



Preparation and evaluation of new uranyl imprinted polymer electrode sensor for uranyl ion based on uranyl–carboxybezotriazole complex in pvc matrix membrane



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ABSTRACT

In this study a new uranyl selective electrochemical sensors were prepared by using uranyl ionic imprinted polymer (IIP). The IIP was prepared by thermal polymerization using acrylic acid as a monomer, ethylene glycol dimethacrylate (EGDMA) as a cross linking and a benzoyl peroxide (BPO) as an initiator. Uranyl–carboxybenzotriazol (UO₂–CBT) complex was used as an active material on the prepared polymer. Several uranyl electrodes were constructed by using different masses of polyvinyl chloride (PVC) matrix. Electrode parameters including slopes, working concentrations, pH, and interferences were evaluated. The electrodes exhibit a Nernstian response with slopes of 23.6 and 28.1 mV/decade for graphite uranyl electrode and liquid uranyl electrode, respectively, over a wide range of concentration from 3×10^{-6} to 6×10^{-2} M and a detection limit of 1×10^{-6} M. It can be used in the pH range of 4.3–10.5 with a response time of less than 60 s. The effect of ions interferences (such as: Cr³⁺, La³⁺, Ba²⁺, Ca²⁺, Cu²⁺, Na⁺, and K⁺) on the electrode response were evaluated. The IIP and nonimprinted polymer (NIP) membranes were characterized by Fourier Transform Infrared Spectroscopy (FTIR) and Scanning Electron microscopy (SEM). The binding capacity and equilibrium dissociation constant were determined by studying the binding isotherm. The concentrations of uranyl ion in the prepared synthetic solutions determined by the standard addition method and the results were satisfactory with errors less than 7%. Finally, the prepared graphite UO₂-IIP sensor was tested to determine the uranyl ions concentrations in a real environmental water sample by standard addition method. The results were compared with those obtained by spectrophotometric method which showed a good agreement. The developed electrode was found to be fast, sensitive and reliable indicated its potential use in measuring the uranyl ion concentration in the field.

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1. Introduction

Uranium contamination can be introduced to the environment naturally from certain types of soils and rocks. Also, it can be introduced from mining, manufacturing activities, coal and other fuel combustion and nuclear power production. This raises the concerns of detecting such contamination in water due to its toxicity. Furthermore, measurement of uranium concentration is having a great importance in nuclear industries due to the application of metal as fuel in nuclear reactors.

There are various techniques have been used to determine uranyl ions concentration, such as determination of uranyl in sea water by spectrophotometric method [1,2],

determination of uranyl adsorption onto poly(acrylamide-g-ethylenediaminetetraacetic acid) hydrogels in the presence of cadmium and lead by polarographic method [3]. The structural properties of UO₂ complex with macro cyclic ligands were studied by NMR [4].

The presence of uranium in both water and soil environment and the limitations associated with measuring uranium by the reported techniques [1–4] create a need to develop a fast, sensitive and reliable method for uranium determination in various environmental samples. Ion-selective electrodes methods developed due to their unique advantages such as speed, sensitivity and ease of preparation. Zamani et al. [5] fabricated new gadolinium potentiometric sensor for lanthanide series determination. Neodymium (Nd(III)) electrode based on azo calix[4]arene was constructed by Menon et al. [6]. Drug selective electrodes were constructed by using chlorpromazine [7] and sibutramine [8]. Various ligands have been used to prepare the uranyl sensors such as

