogen-doped carbon [136-138] were successfully examined for ascorbic acid detection. The GCE was modified using PdNi bimetallic nanoparticles supported the carbon black composite that prepared by the synthesis of PdNi nanoparticles via simple reduction method then PdNi nanoparticles were dispersed in carbon black suspension in order to produce better sensitivity toward ascorbic acid detection than the Pd/C catalyst [139]. Besides, carbon nanofibers decorated with Pd nanoparticles prepared via the electrospinning method by mixing 4.8 wt% $Pd(acetate)_2$ and 8 wt% polyacrylonitrile in DMF solvent and tracked by thermal treatment was used for the electrocatalytic detection of ascorbic acid in real samples with high sensitivity [140].

Table 2. Ascorbic acid electrochemical sensors based on carbon nanomaterials and their composites

Sensor	**LOD (μM)**	**Sensitivity (μA mM^{-1} cm^{-2})**	**Ref.**
Graphene/zinc bismuthate nanorods	0.07	15.44	[141]
Pt nanochains-MWCNT-GNP/GCE	10	32.36	[142]
GO/$TmPO_4$/GCE	39	12.39	[143]

Pt@NP-AuSn/Ni/CFP	13.4	140	[144]
TiO_2-rGO/GCE	1.19	1061	[145]
$ZnCl_2$-CF/GCE	0.02	5.66	[146]
hCNT-4ABA/Au-IDA	0.65	-	[147]
Co_3O_4/nanoporous carbon	0.02	130	[148]
Mesoporous carbon nanorods	2.3	216.91	[149]
Graphene-PtNP/GCE	300	-	[150]
hierarchical bayberry-like-Ni@carbon hollow nanospheres/Rgo	0.37	17.03	[151]
Graphene-AuNP/SPE	1.04	-	[152]
Co_3O_4/Fe_3O_4/C-loaded g-C_3N_4	24.75	33.12	[153]
Au-IDA/hCNT	20	1.3	[154]
Graphene-Ag nanorods	0.88	-	[155]
rGO-SnO_2/GCE	38.7	1200	[156]
Graphene ink	17.8	21.14	[157]
CeO_2/MWCNT/PGE	0.008	-	[158]
ZnO–rGO/GCE	1.2	-	[159]
Carbon black-chitosan ink/GCE	0.1	1950	[160]
GO-XDA-Mn_2O_3	0.6	655.74	[161]
MWCNTs-Cu_2O-Ag_2O	0.011	-	[123]
3D graphene hydrogel/ferrocene hybrid	0.183	-	[162]
AgNP/P(Arg)-GO	0.984	422.53	[163]
Alumina nanofibers graphene foliates/(ANF-C700)	0.117	1060	[164]
PdAu/rGO/GCE	12.5	615.57	[165]
3D graphene hydrogel -AuNP	0.028	216.19	[166]
Fe_3O_4-SnO_2-graphene	0.063	20901	[167]
AuNP-GO/Au-IDA	1.4	0.83	[168]
MoS_2/rGO	0.72	152.11	[169]
Carbon-supported NiNP	5	352	[170]
rGO–ZnO composite	3.71	29.04	[171]
Carbon fibers@ZnO core-shell NP	170.1	57.49	[172]
Carbon fibers/ZnO	156.7	1.802	[173]
PdAg nanoflowers/rGO	0.057	-	[174]
Graphene sheet/graphene nanoribbon	0.23	22	[175]
rGO-CNT/ITO	5.31	-	[176]
MgO nanobelt-graphene-Ta wire	0.03	-	[177]
Graphene–$Gd(OH)_3$	60	116.4	[178]
Pristine graphene	6.45	65.62	[118]
Graphene/wrapped TiO_2	0.512	92.53	[179]
AuCo alloy NP/HS-graphene	4	96.6	[180]
Cu-MOFs-MPC/GCE	3.5	1178.03	[181]
Exfoliated flexible graphite paper	2	10	[182]
Fe_3O_4@ nitrogen doped CNT	0.24	1408	[183]
3D graphene foam/CuO nanoflowers	0.43	2060	[184]
Ru(II)/ZnO/CNT	0.005	502	[185]
Tryptophan/graphene	10.09	149.29	[186]
$NiCoO_2$/C nanoparticles	0.5	549.3	[130]
AuNP/GO	0.1	101.86	[187]

