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Modified screen-printed Electrode as Electrochemical Detector for Noscapine

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In the present study, a La^{3+}doped Co_3O_4 nanoflowers modified graphite screen-printed electrode (La^{3+}doped Co_3O_4 nanoflower/SPE) were prepared and used for voltammetric determination of noscapine. Presence nanoflowers lead to a substantial improvement in current in compare to an unmodified electrode. The diagnostic methods used for the purposes of the study include chronoamperometry, differential pulse voltammetry (DPV) and cyclic voltammetry (CV) approaches. The modified SPE was used to determine noscapine in the range of 0.3 to 600.0 µM (LOD= 0.1 µM).

Keywords: Screen printed electrode, Noscapine, Modified electrode, La^{3+}/Co_3O_4 nanoflower, Voltammetry,

1. INTRODUCTION

Noscapine[(3S)-6,7-dimethoxy-3-[(5R)-4-methoxy-6-methyl-7,8-dihydro-5H-[1,3]dioxolo[4,5-g]isoquinolin-5-yl]-3H-2-benzofuran-1-one] is a phthalideisoquinoline non-narcotic alkaloid. It is derived from the poppy family through potential antineoplastic, antitussive and mild analgesic activities. This factor mostly is used for its antitussive (cough-suppressing) effects. Its major pharmaceutical action, antitussive activity, is reported to be equal to that of codeine. According to the research, noscapine can cause apoptosis in different kinds of cells and it has a powerful antitumor effects on human breast, solid murine lymphoid tumors, and bladder tumors implanted in nude mice. Noscapine presents in opium in concentrations of 2–8% and is the second most alkaloid in it.A wide range of Noscapine concentration is reported in illicit heroin samples: from none up to 46% of the sample weight [1-7].

There are different methods to determine and detect noscapine in plasma, urine, and opium samples containing high performance liquid chromatography (HPLC) [8-10], flow-injection chemilumiescence [11], capillary electrophoresis [12-15] and LC-MS [16-18]. These techniques have different disadvantages such as tedious sample pretreatment steps, time-consuming nature, and expensive operations. However, electrochemical detection techniques are superior to others [19-37].

The screen-printing is a low cost, small size and light weight technology that widely been used for analytical applications and has made possible mass production of electrodes and the development of practical ‘‘in situ’’ analysis [38-46]. In particular, graphite-based screen-printed electrodes (GSPEs), because of low background current, broad potential windows of application and resistance to a wide range of solvents, have been highly suitable to be customized with diverse (Nano) materials [47-51]. Therefore, the screen printed electrode surface can easily be modified. Chemically modified electrodes have a conductive substrate modified with monolayers, electroactive thin films, or thick coatings. The modified electrodes are applicable for certain purposes that a bare conductive electrode does not allow them. Modified conductive substrates allow enhancement of electron transfer kinetics. Meanwhile, surface modifications act as a catalyst, so that trivial changes in surface characteristics affect sensitivity of measurement in electroanalytical applications [52-60].

In this regard, the modified electrodes have been widely used for sensitive and selective determination of the various targets [61-76].

In the recent years, production of noble metal-doped metal oxides,novel nano-scale metal oxides, metal oxide-polymer composites, and metal oxide-CNTs nanocomposites has attracted substantial attention. Novel analytical devices take advantage of nanostructured metal oxides. Given the large surface-to-volume ratio of the nanostructure, these devices are cost-effective and highly sensitive, and also exhibit excellent selectivity when assembled to metal ions with simple design [77-91]. Complex metal oxide with spinel structure are very promising as electro catalysts. However, because of being inexpensive, active, and thermodynamically stable they have some problems in achieving low resistivity and high surface area. Cobalt oxide-based materials can be used in energy storage system, magnetoresistive devices,electrochromic thin films, and heterogeneous catalysis. In addition, Co_3O_4 and other cobalt-based oxides have excellent electro catalytic activity toward various compounds, oxygen evolution. Therefore, they continue to attract significant concern. The electro

catalytic nature of the cobalt oxide film can be attributed to the deposition method. Numerous methods such as, thermal salt decomposition, plasma sputtering, sol–gel and powder immobilization technique so far have been to that end [92-96].

