Review Article

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Contemporary Advances of Electrochemical Sensors in Forensic Applications

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Abstract

The present review article discusses the advances in electrochemical sensing for applications in forensic science to solve criminal conspiracy and cases. The basic principle involved in electrochemical sensing has been discussed along with their technical features and limitations. Furthermore, the significance of electrochemical sensing in the analysis of forensic samples i.e heavy metals, drugs, illicit compounds, biological weapons with suitable illustrations have been presented. The advances in electrochemical sensors have so far documented tremendous findings to assist forensic experts in correlating crime incidents with documents with some drawbacks. This linkage between the success and failure of electrochemical sensing in forensic sciences has tried to establish in interest in the safety and security of a society.

Keywords: Electrochemical Sensors; Forensic Science; Illicit Compounds; Sensing Parameters and Challenges.

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Introduction

Advances in sensing materials, techniques, and mechanisms have immensely improved the analytical science and its applications in different fields like water pollutions, biosensing, atmospheric prediction, and quality controls in various industries.[1] Although, the sensing is an old testing method but the recent advances i.e. portability, unprecedented wide range sensing parameters and stability in very harsh conditions have been made.[2–3] These features revealed its application in various fields like environment, clinics, healthcare, packaging, chemical engineering, automotive, space, and human comforts.[4–5] But, nowadays wide range sensing of different analytes like metals, organic molecules and inorganic molecules have also attracted the interests of forensic experts in analyzing different metals, drugs, liquors, medicine, narcotics, and explosive residue to solve the criminal cases and understand the history of a crime.[6–7]

In the above context, the current integration in size confinements to hybrid materials to dimensional optimization has provided unique features for designing electrochemical sensors to detect heavy metals, narcotics, alcohol in body fluid, and other forensic samples such as gunshot sites.[8] The area of sensing is widely studied, however, several limitations are still inviting the attention of scientists for its advance contemporary applications.[9] In light of the above developments, the present review article discusses the contribution of electrochemical sensors in forensic sciences with suitable illustrations along with the basic principle of electrochemical sensors. To the best of our knowledge. It is a unique focused review article on electrochemical sensing for forensic science.[this line we have already added.

Overview of the electrochemical sensor: The basic principle involved in electrochemical sensing is the monitoring of induced electrical responses develops after the interaction of analytes over sensing electrodes. Fig. 1 is concisely illustrating the fundamental principle of electrochemical sensors in brief.

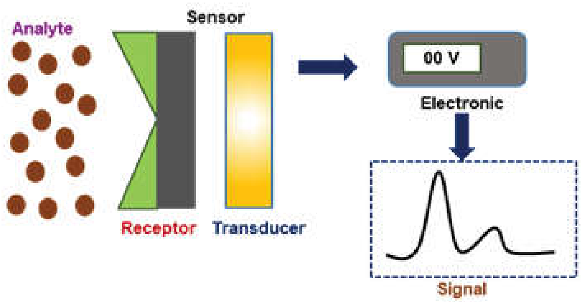

Fig. 1: The basic principle of electrochemical sensors.

The important induced electrical responses monitored in electrochemical sensing are resistance, conductance, and impedance, potential and induced current. These all induced electrical properties are the basis for classifications sensing purposes with their own and demerits. A comparative basic principle of electrochemical sensing is depicted in Table 1.

Table 1: Brief of different electrochemical sensing techniques

S.N.	Methods	Monitored properties	Unit
1	Potentiometry	Potential difference	Volts (V)
2	Conductometry	Resistance	Ohm (Ω)
3	Amperometery	Current as a function of potential	Ampere (I)
4	Coulometry	Current as a function of time	Coulombs (C)
5	Capacitance	Potential Load	Farads (F)

The other methods to classify the electrochemical sensors are interfacial methods and non-interfacial methods. In the interfacial method, the analyte directly interacts on the surface of the electrode and yield electrical responses as a sensing unit in a static (zero external disturbance) condition as well as dynamic, which exploits a redox reaction on the surface to electron transfer from analytes to the sensing electrode. However, in the non-interfacial method interaction took place in the bulk of electrode.