NiO/graphene	0.017	139.43	[115]
rGO/Au nanoplates	0.51	98.59	[116]
ErGO/carbon fiber electrode	4.5	-	[188]
Fe_3O_4-NH_2@graphene	0.074	-	[189]
CoPd/carbon/GCE	0.1	1179	[190]
Gr/CuPc/PANI/SPE	0.063	24.46	[191]
Au@Pd-rGO/GCE	0.02	0.31	[192]
rGO-PAO/CCE	0.3	703.9	[193]
Pd-NP MWCNT-IL/CPE	0.2	3631	[124]
Graphene flowers/carbon fiber electrode	24.7	164	[194]
AgNP/rGO/GCE	9.6	6478.9	[195]
SWCNT-ZnO/GCE	8.5	190	[196]
Pd–Pt/rGO	0.61	-	[197]
Carbon-supported PdNi nanoparticles	0.5	760.6	[139]
Fe_3O_4/rGO/GCE	20	33.5	[198]
SPGNE	0.95	-	[199]
PdNP-GO	-	6.148	[200]
CoPc–MWNTs/GCE	1	444.19	[201]
HNCMS/GC	0.91	422.54	[138]
Graphene/size-selected Pt	0.15	1761.53	[117]
RuOH/MWCNT/GCE	0.087	130200	[202]
graphene doped CPE	0.07	3.316	[120]
Ag hexacyanoferrate NP/CNT	0.42	4847.13	[203]
Graphene nanosheets	120	260.56	[121]
MRAC/pyrolytic graphite electrode	0.3	-	[204]
MWNT-Silica-AuNP/GCE	-	8.59	[205]
Ag/CNT-CPE	12	37.65	[125]
Fe^{3+}Y/ZCME/graphite	1.85	-	[206]

3.3. Bimetallic, metal oxides, and transition metals nanoparticles

In order to benefit from their nanostructures properties, metals were used in the fabrication of modified electrodes with low cost to improve its sensitivity toward biomolecules such as ascorbic acid. Lately, bimetallic/alloy nanoparticles, metal oxides, and transition metal nanoparticles became the most favored nanostructures in the biosensors materials. Thus, a number of bimetallic/alloy nanoparticles, and metals/oxides nanoparticles based ascorbic acid sensors was introduced in the present section. The unusual chemical and physical properties of multi-metallic nanomaterials make these nanomaterials useful for ascorbic acid sensing. Also, we can get morphology variation, particle size variation, and intermetallic charge hybridized multi-metals by metals addition. For example, GCE was modified using Au-Ru nanoshells to enhance ascorbic acid sensing. Remarkably, the high operational stability of Au-Ru nanoshells/GCE was exceptional without any supporting chemicals. The modified electrode was tested for the ascorbic acid determination in vitamin C tablets and urine samples [207]. ZnO nanorodsAuNP prepared via the hydrothermal method and in-situ plasma sputtering-assisted technique was used to modify GCE. The modified electrode showed an enhancement of its sensitivity toward ascorbic acid (sensitivity=264.16 μA mM^{-1} cm^{-2} and linearity= 100 –40000 μM) [208].

The ascorbic acid was detected using Pt alloy coarse grains and Ni spinodal crystals with high sensitivity in 0.1 M KOH solution. Commonly, interfering biomolecules showed strong resistance to bending over this electrode surface. The negatively charged ascorbic acid was attracted to the positively charged electrode via adsorption mechanism [209]. Anodic alumina oxide (AAO) using the template-assisted method was used to prepare nanoporous Fe_2O_3 on Au film that used to modify ITO electrode in order to enhance its sensitivity toward ascorbic acid detection inside 0.2 M NaOH. The modified electrode has an exceptional resistance toward interfering biomolecules [210].

The GCE was modified using Co film via the electrodeposition in order to improve its sensitivity toward ascorbic acid. Different electrochemical techniques were used to study kinetic parameters and electrocatalytic properties. The modified electrode was tested for ascorbic acid detection in juices via amperometric method exhibiting high sensitivity of the modified electrode with limited linearity to micro-molar levels [211]. The naturally low-cost abundant copper metal was used in different applications due to its excellent catalytic and electrochemical properties. So, ascorbic acid sensors were made using copper and its compounds. For example, a tetrahydrofuran and PVC mixture was used to suspend the grounded zeolite type A loading Cu^{2+} and Graphite powder then, the suspension was used to modify Pt electrode to enhance its sensitivity toward ascorbic acid detection in 0.1 M oxalate buffer containing 0.5 M $NaNO_3$. The modified electrode was tested for ascorbic acid detection in chewable tablet and effervescent tablets samples [212]. Additionally, ITO altered with nanostructures of copper metal was employed to modify GCE to be used via differential pulse voltammetry (DPV) to detect ascorbic acid with linearity= 1-40 μM [213]. Furthermore, the GCE was modified using the one-spot hydrothermal synthesized ZnO nanoparticles and used for the electrochemical detection of ascorbic acid. The modified electrode showed LOD= 0.312 μM, and linearity= 1 -800 μM with exceptional an anti-interference, repeatability, and stability nature [214].