In the present paper the synthesis and characterization of La^{3+}/Co_3O_4 nanoflowers followed by its immobilization on the surface of a screen printed electrode to extend a voltammetric sensor introduced for the determination of the noscapine. No study has been reported for this sensor before.

2. EXPERIMENTAL

2.1. Apparatus and chemicals

An Autolabp otentiostat/galvanostat was applied for electrochemical detections. The screen-printed electrode (DropSens, DRP-110, Spain) includes 3 main sections that contain a silver pseudo-reference electrode, a graphite working electrode, and graphite counter electrode. All reagent were purchased from Merck Company.

2.2. Electrode Preparation

As seen in below, La^{3+}/Co_3O_4 nanoflowerswas applied to modify the SPE. Dispersion of 1 mg of La^{3+}/Co_3O_4 nanoflowers with ultrasonication for 1h was used for preparing a stock solution in 1 mL of the water, whereas 2 µl of it was droped on the working electrodes. The solvent was evaporated in room temperature.

3. RESULTS AND DISCUSSION

3.1. Electrochemical Profile of the Noscapine on the La^{3+}/Co_3O_4/SPE

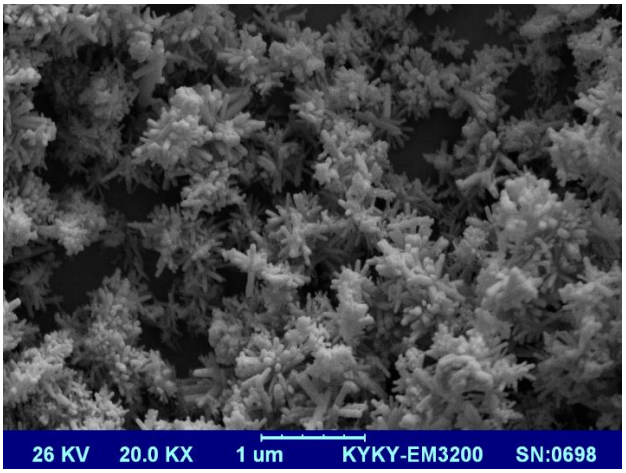

Figure 1. SEM image of La^{3+}/Co_3O_4nanoflowers.

