



Studies on L-histidine capped Ag and Au nanoparticles for dopamine detection



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ARTICLE INFO

Article history:

Received 25 June 2016

Received in revised form 13 September 2016

Accepted 24 November 2016

Available online 26 November 2016

Keywords:

L-Histidine

Dopamine sensing

Ag

Au

Bimetallic nanoparticles

Biosensor

ABSTRACT

This work demonstrates the effective surface functionalization of Ag, Au and bimetallic Ag–Au nanoparticles using L-histidine for colorimetric detection of dopamine (DA) which plays majorly in recognizing the neurological disorder. L-Histidine (L-His) capped Ag, Au, and bimetallic Ag–Au nanoparticles are characterized using physico-chemical techniques. The optical behaviour of nanoparticles has been analysed at various time intervals using UV–Vis absorption spectroscopy. FT-IR results provide the evidence of chemical bonding between L-histidine and metal nanoparticles. Its structure with the capping of L-His was clearly shown in HR-TEM images. The average size of nanoparticles has calculated from TEM image fringes are 11 nm, 5 nm and 6.5 nm respectively, matches with crystals size calculated from X-ray diffraction pattern. Enhanced optical nature of nanoparticles provides the best platform to develop a colorimetric-based biosensor for DA detection. After addition of DA, a rapid colour change has been noted in colloids of nanoparticles. The substantial changes in absorbance and λ_{\max} in metal nanoparticles respect to DA concentration have been observed and formulated. This is one of the successive methods for trace level determination of DA and will be going to a significant material for designing biosensor to determine DA in real extracellular body fluids.

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1. Introduction

Recently metal nanoparticles became more eye-catching for their excellence in detection and imaging purpose [1]. Currently, many types of research on ligand-protected metal nanoparticles are being achieved and investigated. The interesting electro-optical behaviourism of interconnected biomolecules and metallic nanoparticles bring more significance in developing the biosensing materials [2–7]. Among various metal nanoparticles, Au and Ag attract many researchers interest owing to their properties like optical chirality, quantized charging behaviour and photoluminescence along with a wide range of application in the field of sensing, catalysis and labelling [8–19].

Among various amino acids L-histidine (L-His) is a one with aromatic nitrogen and imidazole side chain. Due to its extreme biochemical importance, it is used in various biological process and medical

applications [20]. The three main sites in L-His, which participated in metal adsorption are the carboxylic group, imidazole nitrogen atom, and amino group [21]. Owing to the presence of these sites, L-His acts as one of the strongest metal ion binders. Many investigations have been reported to explore the interaction of L-His with metals and metal oxides like Cd, Fe, Zn, Co, ZnO, Fe₃O₄ [22–26].

Dopamine (DA) is a catecholamine neurotransmitter, which plays a crucial role as chemical messengers in central nervous system, hormonal, cardiovascular and renal systems. Recently, many attentions are paid to trace level determination of DA, because small changes in concentration coupled with various disorders like Alzheimer's disease, epilepsy, Parkinson's disease, schizophrenia, and pheochromocytoma etc. The selective and sensitive approach for DA level determination is vital for diagnosing diseases. It has been shown that metal nanoparticles having better sensing activity [27–29]. Metal nanoparticles like magnetic Fe₃O₄ with Au nanoparticles have been utilized for DA sensing and detection [30].

In this present work, the detection of DA using L-His capped metal (Ag, Au) nanoparticles has been studied by analysing the SPR peak shift after addition of DA. The optical properties, crystalline pattern and the morphological structure of L-His capped metal nanoparticles (Ag, Au, Ag–Au) have been investigated. And the comparative DA sensing behaviours of synthesised metal nanoparticles has been studied at

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Table 1
Synthesis of nanoparticles with various parameters.

| Sample name | Nanoparticles | L-His solution | Ag ⁺ solution | Au ³⁺ solution | NaOH solution | Colour change |
|-------------|--------------------|----------------|--------------------------|---------------------------|---------------|---------------|
| i | L-His capped Ag | 30 mL | 500 μ L | — | 10 mL | Yellow |
| ii | L-His capped Au | 30 mL | — | 500 μ L | 10 mL | Pink |
| iii | L-His capped Ag-Au | 30 mL | 500 μ L | 500 μ L | 10 mL | Orange |

the different concentration of DA. The relation between the wavelength of maximum absorbance (λ_{max}), absorbance, and DA concentration are formulated based on sensing property. So for many electrodes and colorimetry based DA sensor has been developed [31]. To our knowledge, it is a simple, easy and handy method for detection of DA concentration using biomolecules assisted bi-metallic nanoparticles.

2. Experimental procedure

2.1. Materials

Aurochloric acid (HAuCl_4) and silver nitrate (AgNO_3) were purchased from Sigma-Aldrich, India and used as initial precursors for synthesizing L-His capped Ag, Au, and Ag-Au nanoparticles. L-Histidine ($\text{C}_6\text{H}_9\text{N}_3\text{O}_2$) and sodium hydroxide (NaOH) were obtained from Merck Specialities Private Limited, India. All chemical reagents of analytical grade are used as received without any further purification.

2.2. Synthesis of L-histidine capped nanoparticles

L-His solution has been prepared by dissolving 10 mM L-His in 100 mL double distilled water (DD). By dissolving 30 mM of AgNO_3 and HAuCl_4 in 2 mL DD respectively. Like the same NaOH solution has been prepared by dissolving 25 mM of NaOH in 50 mL DD. Finally, the preparation of three different nanoparticles has shown in Table 1.

