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Full Paper

A Modified Carbon Paste Electrode for Acetaminophen based on the Application of $YbVO_4$ Nanoparticles Prepared using a New Capping Agent

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Abstract- $YbVO_4$ nanoparticles were prepared using sodium tetraphenylborate ($Na(B(C_6H_5))$) as a novel capping agent. The effect of the capping agent on the morphology and size of the product particles was studied. The characterization of the particles was performed using X-ray diffraction (XRD), transmission electron microscopy (TEM), and vibrating sample magnetometery (VSM). The synthesized of $YbVO_4$ NPs nanoparticle was used to investigate the electrochemical behavior of acetaminophen in an aqueous medium using the linear sweep voltammetric (LSV) technique.

Keywords - $YbVO_4$, Precipitation method, Electrochemical, Acetaminophen

1. INTRODUCTION

Because size and shape of nanoscaled materials including nanospheres, nanoplates, and nanorods have a major role to play in the application of such compounds, they have been

received much attention in recent years [1-13]. Until now, some procedures have been suggested for fabrication of nanomaterial [6, 14-19]. However, from all of known methods, it has been confirmed that precipitation process is a simple route [20-22].

Lanthanide vanadates (MVO_4) as important compounds made from vanadium have been focused by a numerous researchers owing to their different structures which, in turn, can stem from the fact that vanadium have different coordination and valence numbers, and polyhedral coordination forms. Vanadates could be employed in varied field including ion-exchange, lithium ion batteries, water splitting, antibacterial additive, catalyst, photocatalyst, photoluminescence devices, supercapacitors, luminescence and gas sensors [23-28].

In this research $YbVO_4$ NPs were prepared using a new approach using $Na(B(C_6H_5)$ as surfactant, for the first time. The crystalline composition and morphological properties, of the products were evaluated through XRD, FTIR, energy dispersive X-ray microanalysis (EDX), transmission electron microscopy (TEM) and vibrating sample magnetometers (VSM). After synthesizing and optimizing the conditions, the prepared nanoparticle was used for studying and electrochemical measurement of acetaminophen as an anti-nociceptive and anti-nocentric drug.

2. EXPERIMENTAL

2.1. Chemicals and instrumental analyses

$Yb(NO_3)_3.6H_2O$, NH_4VO_3, and $Na(B(C_6H_5)$ were from Merck Co. The materials were not treated before use. XRD analyses were conducted on a Philips-X'pertpro spectrometer equipped with Ni-filtered Cu Ka radiation. Fourier transform infrared spectra were recorded on a Nicolet Magna- 550 instrument. The FTIR samples were loaded into the spectrometer using KBr pellets. EDX analyses were performed at an acceleration voltage of 20 kV. TEM images were recorded with a Philips EM208 instrument at 200 kV. VSM analyses were performed using a Meghnatis Daghigh Kavir Co. instrument (Kashan Kavir, Iran) under ambient conditions.

2.2. Synthesis of $YbVO_4$ NPs

The typical procedure for the preparation of the nanoparticles involved dissolving 1 mmol of $Yb(NO_3)_3.6H_2O$ dissolving in 30 mL of water. Then a volume of a water solution containing 3 mmol of $Na(B(C_6H_5)$ was added to this solution followed by adding 1 mmol of NH_4VO_3. This mixture was kept stirring at 70 °C for 60 minutes and the resulting yellow solid was filtered, desiccated at 100 °C, and then calcined at 500 °C for one hour.

2.3. Electrochemical procedures

Unmodified and modified carbon past electrodes (CPEs) were prepared using a mixture of graphite powder and paraffin oil (and the modifier in the case of the modified electrode). The unmodified or modified pastes were then filled into a part of a polyethylene tube. A thin Cu wire was inserted into the tube and then the paste from the opposite employed inside the tube connecting to the composite to form electrical contact. Finally, the outer surface of the paste accessible through the opening f the tube was smoothed manually by polishing.