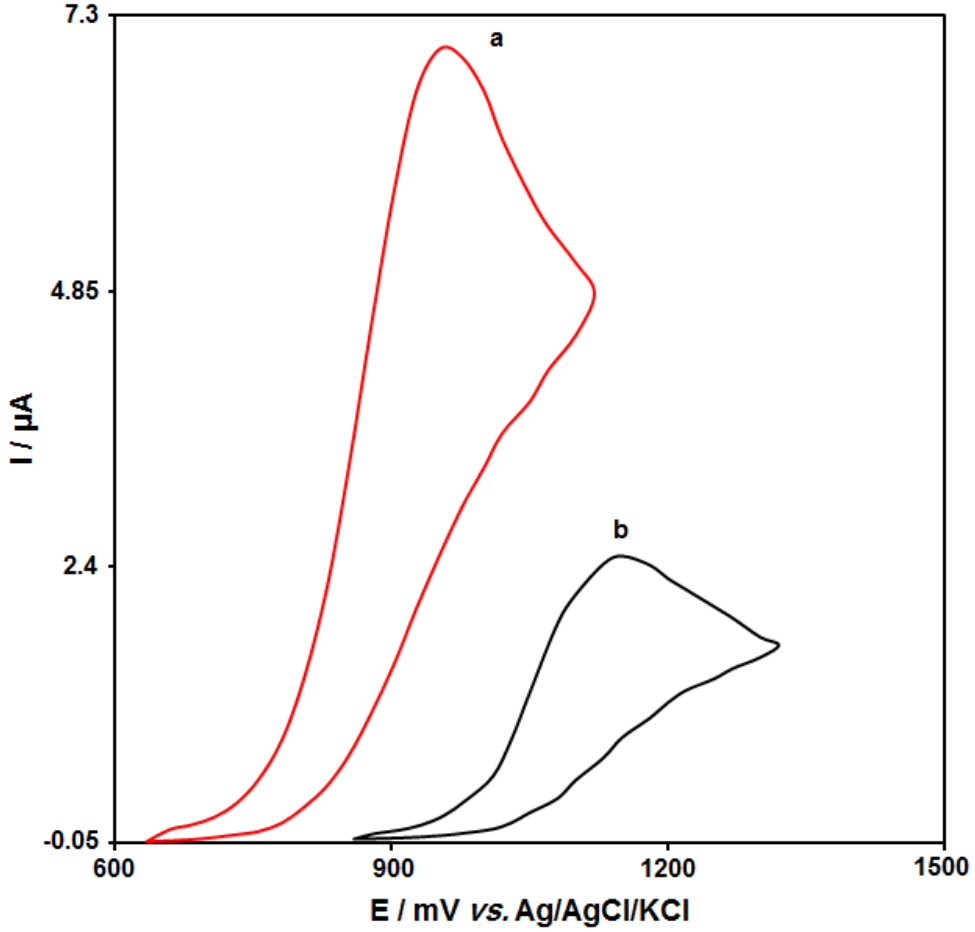

Figure 2. Cyclic voltammograms of (a) modified SPE and (b) bare SPE in the presence of 100.0 µM noscapine at the scan rate 50 mVs^{-1}.

A typical SEM image of La^{3+}/Co_3O_4 nanoflowers is shown in Fig. 1. Figure 2 shows responses of CV to electro-oxidation of 100.0 µM noscapine at the unmodified SPE (curve b) and modified SPE (curve a). The peak potential occurs at 950 mV, which is around 200 mV more negative than the unchanged SPE.

3.2. Effect of scan rateon Results

Findings indicated increasing in the current of the peak currents (Ip depends on $\nu^{1/2}$), therefore the mechanism is diffusion controlled [97].

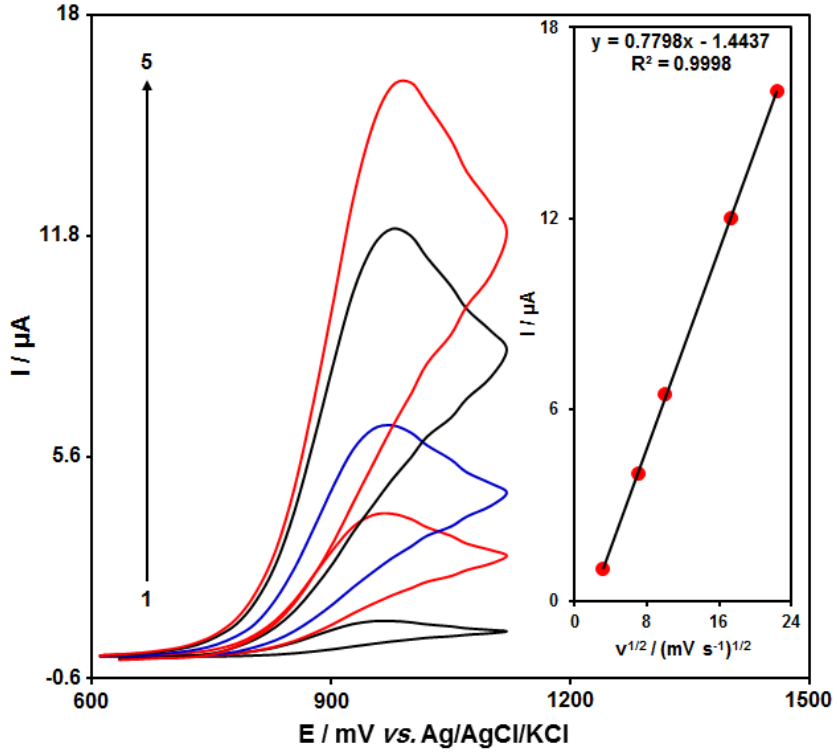

Figure 3. CV of modified SPE in the presence of 50.0 µM noscapine at various scan rates (10, 50, 100, 300 and 500 mV s^{-1}). Inset: variation of anodic peak current vs. $v^{1/2}$.