These three different samples are maintained at 60 °C, and the reaction has been terminated after attaining the respective colour changes as noted in Table 1.

2.3. Procedure for dopamine sensing

Different concentrations of DA have been prepared by dissolving 10 μ M, 15 μ M, 20 μ M and 25 μ M in 100 μ L of DD respectively and are tested against L-His based Ag, Au and Ag-Au nanoparticles. Real-time colour change has been noted from yellow to dark yellow for Sample i. Similarly, visible colour change from pink to violet and orange to red respectively occurs for Sample ii and Sample iii after addition of DA at different concentration.

2.4. Characterization

Optical absorption spectra have been recorded using a JASCO V-650 spectrophotometer in the UV-Visible wavelength range of 200 nm–900 nm and images of respective colour changes during reactions has been photographed with a Canon digital camera. The surface morphology has been studied using Atomic force microscope (AFM Park System, 100XE) at different magnifications range with silicon nitride cantilever tip height 10–12 nm in the contact mode. TEM samples are prepared by dropping solution onto a carbon coated copper grid (3 mm, 200 mesh), and images have been captured with Tecnai (FEI) high-resolution transmission electron microscope

(HR-TEM). The crystallinity and phase changes have been obtained by Rigaku Ultima III X-ray diffraction (XRD) with Cu-K α radiation (1.500 Å, 40 kV, 30 mA). The fingerprints of functional group bindings are recorded in the spectral range of 400 cm^{-1} –4000 cm^{-1} using a PerkinElmer FTIR spectrophotometer.

3. Results and discussion

3.1. Spectral characteristics

UV-Vis spectra for different nanoparticles are measured at different time intervals during the experimental procedure are shown in Fig. 1. The surface plasmon resonance (SPR) peak of L-His capped Ag (Fig. 1(A)) nanoparticles and L-His capped Au (Fig. 1(B)) are at 415 nm and 539 nm respectively. Whereas for L-His capped Ag-Au nanoparticles (Fig. 1(C)), the broad SPR peak exhibits in the range of 430–540 nm confirm the presence of both Ag and Au nanoparticles. Similarly, there is an existence of L-His peak in all three synthesised nanoparticles at 282 nm, 283 nm, 279 nm, respectively shows the capping behaviour of L-His above the nanoparticles. This proves the L-His acts as both capping and reducing agent. As time increases, nucleation starts and further growth of nanoparticles taken place which is directly related to the absorption as well as yield. After attaining the growth, absorption gets saturated with the maximum yield at 60 min, 120 min and 60 min for Ag, Au, and Ag-Au nanoparticle respectively shown in Fig. S1 (see Supplementary). This show the ions get reduced completely and further capping agent prevents the aggregation of respective nanoparticles.

The available chemical bonds and its bindings between L-His and metal nanoparticles have been studied using Fourier transform infrared spectroscopy. In the spectrum of L-His Fig. 2(A), the absorption position at 675–750 cm^{-1} , 1124 cm^{-1} , 1404 cm^{-1} , 1490 cm^{-1} , 1640 cm^{-1} , 2080 cm^{-1} , 3200–3500 cm^{-1} corresponds to bending vibration (δ) of sp^2 CH group, stretching vibration (ν) of CN in imidazole group, symmetric stretching (ν_s) mode of COO^- group, symmetric NH bend, $\nu(\text{C}=\text{C})$, $\nu(\text{N}-\text{H})$, and $\nu(\text{O}-\text{H})$ of H_2O , respectively [32–34]. In Fig. 2(B), the presence of COO^- group at 1404 cm^{-1} in L-His has been shifted slightly to 1385 cm^{-1} , 1384 cm^{-1} , and 1388 cm^{-1} in L-His capped Ag, Au, Ag-Au nanoparticles respectively, indicates the interaction of carboxylic group of one L-His molecule with amine group presents in neighbour L-His molecule. Whereas the vibrations in the spectral position 1124 cm^{-1} and 1490 cm^{-1} in L-His are absent in metal nanoparticle spectra, proves the adsorption of L-His on the metal surface through amine group and nitrogen atom present in the imidazole group. This result indicates the binding as well as the capping of L-His over metal nanoparticles.

3.2. XRD characteristics

XRD pattern of L-His capped Ag, Au, Ag-Au nanoparticles has shown in the Fig. 3. This graph shows diffraction pattern of L-His capped Ag nanoparticles having a face-centered cubic lattice matches with JCPDS No: 01-087-0597. Similarly, the XRD pattern of L-His capped Au nanoparticle matches with the JCPDS No: 00-002-1095 and having a cubic system of the face-centered lattice. Whereas in the diffraction pattern of L-His capped Ag-Au nanoparticles, some peaks marked with (#) are matches with the JCPDS No: 00-041-1402 corresponds to the Ag, hexagonal systems having primitive lattice. While some others are matches with the JCPDS No: 00-004-0784, which corresponds to the Au, cubic systems having a face-centered lattice. The mean crystal size has calculated using Debye-Scherrer's formulae $D = 0.9\lambda/\beta\cos\theta$ where λ is the used X-ray wavelength and β is full width at half maximum (FWHM).

Fig. 1. UV-Vis absorption spectra recorded at different reaction intervals (A) L-His capped Ag nanoparticles, (B) L-His capped Au nanoparticles, and (C) L-His capped Ag-Au nanoparticles.

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