3. RESULTS AND DISCUSSION

Considering XRD pattern of nanomaterial, one can obtain chemical structure and crystal size of the compounds [29-36]. The XRD results for $YbVO_4$ NPs prepared in the presence of $Na(B(C_6H_5))$ as a surfactant can be viewed in Fig. 1. For bare $YbVO_4$, three peaks are seen at 25.07° ((200) line), 33.67° ((112) line), and 49.92° ((312) line). These fully agree with reference data from JCPDS No 01-072-0860 file for tetragonal (space group I41/amd) phase. Using the XRD data (Fig. 1) and Scherrer's equation, the crystallite diameter (D_c) is given by (37):

$$D_c = K\lambda/\beta\cos\theta \quad (1)$$

Where β is the width of a line at the half intensity maxima, K is a shape factor (about 0.9), and λ represents the wavelength of the X-ray radiation applied. Accordingly the domain sizes of the crystals were determined as 11 nm, for $YbVO_4$.

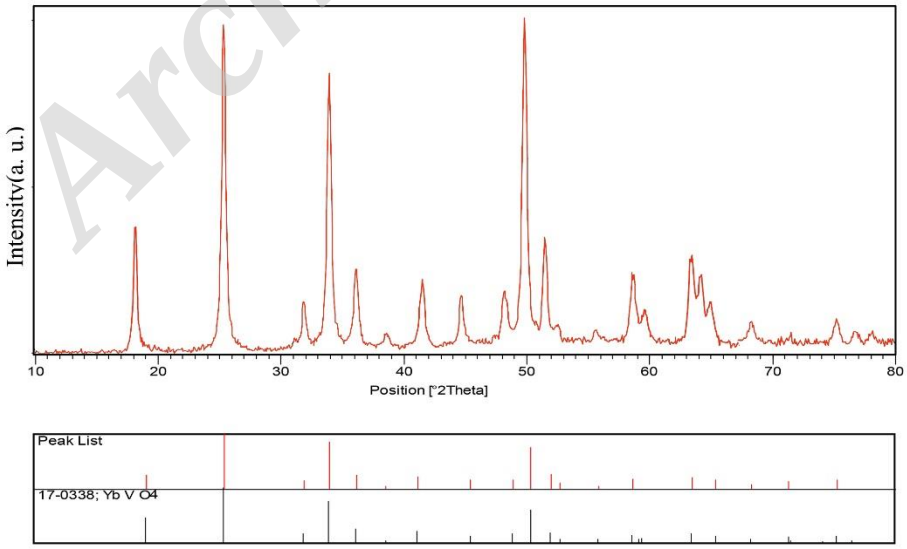

Fig. 1. The XRD pattern obtained for $YbVO_4$ NPs prepared using tetraphenylborate

EDS results (e.g. Fig. 2) indicated the $YbVO_4$ NPs were purely composed of vanadium, yttrium, and oxygen.

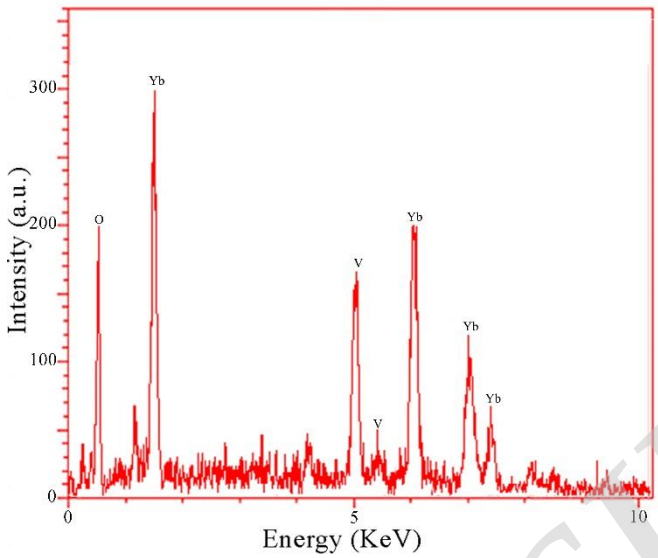

Fig. 2. EDS pattern obtained for the $YbVO_4$ NPs prepared using tetraphenylborate

Na(B(C6H5) is employed as a new surfactant for fabrication of $YbVO_4$ NPs. The TEM images display that prepared $YbVO_4$ NPs in the presence of $Na(B(C_6H_5)$ has a sphere-like morphology, with the size up to 20-80 nm (Fig. 3).