3.3. ChronoamperometricAnalyse

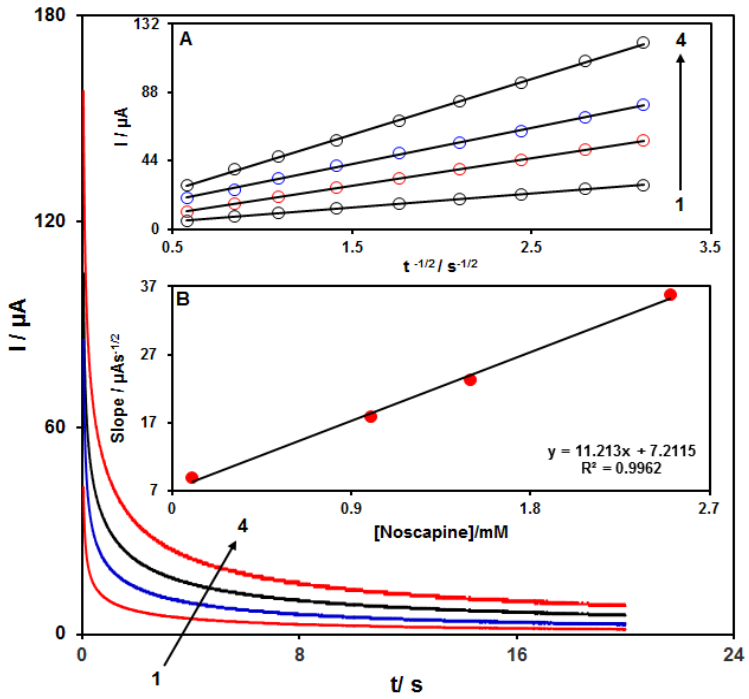

Figure 4. Chronoamperograms obtained at La^{3+}/ Co_3O_4/SPE for different concentration of noscapine (0.1, 1.0, 1.5 and 2.5 mM) (A) Plots of I vs. $t^{-1/2}$ obtained from chronoamperograms1–4. (B) Plot of the slope of the straight lines against noscapine concentration.

Chronoamperometric measurements of noscapine at La^{3+}/Co_3O_4/SPE were conducted by adjusting the working electrode potential at 1 V (Figure 4). According to the Cottrell equation [97], mean values of D were 1.1×10^{-5} cm^2/s for noscapine.

3.4. Calibration Curves

DPV was used for quantitative analysis of noscapine (Figure 5).

It was found that the peak currents of noscapine at modified SPE surface linearly depended on noscapine concentrations in the range of 0.3 to 600.0 µM, while detection limit (3σ) was achieved to be 0.1 µM.

Table 1.shows a comparison of voltammetric techniques for the detection of noscapineat the prepared electrode in this work and some other works.