The composition, structure, morphology, and size of materials used in sensing electrodes are other important features to improve electrochemical sensing. In this regards several polymers, biopolymers, ceramic, metals and carbon compounds, are exploited with their inherited advantages and limitations. Currently, the blending of size reduction, polymer matrix, and inorganic catalytic molecules yield polymer nanocomposite, metal-organic framework, hydrogel, aerogel, xerogel for advanced electrochemical sensing in the efficient monitoring of forensically significant compounds like metals, narcotics, drugs, and explosives. In this regard, Shukla etal., has designed an integrated composite from chitin and polyaniline for efficient electrode materials.[10] The grafting chitin with polyaniline develops free interacting carbonyl groups present in the chitin matrix to interact with metal ions like copper. Thus, the interaction between cupric ion with chitin grafted polyaniline develops induced potential for efficient sensing of copper metals in natural (laboratory and groundwater) as well as artificial samples with negligible interference. The designed sensing setup and sensing parameters by authors are shown in Fig. 2.

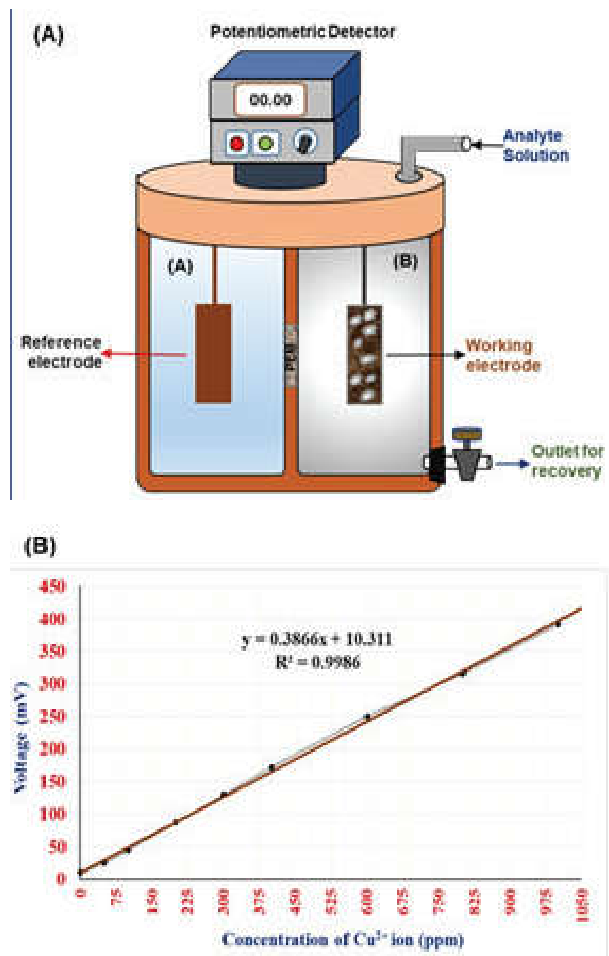

Fig. 2: Potentiometric sensing setup (A) and B is the sensing behaviour of cupric ion.[10]

Table 2: Representative electrochemical sensing materials along with properties.

S.N.	Composition	Objective	Sensing properties	Ref.
1	Nano materials	Use of nano materials	H_2O_2, H_2S and NO inside cells	11
2	Mesoporous Materials	Organic-inorganic hybrid mesostructures	Direct and electrochemical	12
3	Nanostructured metal oxide	Size confinement effects	Chemical, bio and gas sensor	13
4	ZnO nanostructure	Specific surface area and high catalytic efficiency	electrochemical sensors and biosensors	14
5	Metal oxides	Composite sensing and flexible/wearable	pH sensors	15
6	Metal Oxide	Size confinements	Heavy metals	16
7	Metal organic Frameworks	Chemically Stable	Wide range analytes	17
8	Conducting Polymer Nanostructures	Stable and functionalization	DNA, proteins, peptides, and other biological biomarkers	18
9	Conducting Polymers composite	Small dimensions; and large surface area	Chemical and bio sensor	19
10	Wide range	Wearable	Forensic sample and clinical samples	20

Similarly, several pristine and hybrids materials-based electrodes are exploited in electrochemical sensing using different transducers. Another modification in sensing materials is size confinements at the nano level and dimensional optimizations. The reduction in size improves the surface area and the dimension optimization channelizes the better response conduction in efficient electrochemical sensors. Some of the important reviews on electrochemical sensing materials are listed in Table 2 along with their composition, significant properties and sensing parameters.