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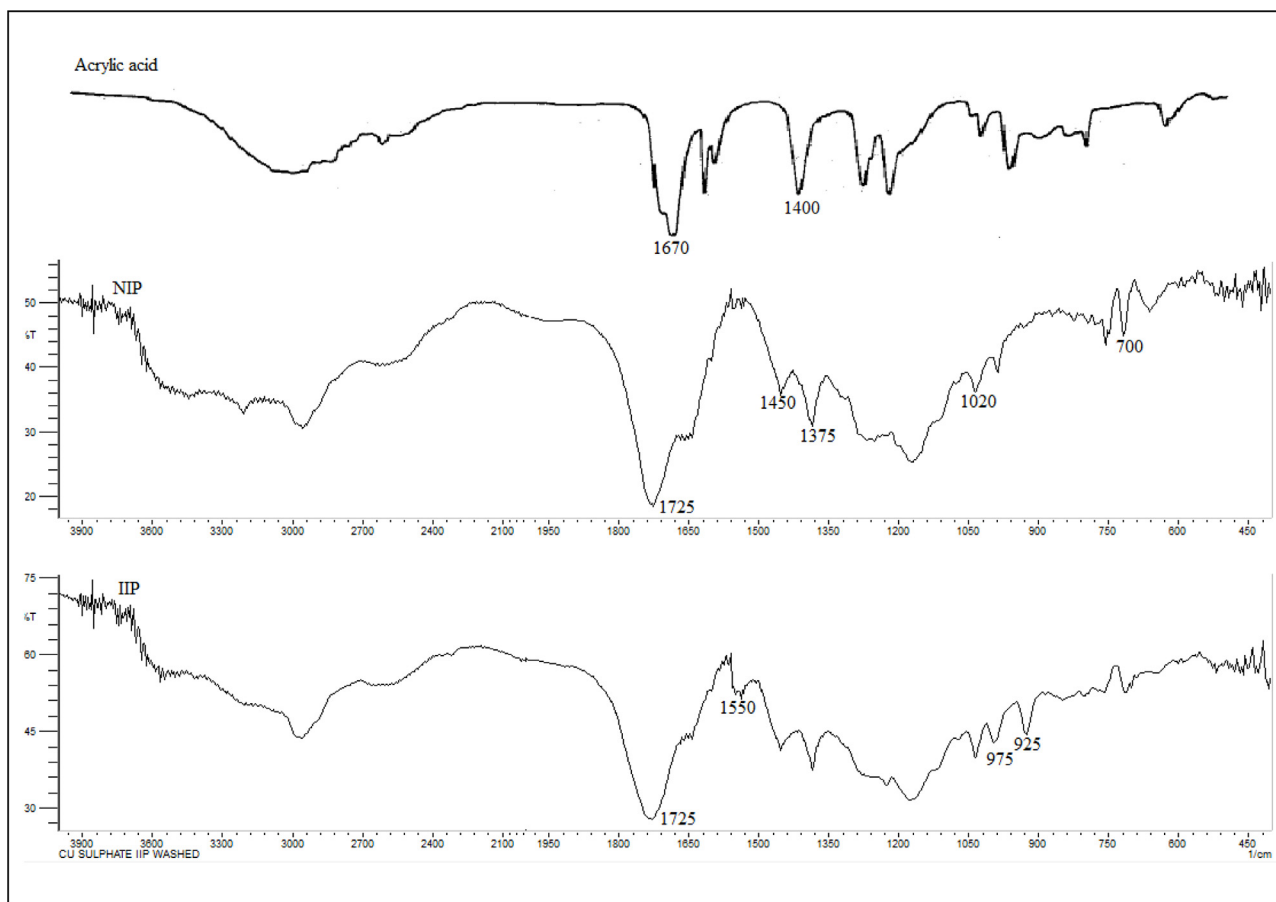


Fig. 1. FTIR spectra of acrylic acid, NIP, and IIP.

1-(4-carboxyphenyl)-1H-1,2,3-triazole-4-carboxylic acid [9], and crown ether [10]. Shokrollahi et al. [11] prepared and studied the electrodes parameter for uranyl sensors based on bis(2-hydroxy acetophenone) ethylene diimine. The construction and performance characteristics of uranyl membrane electrodes based on amino (tri-methyl) phosphate were developed [12], and the developed membrane electrodes found to be selective towards uranyl ion over a large number of interfering cations. Nassory et al. [13] were prepared uranyl selective electrodes using crown compounds in different plasticizers, and used for determination of uranyl ions in uranium samples. Metilda et al. [14] prepared ion imprinted polymer (IIP) materials for uranyl ion by forming binary 5,7-dichloroquinoline-8-ol (DCQ) or 4-vinylpyridine (VP), the synthesized polymer particles were characterized by FTIR, elemental analysis (CHN), X-ray diffraction and pore size analysis. Molecularly imprinted polymer (MIP) have been prepared and fully characterized, as an example diphenylamine MIP [15], metoprolol [16], dopamine [17] and dextromethorphan [18].

Ion-imprinted polymers (IIP) has a promising properties such as short response time, good sensitivity and selectivity, good stability, low cost and ease of preparation. Therefore, the research activity in the field of ion-imprinted polymers is increasing drastically. Ion-imprinted polymers properties are remarkable because of their high selectivity towards the target ion due to a memory effect resulting from their preparation process [19]. The size and shape of the generated IIP cavities and the affinity of the ligand for the imprinted metal ion are responsible for the high selectivity of IIP. Furthermore, a favorable environment for the template ion

rebinding can be generated by maintaining the complex geometry through the crosslinking and leaching steps. The IIPs have good stability and can be reused without loss of activity, due to highly crosslinking polymers. The versatility of the ion-imprinting technology makes it possible to forecast significant improvements.

Although bulk polymerization has been extensively used to produce IIPs in a very easy fashion, it clearly appears that the control of their shaping is a crucial issue. The optimization of the performances of the materials, eventually incorporated in sophisticated devices, will depend on that point. As far as particles are desired, the suspension and precipitation polymerization techniques are attractive alternatives. They still require some new developments to control the particles characteristics in terms of size, shape and porosity. Surface imprinting is also a promising route to prepare highly efficient IIPs, especially with the generation of hybrid materials. It can be anticipated that a trend will be to assure a better control of the IIPs elaboration [19].

Recently, our research group fabricated several molecularly imprinted polymer (MIP) sensors for determination of azithromycin determination [20]. This study aimed to investigate new graphite uranyl ion selective electrodes which manufactured using uranyl-carboxybenzotriazole complex based on PVC matrix membrane and uranyl ion-imprinted polymer. These new ion selective electrodes will be used as a sensor, which provide a simple and sensitive method for uranyl determination in the presence of interfering metal ions in environment samples and uranium salts.

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