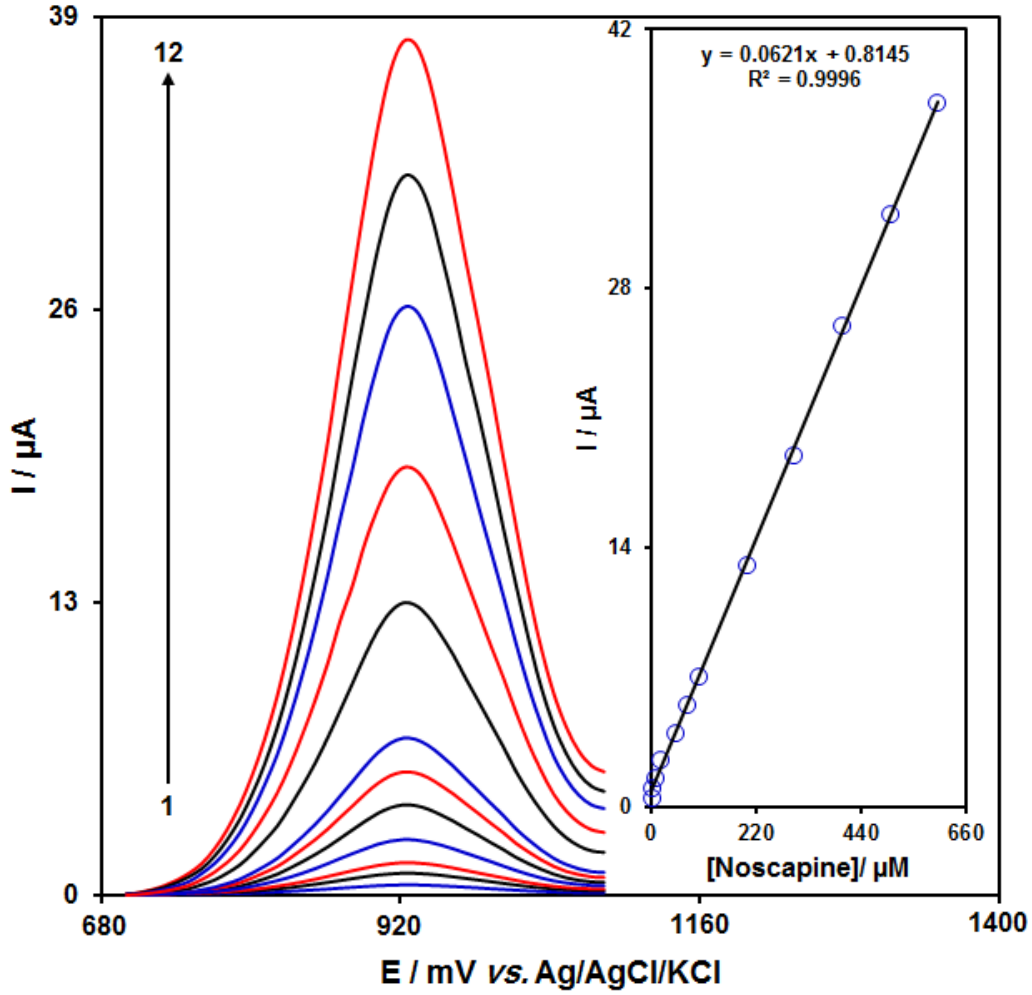

Figure 5. DPVs of La^{3+}/ Co_3O_4/SPE in solution containing different concentrations of noscapine. (0.3, 3.0, 10.0, 20.0, 50.0, 75.0, 100.0, 200.0, 300.0, 400.0, 500.0 and 600.0 µM). Inset: plot of the peak current as a function of noscapine concentration.

Table 1. A comparison of the efficiency of various modified electrodes reported for the detection of noscapine.

Electrode	Modifier	Method	Limit of Detection	Linear Range	Ref.
Carbon Paste Electrode	Alumina-borate oxide fibers/reduced graphene oxide (AlBFs/RGO)	DPV	0.049 μM	0.07-300 μM	5
Glassy carbon electrode	Graphenenanosheets (GNSs)	DPV	0.2 μM	1.0–35.0μM	7
Glassy carbon electrode	Multiwall carbon nanotubes (MWNTs)	Voltammetry	0.08	0.4-100.0	34
SPE	La^{3+}/Co_3O_4 nanoflowers	DPV	0.1 μM	0.3-600.0 μM	This work

3.5. Real Samples Analysis

The method illustrated above was used to evaluate La^{3+}/Co_3O_4/SPE usability for determining noscapine in noscapine tablet and urine samples. Therefore, the standard addition technique was applied (Table 2).

Table 2. The application of La^{3+}/Co_3O_4/SPEfor determination of noscapine in noscapine tablet and urine samples(n=5). All concentrations are in μM.

Sample	Spiked	Found	Recovery (%)	R.S.D. (%)
Noscapine tablet	0	2.5	-	3.2
	2.5	4.9	98.0	2.4
	5.0	7.7	102.7	2.5
	7.5	10.1	101.0	1.8
	10.0	14.9	99.3	2.2
Blood serum	0	-	-	-
	5.0	5.1	102.0	1.7
	10.0	9.9	99.0	2.9
	15.0	15.2	101.3	2.3
	20.0	19.7	98.5	3.4

4. CONCLUSION

Taking advantage of La^{3+}doped Co_3O_4 nanoflowers, present study examines modified screen printed electrode. The findings of the study showed that modified screen-printed electrodes have an excellent analytical performance in determining noscapine in apple juice and water specimens. An

excellent linear relationship was also observed between the peak currents and noscapine concentration. The detection limit for noscapine was 0.1 μM.

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