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Green synthesis of curcumin-silver nanoparticle and its modified electrode assisted amperometric sensor for the determination of paracetamol

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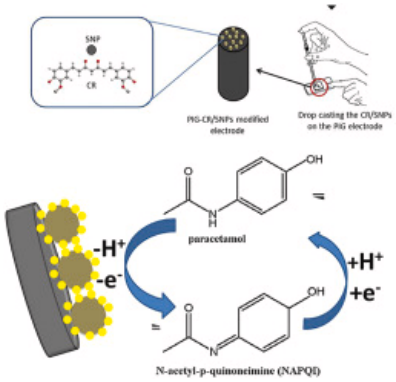
Abstract

Contamination of paracetamol, a primary analgesic was wide spread in the water system that affects the eco-system. High-dosage of paracetamol to humans cause organ damages and showed adverse effect. It is important to monitor the paracetamol concentration in environmental and human samples periodically. Conventional methods associated with chromatography is found to be high-cost, time consuming and requires high-end instrumentation, Herein, we investigated the role of curcumin during bio-synthesis of silver nanoparticles. The curcumin functionalized silver nanoparticles were further chemically modifying on the electrode surface and the resulting modified electrode was applied for electrocatalytic oxidation of paracetamol. The experimental finding proved that the modified electrode is capable of sensing paracetamol by applying oxidation potential 0.4V. Both the synthesised material and modified electrode surface were characterized for its physic-chemical properties using spectroscopy and microscopy techniques. The HR-TEM, FESEM and AFM results showed that the distribution of nanoparticle with the size range from 25 to 70nm and the UV–Vis and Raman spectrophotometer characterization confirms the coordination between SNP and curcumin. Under optimized condition, in 0.1M NH_4Cl (pH 7) at the scan rate of 50mVs^{-1}. The modified electrode enhanced the sensitivity towards the detection of paracetamol in trace level. The modified electrode is capable of sensing paracetamol in a linear range between 0.59×10^{-6} and 342.1×10^{-6}M, with LOD of 0.29μM, and linear regression equation of y=0.092x+502.6 with a correlation coefficient of R^2=0.996.

Graphical abstract



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Introduction

Water contamination is wide spread in the urban and rural area. Due to industrialisation, increase in domestic sewage, and hospital waste, there are several active pharmaceutical pollutant quantified in waste water system. This affects the environment and ecosystem adversely. Therefore it is important to develop a technology which can quantify, separate and degrade the pollutant from water system. Development of miniaturized, cost-effective devices would be feasible for industry and hospital to monitor the discharge of pharmaceutical pollutant in their effluent. This will help for the pharma pollution control and waste water treatment (Sivaranjanee et al., 2022). The active pharmaceutical compound paracetamol (PAR) chemically named as acetaminophen is extensively used over long period of time and considered as the primary analgesic for chronic pain management. Several international agencies have recommended the use of PAR. It is very effective, well tolerated and suitable for all varieties of population. It is effective for both acute and chronic pains like head ache, migrane, soft tissue injury, pain after dental injury and as an anit-pyretic agent. It is also used in combination with opioid analgesia for the management for severe pains as in the case of cancer pain (P, Michael Yelland *et al.*, 2005). PAR is thought to be selectively functioning by stopping prostaglandin release and it also contributes some peripheral analgesic effect. But still the exact mechanism of action has to be confirmed (P, Michael Yelland *et al.*, 2005). Although PAR can be considered as an effective pain reliever, over doses of the same may lead to hepatoxicity due to hepatocyte apoptosis which has been proved in experimental animals (Lorz et al., 2004; Hale Z. Toklu et al., 2006) and sometimes found to be fatal (Hale Z. Toklu et al., 2006). Nephrotocity have also been seen in animals subjected to high doses of PAR due to the induction of apoptosis in the renal tubular epithelial cells (Milena Ściskalska et al., 2014) (Lorz et al., 2004). Apart from heaptotoxicity and nephrotoxicity PAR overdose can induce skin rashes and pancreas inflammation (Sudip Biswas *et al.*, 2015). Deliberate or accidental PAR overdose can be detected and treated with N-acetylcysteine which is the antidote for PAR overdose.Plasma concentration of PAR is measured in patients for the treatment who has been poisoned deliberately and ingested concentration of the drug is measured in case of accidental overdose (Paul I Dargan et al., 2002). Since the amount of drug overdose and the duration plays an important factor in the treatment, it is essential to detect the concentration of the drug rapidly and selectively.