Applications

The potential of electrochemical sensing has been extensively endorsed by the scientific community by precise chemical and compositional analysis of different forensic samplesqualitatively and quantitatively. Although the most scientific evidence to study as well as solve the crime is a compositional analysis of forensic residues like dead bodies, fingerprint soils of crime sites, and atmosphericgases. In this context, several techniques are already in practice to establish justice against crime but the development of sensing science has added several advantageous features in facilitating the criminal investigation to quick judgement and thus ensuring the law and order.[21]

Metal detection

The metals and its weapons are used in crimes since ancient times for various purposes but, with time their use has taken several destructive forms in doing criminal offenses as poison, weapon, arm, bombs, and ammunition. Recently the uncontrolled discharge of metals in water and edible items is also creating serious criminal cases towards the society.[22] Some of the poisonous metals are arsenic, antimony, barium, mercury, copper, lead, and thulium. In this regard, several electrochemical sensors are used to sense different heavy metals present in water and soil, collected from crime sites, and edible items with the help of several sensing materials and setups. Hence, the precise and accurate sensing of metals is always beneficial in solving and controlling crimes in society. Ott et al., has reported the use of electrochemical sensors for simultaneous monitoring of different metals i.e., lead, antimony and copper present in gunshot sites with a high population using square-wave anodic stripping voltammetry and screen-printed carbon electrode. The measurement is based on monitoring of potential shift in the redox potential of different metals i.e. lead ~0.784, antimony ~0.401 and copper ~0.282 v.[23] Similarly, the use of crime weapons like knives is having their composition which depends on the types and nature of their impact. In general, the knives used on homicide are consisting of iron, chromium, phosphorous, molybdenum, and vanadium. These weapons are creating serious problems in humans as well as leaving their own impacts on the surface and body. The presence of metal accumulated in the soft tissue of the human body reveals its chemistry to deteriorate the body functioning as well as metal toxicology. The main sources of metal poisoning are medication, the use of contaminated food and water, ingestion of herbicides, pesticides, fungicides and occupational exposures.

The presence of metals in water, edible items, and body fluids are extensively investigated by electrochemical sensors using various sensing materials. In the interfacial method, amperometry and potentiometry setups are used for sensing different heavy metals like lead, iron, copper, arsenic, chromium, and mercury. In general, these metals electrostatically interact on the charged electrode and develop induced potential or current in proportion to the concentration heavy metal ions present in testing fluid but sensing of simultaneous multiple metals are need of the time to improve the analytical chemistry and forensic science. In an innovative step, Durai et al., has reported, aluminium ferrite modified glass electrode for simultaneous monitoring of trace level heavy metals (cadmium, lead, copper and mercury (Hg2+) present in human blood serum using differential pulse anodic stripping voltammetry (DPASV) technique in the potential range of −1.2 to 0.4 V with excellent sensing parameters i.e limits of detection values 1.5 nM for Hg, 4 nM for Cd, 1.6 nM for Pb and 0.5 nM for Cu2+. The scheme of the sensing mechanism has been illustrated in Fig. 3 along with results.[24]

Some other important electrochemical sensors for heavy metal in human body fluid, water, edible item and soil collected from crime and gunshots are given in Table 3 with salient features.

Illicit organic compounds

Currently, the uses of organic compounds in criminal cases are exponentially increasing as narcotics, drugs, medicines, poison, and liquors. Although the narcotics and drugs are in practice since ancient times, but adulteration, controlled monitoring, bad practice, and the criminal mindset of society is using these compounds too much knowingly and unknowingly for crimes. Some time the narcotics are having medicinal importance but their uncontrolled consumption is fatal. Hence, their monitoring is important for solving several criminal