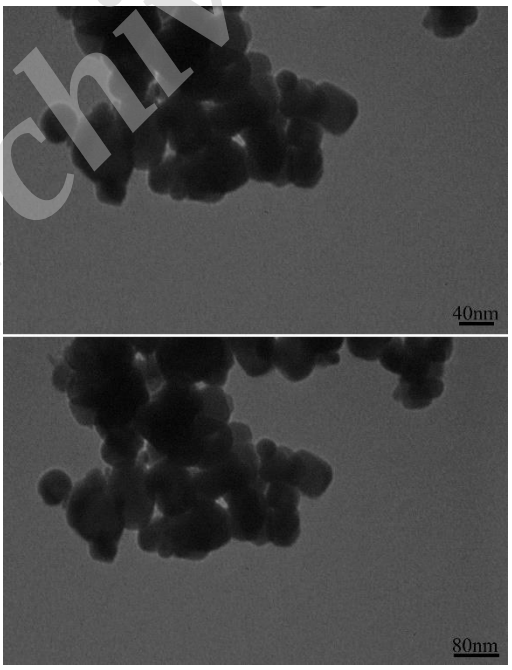

Fig. 3. TEM image of $YbVO_4$ NPs obtained in the presence of tetraphenylborate

The magnetic characteristics of the $YbVO_4$ NPs were determined using VSM at 300 K and the results have been displayed in Fig.4. It suggests that sample has paramagnetic behavior with magnetization value of 0.042 emu/g.

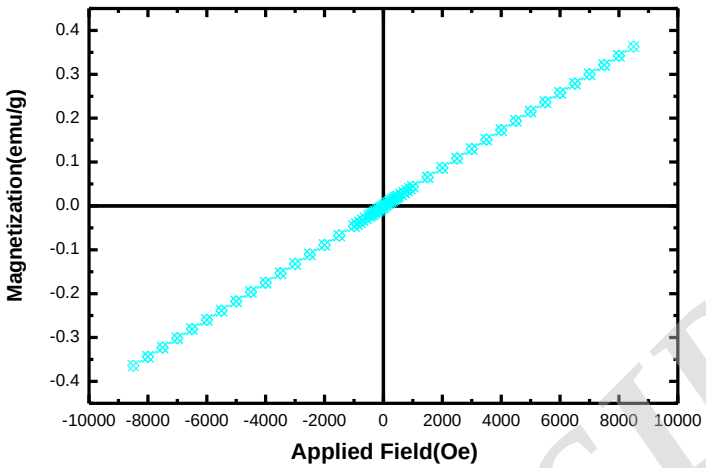

Fig. 4. VSM curve of of $YbVO_4$ NPs obtained in the presence of tetraphenylborate

To evaluate the chemical composition and quality of the $YbVO_4$ NPs synthesized in the presence of $Na(B(C_6H_5)$, FT-IR studies were conducted from 400 to 4000 cm^{-1} (Fig. 5). Accordingly, the band at 3438 cm^{-1} reflects the presence of hydroxyl (-OH) groups. The bands at 451 and 810 cm^{-1}, on the other hand, were attributed to the respective stretching and bending vibrations of Yb-O and V-O bonds [38].

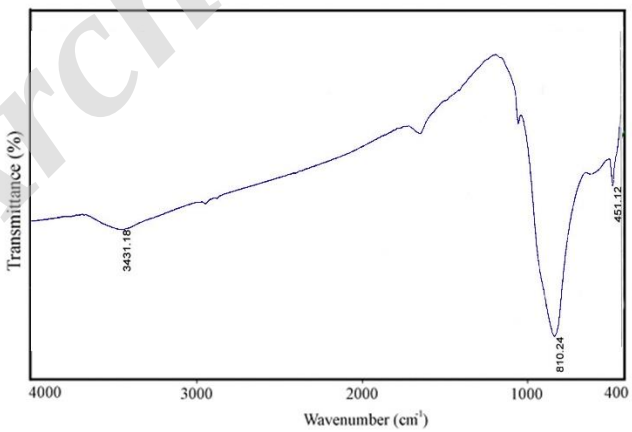

Fig. 5. FT-IR spectra recorded for the $YbVO_4$ NPs prepared using tetraphenylborate

Considering the great responsiveness and excellent performance of various nanoparticles and nanostructures in electrochemistry, many electrochemical scientists have focused on this field especially preparing the modified electrode using various nanostructures as sensing probe

for determination of different compounds [39-48]. Therefore, in this work the synthesized $YbVO_4$ NPs were also used to measure the acetaminophen. Fig. 6 shows the LSV of unmodified and modified electrodes of $YbVO_4$ NPs. Observations indicate that the current of acetaminophen at the modified electrode surface is 3.8 times greater than the unmodified electrode surface in phosphate buffer with pH= which represents a very good application of this synthesized compound for electrochemical measurement of acetaminophen.