Many methods have been used towards selective determination of PAR such as liquid chromatography, spectrophotometry, etc., but they are very expensive, time consuming and lack sensitivity and selectivity (Ayyadurai Kannan et al., 2017; Sudip Biswas *et al.*, 2015). The electrochemical oxidation of PAR is mainly due to the phenolic hydroxyl group present in it (Jinchun Songa *et al.*, 2011). As PAR is an electrochemically active species, it can be detected easily by electrochemical analysis rapidly with high accuracy in trace quantities (Ayyadurai Kannan et al., 2017; Sudip Biswas *et al.*, 2015). The metabolic products of PAR often co-exist along with ascorbic acid and dopamine which exert physiologic importance (Xianlan Chen *et al.*, 2016, Sudip Biswas *et al.*, 2015). Electrodes modified with Poly-4-amino-6-hydroxy-2-mercaptopyrimidine (Poly-AHMP) (Ayyadurai Kannan et al., 2017) porous nitrogen doped graphitic carbon (Sudip Biswas *et al.*, 2015) and Au/ZnO/N-doped graphene nanocomposite (Xianlan Chen *et al.*, 2016) detection of PAR with co-existing species like ascorbic acid and dopamine selectively with high sensivity (Uday Pratap Azad et al., 2015). This infers that electrochemical detection of PAR using modified electrodes is highly desirable over other conventional methods.

Nanomaterial has contributed towards the development of electrochemical sensor for nanoscale detection (Mary Isabell Sonali et al., 2022; Padmalaya et al.,, Padmalaya et al., 2022). Among them colloidal silver nanoparticles have attracted much attention in electrochemical sensor development due to their ease of synthesis, small size, high surface area, electrically conductive and redox mediating property (Rameshkumar et al., 2014). Silver nanoparticles (SNPs) act as an electrically conductive substrate for preparing chemically modified electrodes (Li Lin et al., 2008). SNPs are inexpensive when compared to other nanomaterials and have good physical and chemical properties. SNP coated surfaces are capable of improving the electrochemical and optical properties of the substrate (Xing et al., 2011; Amiri et al., 2015). They have superior properties like high catalytic performance, good capacitance, high conducting nature, and surface enhanced Raman scattering properties (Wang et al., 2009). SNPs are capable of acting as catalysts for oxygen reduction. A SNP coated glassy carbon electrode (GCE) has been found to be effective in the determination of dissolved oxygen (Tsai et al., 2010). They have good affinity towards heme-protein and have been employed for the electro-analysis of cytochrome *c* protein. When compared to bulk silver electrode, SNP functionalized electrode exhibited enhanced electron transfer between cytochrome *c* and the electrode (Lin *et al.*, 2008). Colloidal SNPs are capable of interacting with L-cysteine molecules *via* the amino and carboxyl groups. Self-assembled monolayer of L-cysteine decorated SNPs were used to modify gold electrode and used for the electrocatalysis of neutral red dye (Wang et al., 2009). A SNP/Nafion (cation exchange polymer) film modified GCE was employed successfully for the determination of Chromium (VI) in real sample. The modified electrode was found to be highly sensitive and interference by other heavy metals such as cadmium, copper or mercury was effectively eliminated (Xing et al., 2011). SNP modified GCE have been used for the determination of mercury in drinking water in trace quantities (Suherman et al., 2017). Chitosan stabilized SNP modified GCE was found to be effective in the determination of pesticides such as pendimethalin (in tap and mineral water) and ethyl parathion (in lettuce and honey). Due to the electrocatalytic activity of SNPs, the modified electrode was capable of detecting nitrogen containing compounds more effectively than bare electrode (Alves de Lima et al., 2016; Pal and Ganesan, 2010, Shivakumar et al., 2017).