3.4. Noble metals and its nanostructures ascorbic acid sensors

Compared to other nanomaterials, the noble metals have various properties (excellent stability at ambient conditions, biocompatibility, catalytic activity, easy size tenability and high surface/volume ratio) which enhance their using in different application fields like biochemical sensing, therapeutics, bioimaging, immunoassays, and molecular diagnostics. Depending on noble metals nanostructures properties, these nanoparticles have been used for biomolecular electrochemical sensing like glucose [215], tirapazamine pharmaceutical compounds [216], dopamine [217], and ascorbic acid [218]. Ascorbic acid was detected using many noble metal nanoparticles modified electrodes with high sensitivity and low overpotential providing high rate of electron transfer, high surface active area, and rapid current response. For example, Pd nanostructures catalysts were used for ascorbic acid detection due to their higher abundance and lower cost than other metals. The GCE was modified using Pd nanowires to enhance its sensitivity toward ascorbic acid with LOD=0.2 μM and linearity= 25-0.9 mM [219]. The effective in catalysis, electron-rich core, high surface/volume ratio, high stability, numerous surface functionalities, narrow size distribution, and biocompatibility of gold nanoparticles allowed their use in different biomedical applications. The GCE was modified using self-assembling Au nanoparticles for the simultaneous detection of ascorbic acid in the presence of competitive uric acid with acceptable results [220]. Also, GCE modified with Au nanoparticles was used for the detection of dopamine and ascorbic acid with low detection limit and high selectivity [221]. Additionally, the ascorbic acid detection mechanism was discussed over the Au electrode modified with mercapto carboxylic acid in which two electrons and two protons involved mechanism at acidic pH while found to be two electrons and one proton at basic pH [222]. Similarly, Pt electrodes were used for biomolecular detections due to their high mass transport characteristics and rapid transfer of electrons. Due to their ability to adsorb ascorbic acid on its surface via hydrogen adsorption, Pt electrodes were used for the electrochemical detection of ascorbic acid in several studies [223, 224]. Also, ascorbic acid was detected in apple juice using Pt electrodes by voltammetric technique [225]. In another study, ascorbic acid was detected by amperometric technique over Pt electrodes in which the method sensitivity was detected using UV and titrimetric methods [226]. However, the lack of studies related to the noble metals applications, is due to the high costs and the limited worldwide supply. Additionally, noble metal loses its high surface area that benefit it by irreversible agglomeration via Van der Waals interactions. Metals/metal oxides, bimetallic nanoparticles, noble metals nanoparticles based sensors for ascorbic acid detection were summarized in Table 3.

Table 3. Ascorbic acid electrochemical sensors based on transition metal nanoparticles and their composites

Sensor	**LOD (µM)**	**Sensitivity (µA mM^{-1} cm^{-2})**	**Ref.**
AgNP-Psi	0.83	1279	[227]
Hierarchical NiO/ITO	1.127	760	[228]
CdO/SPCE	0.0535	420	[229]
Branch-trunk Ag	0.06	35.512	[230]
RuO_2/Au	11.6	342.8	[231]
Au-Cys-Bt/GCE	0.87	30.0	[232]
$Cu(OH)_2$ nanorods/SPE	-	268	[233]
AuNP-SPCE	-	-	[234]
CL-TiN/GCE	1.52	6.073	[235]
Nano RuOx/Ni	-	296	[236]
Cobalt ferrite (CoFeGCPE)	0.15	-	[237]
AgNP/CPE	0.1	67.2	[238]
ZnO-Au/GCE	4.699	264.16	[208]
Mn-SnO_2 nanoparticles	0.058	10.92	[239]
hnp-PtTi alloy	24.2	436.31	[240]
CuO nanoneedle/SPE	88	107	[241]
CuO hollow sphere/SPE	90	533	[241]
Fe_2O_3/Au film	1	1281.9	[210]
Fe(III)-NClino/CPE	0.00024	-	[242]
CuO-SPE	88	107	[243]
BMITFB/Ni NP/CPE	0.04	0.158	[244]