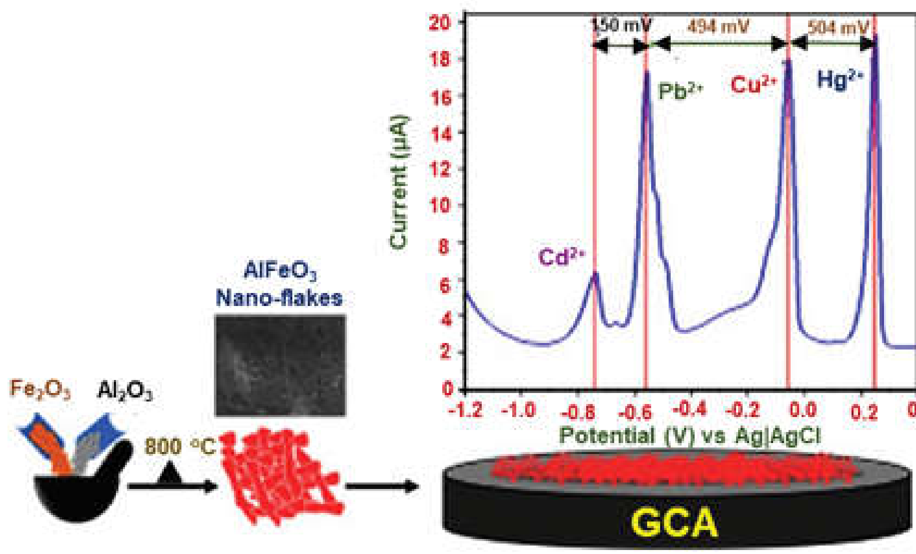

Fig. 3: Multiple metal sensing in human blood using electrochemical sensing[24].

Table 3: Electrochemical sensor for heavy metals.

S.N.	Composition	Transducer	Sensing metals	References
1	Graphene/PLA	Voltammetric	Simultaneous lead and antimony	25
2	Mercury-coated graphite	Anodic stripping voltammetry	Lead and antimony	26
3	Acetylcholinesterase (ACh) and acid phosphatase	Chronoamperometric	As (III)	27
4	Graphene oxide	Anodic stripping voltammetry	Zn in seminal fluid	28
5	Calix[4]pyrrole capped gold nanoparticles	Electrochemical sensing	As(III)	29
6	Ultrathin mercury film	Voltammetry	Barium	30
7	Nafion/ionic liquid/graphene	Anodic stripping voltammetry	Zn, Cd and Pb	31
8	Phthalocyanine	Amperometric	Lead	32

cases and crime related issues. Several sophisticated instruments are used for their monitoring with some limitations. Thus, the development of specific, reliable, and simple methods to detect illicit drugs in biological samples is the utmost requirement.[33] Gandhi et al., described an effective immunosensor for monitoring illicit drug abuse.[34] The finding has offered a low-cost detection of narcotics; thereby, providing a confirmatory platform to complement the existing analytical methods. Generally, electrochemical sensors monitor the current or voltage generated by oxidation or reduction of these illicit drugs. Shukla et al., has monitored residual pesticides i.e malathion and drug paracetamol in water samples after monitoring the induced potential on a suitable electrode. The potential was developed in the case of malathion after surface interaction between sulphur and polyaniline, due to oxidation of paracetamol by iron present iron oxide encapsulated in chitosan-grafted polyaniline hybrid matrix.[35-36] The scheme of potentiometric sensing of paracetamol in water is shown in Fig. 4.

Some other important electrochemical sensing of illicit organic compounds are given in Table 4 along with transducer and properties.

Applications in biological weapons

Bioterrorism is another most fatal kind of abuse in human society and has currently been used in different ways. Biological warfare includes germs like bacteria, viruses, insects, and fungi, which are more potent than conventional and chemical weapons. Furthermore, the current progress in biochemistry, biotechnology, and genetic engineering has made science simpler in developing biological warfare with more fatal characteristics. Lately, these weapons have become more sophisticated in use with severe impacts on