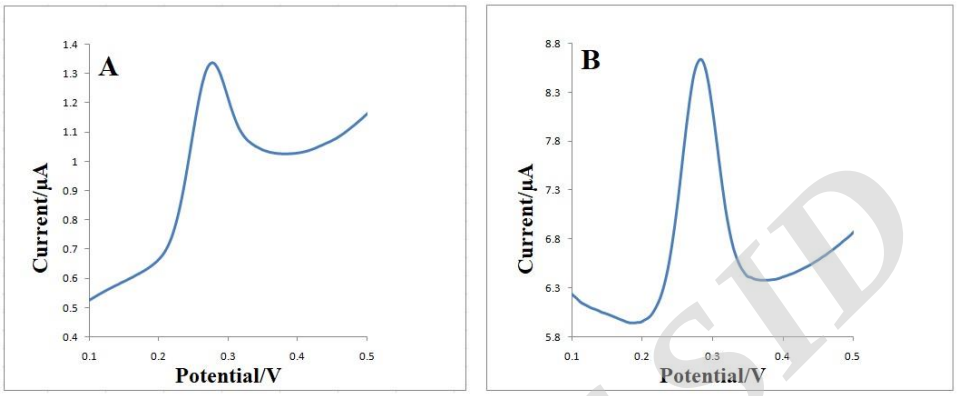

Fig. 6. LSVs of 15 µM of acetaminophen using unmodified CPE (A), and the carbon past electrodes modified using $YbVO_4$ NPs; (B) in 0.1 M PBS (pH=7), scan rate 50 mV/s

To better study the behavior of the modified CPE and determining the optimal conditions for its performance, the influence of pH on the electrochemical response of acetaminophen was monitored. Fig. 7 shows the LSVs in the phosphate buffer solution with pHs 2-9. The maximum current is related to the pH=7, so this pH was used in the following steps of this work. Also, the slope of pH values versus potential is 58 which is close to 59, so it can be concluded that an equal number of electrons and protons were involved in the reaction.

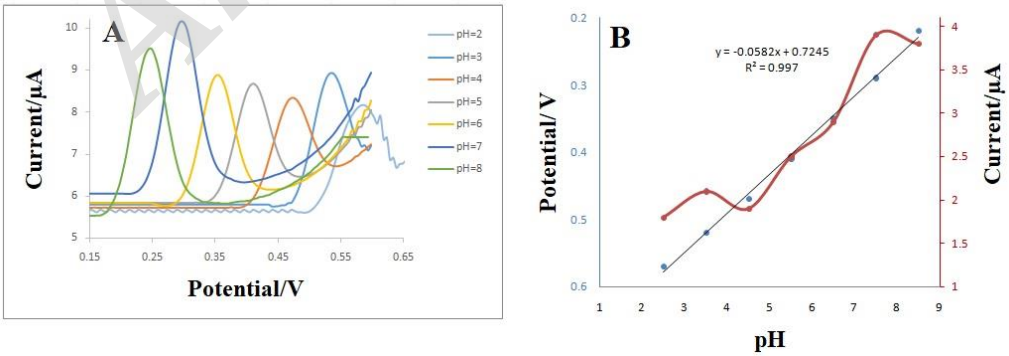

Fig. 7. (A) LSVs of 20 µM acetaminophen at the modified carbon past electrodes of $YbVO_4$ NPs in various pHs (from 2 to 9) of buffer solution; B) Dependence of current and potential (Scan rate=50 mV/s)

The relationship between changes in the scan rate and the oxidation signal for acetaminophen was investigated. Fig. 8 shows the LSVs recorded using in 0.1 M PBS (pH=7), while applying scan rates in the range of 10-400 mV. It was observed that a plot of the anodic peak currents against the square root of scan rate (Fig. 8B) was linear for scan rates of 20-400 mV/s. This is a typical behavior for processes with a diffusion-controlled nature.