Ag NP/CPE	600	-	[245]
Copper vanadate nanobelts	0.14	960.44	[246]
hnp-PtCu alloy	25.1	120.68	[247]
IrOx nanofibers	0.4	194.4	[248]
NiHCF/LDH/Au	21	55.73	[249]
MgO nanobelt/GCE	0.2	197.18	[250]
Chitosan/Ag-Au nanotubes	2	-	[251]
Au-Ru nanoshells	2.2	426	[207]
Gold-organosilica/CPE	1.54	78.63	[252]
$CuGeO_3$ nanowires	24	-	[253]
$Cu_4(OH)_6SO_4$ nanorods	6.4	17.53	[254]
Nano Au/TiO_2/Ti	-	-	[255]
Ni foil	-	169	[256]
Pt foil	-	178	[256]
Au-Pt alloy/ITO	1.0	-	[257]
Ni-Pt alloy	-	333	[209]
ultrathin Pd nanowire	0.2	166.5	[219]
Co film/GCE	0.2	1305.7	[211]
NiHCF/Au	25	-	[258]
βCD-nano Au/Fc-ITO	4.1	1764	[259]
Pt/Au/GCE	-	114.2	[260]
RuO_2/SPE	-	-	[261]

3.5. Other nanomaterials for ascorbic acid sensors

Many efforts have been done to develop high sensitive, cost-effective, and simple nanomaterial based sensors for the determination of ascorbic acid. Recently, the ascorbic acid was detected with excellent catalytic activity over a hexagonal boron nitride sensor with LOD=3.77 μM and linearity= 30-1000 μM [262]. The study showed that the boron nitride was synthesized via the combustion method at low temperature and carbothermal reduction tracked with nitridation. The senor additionally, was used to detect ascorbic acid in the presence of dopamine and uric acid without interference. In another study, the ascorbic acid was oxidized over a sensor of ketjen black encapsulated porous coordination network using DPV technique with linearity= $5.5X10^3$ - 0.2 μM [263]. Similarly, the ascorbic acid was detected in pharmaceutical samples and food samples over a modified GCE with Fe_3O_4NP, boron nitride, and 1-butyl-3-methyl imidazolium tetrafluoroborate IL [264]. Also, the ascorbic acid was detected over modified GCE with higher catalytic activity than unmodified GCE. Here, the modification process occurred by Bi_2S_3 nanoparticles modified titanate nanofibers to produce ascorbic acid sensor with sensitivity= 38 μA mM^{-1} cm^{-2} and linearity=10 mM [265]. The modified electrode showed lower current density indicating high electroanalytical performance than the unmodified electrode due to nanoparticles' high surface area. Additionally, the GCE modified with nanorods of bismuth sulfide synthesized via hydrothermal method was used for the electrochemical detection of ascorbic acid in phosphate buffer with LOD=0.083 μM, and linearity= 1-1000 μM [266]. The ascorbic acid was determined in 0.1 M KH_2PO_4 using CV over GCE modified with bismuth oxide nanoparticles. The modified electrode was also tested for real samples including fruit juices and tablets of vitamin C. [267].

4. CONCLUSION

The synthesis of different advanced nanomaterials such as metals, metal oxides, graphene, and CNTs are associated with nanotechnology applications advancements. Biocompatibility, excellent conductivity ranges, and high surface area of these prepared NMs allowed their use in different applications with high potency. One of these fields is the electrochemical sensors in which NMs were experienced with excellent results and high efficiencies such as the detection of biomolecules in real biological samples as serum and urine. Ascorbic acid electrochemical detection in real samples is considered to be a challenge for researchers due to its interference with dopamine. So, in the current review, the ascorbic acid sensors based on different nanomaterials such as carbon NMs, conducting polymers, noble metals, and metal/metal oxides NPs were discussed and their sensitivity was notified. The review showed that NMs can be considered as promising materials in the fabrication of biomolecules electrochemical sensors and approved its ability to solve many sensor problems such as fouling, reproducibility, selectivity, sensitivity, device failure in long-term stability, real-time continuous monitoring, and analysis of multiple targets. Thus, researchers can develop the electrochemical sensors using nanotechnology and overcoming the biofouling via the development of the electrode surfaces resistance, the enhancement of the reaction rate over the electrode surface by the addition of catalysts to the sensor materials, and the development of high surface/volume ratio nanomaterials and the use of these materials to enhance sensors performance through the modification of sensor geometry and substrate materials.

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