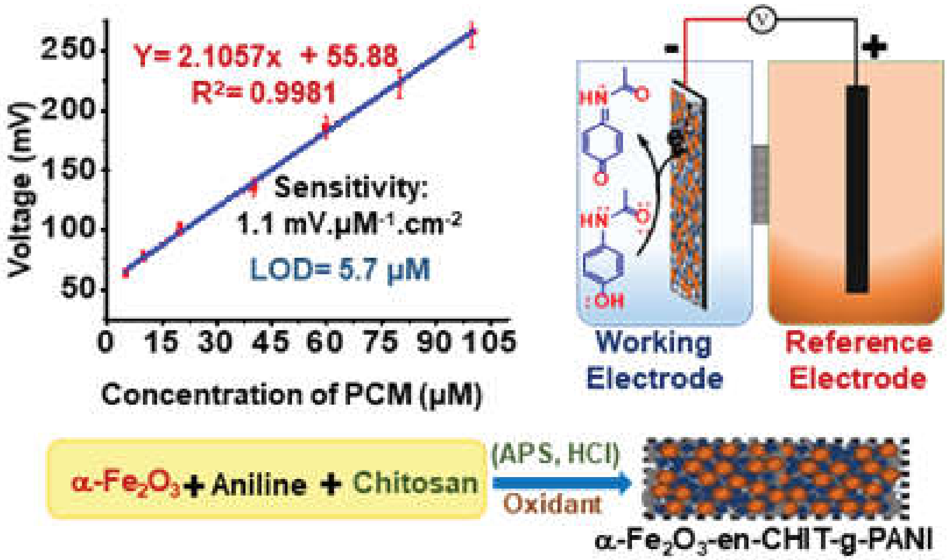

Fig 4: Potentiometric sensing of paracetamol over iron oxide encapsulated in chitosan-grafted-polyaniline.[36]

Table 4: Electrochemical sensing of illicit organic compounds.

S.N.	Composition	Transducer	Sensing compound and properties	References
1	Carbon nanotube β-cyclodextrin	Linear sweep voltammetry	Cocaine with detection limit of 1.02 μM	37
2	Carbon electrodes/ gold/graphene oxide	Voltammetry	Carbofuran in sensing range linear range of 1–250 μM	38
3	Graphene-Au	Differential pulse voltammetry	Carbaryl and detection limit of 0.0012 μM	39
4	Schiff base complex	Linear sweep voltammetry	Cocaine hydrochloride	40
5	Gold electrode	Voltametry	Methanol and ethanol	41
6	Pd impregnated graphene	Cyclic voltammetry	Cocaine	42
7	Catechol-Attached Polypeptide	Differential pulse voltammetry	Cannabinoids	43
8	Au/Fe_2O_3/MWCNT	Voltammetry	Methamphetamine	44

the health and life of organisms. Therefore, the monitoring of these weapons is highly important and many electrochemical based sensors are being used to detect the biological warfare substances used in crimes for individuals or in groups.[45] Some of the used biological weapons are microbes i.e., bacteria, virus and toxic chemical substances. The plant-based toxic substances are also used which are categorized into alkaloids, glycosides, tannins, proteins, oxalic acid and oxalates and volatile oils. For example, mushroom poisoning is caused by wild varieties of mushroom-like Amanita group members which produce amatoxins and muscimol that would affect the health of human beings. The phytochemicals adverselyaffect human health like white color latex of the plant, i.e. Calotropisprocera acts as a poison if taken inthe excessive amount.[46] In this context, several electrochemical sensors have been developed to detect biological warfare including algal toxins such as Cylindrospermopsin, Anatoxin-a, Brevetoxins, and cyanotoxins in water.[47] Recently, a DNA based biosensor has developed to detect the fungal toxin (mycotoxins) i.e. Ochratoxin A from plant-based food and beverages.[48] Thus, developing the biosensors especially to detect some plant-based chemical substances, which are poisonous to human beings or other organisms, isimmensely important in the field of drug analysis and forensic toxicology. Electrochemical sensors are also used for sensing themicrobes after monitoring induced potential and current due to electrochemical interactions. For example, the microbial activity induces the impedance of the system and has been used for monitoring the temperature-dependent activeness of microbes.[49] A simple electro sensing strategy has been reported by Joshi to monitor the virus in saliva through change in impedance over reduced graphene. The sensors exhibit high stability and reproducibility due to the high adhesion of sensing materials due to the presence of phenolic (-OH) moiety with the limits of detection of 26 and 33 plaque-forming units.[50]

Conclusion

The brief principles and applications of electrochemical sensing in forensic sciences have been discussed for three types of illicit materials i.e., heavy metals, organic compounds, and biological warfares. The sensing mechanism and role of sensing materials have been discussed with suitable illustration. Furthermore, the sensing properties with an emphasis on highlighting the importance for the scientists to amplify research and applications have also been discussed.

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