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Studies on adsorption property of novel composite adsorption material PEI/SiO₂ for uric acid

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Abstract

Polyethyleneimine (PEI) was grafted onto the surface of silica gel particles via the coupling effect of γ -chloropropyl trimethoxysilane (CP), the composite adsorption material PEI/SiO₂ with strong adsorption action for uric acid was prepared. Static adsorption experiment results show that macromolecules of PEI grafted onto silica particles have very strong adsorption ability by right of strong physical adsorption induced by the hydrogen bond interaction between PEI and 2,6,8-trihydroxypurine which is a tautomer of uric acid, and chemical adsorption of a certain extent caused by Schiff base reaction. The isothermal adsorption fits to Freundlich equation, and the maximum adsorption amount reaches 84.9 mg/g. The pH value of the medium influences greatly the sorption, in neutral solution (pH=6–7), PEI/SiO₂ displays strong adsorption ability for uric acid, whereas in acidic and basic solution, the adsorption is weak. Temperature also has some influence on the adsorption property of PEI/SiO₂, and the adsorption capacity increases with rising temperature.

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1. Introduction

Uric acid is a catabolism product of nucleoprotein and nucleic acid. There is a great deal of excess uric acid in blood of patients with kidney failure. The cumulated uric acid can be removed effectively by using blood purification techniques so that the toxicosis symptom of patients can be mitigated. It is an attractive research subject in the field of biomedicine engineering to prepare polymer adsorption material with high property in order to eliminate excess endogenesis toxin [1-4], and it is also a key to develop blood purification techniques. Different polymer adsorption materials were used to eliminate small molecular endogenesis catabolism products, such as uric acid. Some researchers adopted adsorption resins with non-polarity and weak polarity, which exert sorption through hydrophobic interaction and remove uric acid to a certain extent [5,6]. Someone used crosslinking polystyrene microspheres chemically modified with multiethylene-multiamine [7], however, the adsorption capacity was lower and the adsorption mechanism was not discussed.

Polvethyleneimine (PEI) is a typical water-soluble polvamine, and there is a large quantity of nitrogen atoms of amino groups on the macromolecular chains. The molecules of commercial PEI often have branch chains (Branched PEI), and it contains primary, secondary and ternary amino groups in a ratio of 1:2:1 approximately [8]. PEI macromolecules have strong property of conferring electrons, display very strong protophilia owing to the abundant nitrogen atoms on macromolecules, and the strong hydrogen bond interaction can be produced between PEI and proton donors. Besides, majority of amino groups on the molecular chains is in protonize state in aqueous solution of pH<10, so PEI is one kind of cationic polyelectrolyte [9,10]. These characteristics of PEI have attracted wide attention of scholars, and PEI has been used in separation, purification and immobilization of biomacromolecules [10], in constructing pH sensor and biosensor [11,12], in drug releasing system [13,14] and so on.

Uric acid and 2,6,8-trihydroxypurine are tautomer to each other, and hydrogen bond can form between the hydroxyl groups in molecular structure of trihydroxypurine and electron donors. By designing the chemical structure of the adsorption material, in this paper, macromolecule PEI was grafted onto silica gel surface via the action of a coupling agent, the

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concentrating effect of functional groups of PEI was combined with a high specific area and high mechanical strength of silica gel particles, and novel composite adsorption material of PEI/ SiO₂ for medical application was prepared. It was expected that PEI/SiO₂ would produce strong sorption for uric acid by right of the hydrogen bond interaction between PEI and uric acid (trihydroxypurine). The experiment results of static adsorption show that PEI/SiO₂ has very strong adsorption and removal effect for uric acid indeed; the maximum adsorption amount reaches 84.9 mg/g. This paper still probed deeply into the adsorption mechanism of PEI/SiO₂ for uric acid, examined the effects of the pH value of medium, temperature on sorption, and it is expected to supply some theoretical references for developing blood purification materials. The blood compatibility of PEI/SiO₂ needs to be studied further.