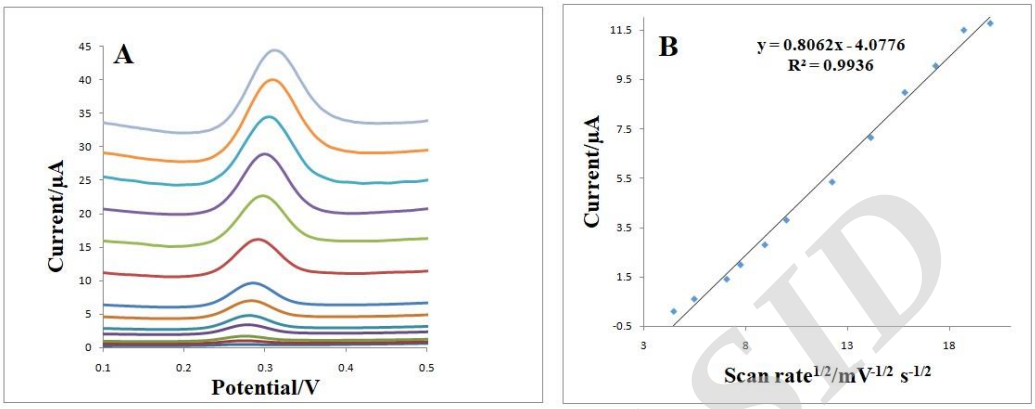

Fig. 8. Influence of varying the scan rates on the LSVs recorded for a 15 µM acetaminophen solution, in the range 20-400 mV/s; (B) the plot of changes in the anodic peak current against /root of scan rate recorded in a 0.1 M PBS (pH=7)

The LSV technique was used furthermore for analytical determination of acetaminophen. Fig. 9 shows the LSVs in the range of 5 to 50 µM of acetaminophen. The linear relationship is observed in this range and obtained detection limit is also 2.1 µM.

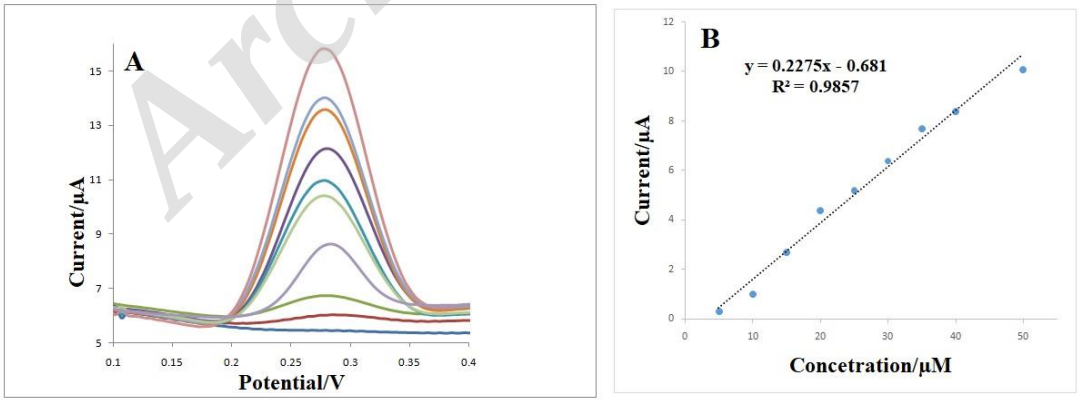

Fig. 9. LSVs for various concentrations of acetaminophen (5-50 µM) in 0.1 M phosphate buffer (pH=7); (B) The corresponding current *vs*. acetaminophen concentration plot recorded at 50 mV/s

4. CONCLUSIONS

Yttrium vanadate ($YbVO_4$) nanoparticles (average size= 40–60 nm) were prepared through a precipitation method and the crystal properties and morphology of them were studied using TEM and XRD techniques. Magnetic measurements on the as-synthesized $YbVO_4$ NPs indicate a paramagnetic property. Finally, the carbon paste electrode was modified with $YbVO_4$ NPs and used for electrochemical measurement of acetaminophen. At pH=7, the highest current was obtained for acetaminophen with modified electrode and detection limit was 2.1 μM.

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