Bio-treatment of waste water system using microalgae consider to be cost-effective and the role of microorganism is multifunctional (Chai et al., 2021). Recent research focused on bio-synthesis of nanomaterials using biomolecules or microorganism (Rambabu, K,2020) as functionalizing, reducing agent and its application towards electrochemical sensor development and waste water treatment is of emerging interest (Chia et al., 2018). To overcome the demerits associated with conventional technique and consider to be an eco-friendly approach there is a need for development of electrochemical sensor (Parkavi et al., 2022; Subhasri et al., 2022). The role of functionalized SNPs is wide spread in electrochemical sensor application. Also the electrocatalytic behaviour of curcumin has equally contributed for the electrochemical sensor development. Several chemically modified electrode using curcumin in the form of Ni (II)-curcumin complex (Aleksander Ciszewski, 1995; M. Yousef Elahi et al., 2007; S. Majdi et al., 2007*;* Sasikumar Ragu et al., 2016), polycurcumin (Balamurugan Devadas et al., 2014), curcumin-carbon composite (Zheng and Song, 2009), and curcumin functionalized gold nanoparticles (K. Krishna Kumar et al., 2019) were developed and used for sensing various organic and in-organic compounds. Curcumin has the tendency to form complex with metal ions, also it has the property to reduce the bulk metals into stable metal nanoparticles without using any toxic reducing agents. As a green chemistry approach curcumin was used as a reducing agent and functionalizing agent during synthesis of SNPs. The present work focused on developing chemically modified electrode using CR-SNPs nanomaterials. The physico-chemical properties of CR-SNPs nanomaterial and CR-SNPs modified electrode were studied using spectroscopic, microscopic and electrochemical techniques. The CR-SNPs modified electrode was used for electrochemical determination of paracetamol using amperometic technique. So far there is no literature reporting the application of green synthesised curcumin functionalized silver nanoparticles for the electrochemical sensing of paracetamol. The physio-chemical characterization results showed that the green synthesised CR-SNP is in nano-size with large surface area and enhanced electrocatalytic property. As such SNP alone is not stable in solution or solid surface due to photocatalytic oxidation of SNP. But upon functionalizing with CR, the stability of the CR-SNP is found to highly stable. Herein, for the first time we are reporting the application of CR-SNP modified electrode towards determination of paracetamol. The CR-SNP modified electrode exhibited excellent electrochemical behaviour towards electrocatalytic oxidation of paracetamol which showed stable behaviour for multiple analysis. The CR-SNP mediated the oxidation process of paracetamol by enhancing the peak current and favours for its sensitive detection. The optimized methods can be taken for real time analysis of paracetamol in water system and industrial effluent from neighbouring pharmaceutical industries.

Section snippets

Chemicals and reagents

Spectroscopic grade graphite rod purchased from Sigma-Aldrich was used as a working electrode. Paracetamol was purchased from Sigma-Aldrich. All other chemical used in the study was procured from SRL Chemicals, India.…

Instrumentation

The material and modified electrode surface characterization were performed using spectrophotometric and microscopy analysis available at Central Instrumentation Laboratory, Vels institute of science, technology and advanced studies and National centre for nanoscience and…

UV–visible spectrophotometer of CR, SNP and CR-SNPs nanomaterial

The optical properties of curcumin (CR), citrate capped silver nanoparticles (cSNPs) and curcumin functionalized silver nanoparticles (CR-SNPs) were measured using UV–visible spectrophotometer. Fig. 1 (i) showed the absorption spectrum of **(a)** CR, **(b)** cSNPs, and **(c)** CR-SNPs. For the absorption spectra of CR (graph a), a highly intense peak at λ_{max} 425 nm was observed **(Kim HJ *et al.*, 2013 & Kim YJ *et al.*, 2013).** The absorption maxima (λ_{max}) for cSNP (graph b) was found at 420 nm which was due to…

Electrocatalytic oxidation of PAR using CR-SNPs electrode

For a known concentration of PAR in 0.1 M NH_4Cl (pH 7) solution, the CV measurement was recorded using SNP electrode and CR-SNPs electrode. Fig. 8 (i) shows the CV **curve a** for SNP electrode and **curve b** for CR-SNPs electrode in 0.1 M NH_4Cl (pH 7) solution containing 78.06×10^{-6}M of PAR. The potential of the electrodes were scanned between −0.4V and +1.0V at 50 mVs^{-1}. The material design for modifying the electrode surface and its sensing behaviour are based on its redox behaviour **(Athimotlu** …

Conclusion

As a green chemistry approach, a novel electrochemical sensor was developed using curcumin functionalized silver nanoparticles modified electrode. Synthesis of silver nanoparticle using curcumin added advantage and consider as eco-friendly approach in development of non-toxic sensor system. The combination of metal (silver) nanoparticle with curcumin in polymeric form is a new class of metallopolymer nanoparticles**.** The electrochemical performance of the CR-SNP towards electrocatalytic oxidation …

Credit author statement

K. Krishna Kumar: Investigation; Data curation; Resources; Writing – original draft; M. Devendiran: Data curation; Formal analysis; Resources; P. Senthil Kumar: Conceptualization; Methodology; Validation; Supervision; R. Suresh Babu: Data curation; Formal analysis; Resources; S. Sriman Narayanan: Conceptualization; Methodology; Validation; Supervision…

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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