2. Experiments

2.1. Materials and instruments

Silica (120–160 mesh, Qingdao Ocean Chemical Limited Company, Qingdao in China) was of agent grade, γ chloropropyl trimethoxysilane (Yongchang Chemical Limited Company, Naking in China) was of analytical grade, and polyethyleneimine ($M_r=2 \times 10^4$ to 5×10^4 , Qianglong Chemical Limited Company, Wuhan in China) was of chemical grade. Methane sulfonic acid, sodium tungstate and other chemicals used were all of commercially analytical grade, and were purchased from Chinese companies.

Used instruments were as follows: Unic-2602 UV spectrophotometer (American Unic Company), 8400S Shimadzu FTIR spectrometer (Japanese Shimadzu Company), DDS-11Ar digit conductivity meter (Leici Instrument Limited Company of Shanghai), PHS-2 acidimeter (The Second Analytical Instrument Factory of Shanghai), TG16-WS high speed centrifuge (Changsha Xiangyi Centrifuge Factory, China), and THZ-92 constant temperature shaker equipped with gas bath (Boxun Medical Treatment Equipment Factory of Shanghai).

2.2. Preparing composite adsorption material PEI/SiO₂

The preparing process was as follows: firstly, silica gel particles was treated for activating by using aqueous solution of methane sulfonic acid; secondly, activated silica gel reacted with γ -chlopropyl trimethoxysilane (CP) at 80 °C by using xylene as solvent into which a small quantity of water was added and chlopropylation silica (CP-SiO₂) was prepared; finally CP-SiO₂ was added into PEI aqueous solution with certain concentration, then reaction was carried out at 90 °C for certain time, PEI was grafted onto silica gel surface by coupling mode, and the novel composite adsorption material PEI/SiO₂ was prepared. The chemical structures of CP-SiO₂ before and after grafting were characterized by infrared spectrum in order to confirm that PEI had been grafted onto silica particle surface. The amount of amino groups on PEI/SiO₂ was determined using the conductivity titration method

in which hydrochloric acid was used as titrant, and the grafting amount (g/100 g) of PEI was calculated further. Through varying the concentration of PEI solution during grafting reaction in a coupling manner, PEI/SiO₂ with different grafting amount of PEI were prepared.

2.3. Plotting standard curve of uric acid [15]

First, salkowskis solution was prepared through the reaction of sodium tungstate and phosphoric acid. Certain amount of dried uric acid was weighed and dissolved in distilled water, the solution was transferred into a measuring flask, and was made at a constant volume. Uric acid aqueous solutions with different concentrations were prepared by diluting with distilled water. 0.5 mL of salkowskis solution and 0.5 mL of Na₂CO₃ solution were added into 3 mL of uric acid solution; the mixed solution was placed for 15 min, and its absorbency was measured on Unic-2602 UV spectrometer at 680 nm that is the character absorption spectrum of tungsten blue, which is the product of the reaction between uric acid and salkowskis. After measuring the absorbencies of various solutions, the standard curve of uric acid was plotted.

2.4. Measuring kinetic curve of adsorption

A certain amount of uric acid solution with 2 mg/mL of concentration was taken with pipet and transferred into a conical flask with a plug, and about 0.2 g of PEI/SiO₂ weighed accurately was added. The suspension was shaken on shaker, after a certain time interval the suspension was taken out and separated centrifugally. The uric acid concentration of the supernatant was determined by using the spectrophotometric method according to the standard curve, and the adsorption amount of uric acid was calculated according to Eq. (1). The curve of adsorption amount against time was plotted, the adsorption rate was examined, and the time in which the adsorption reached equilibrium was determined.

$$\Gamma = \frac{(C_0 - C)V}{m} \tag{1}$$

In Eq. (1), C_0 (mg mL⁻¹) and *C* are the initial (without absorbent) and final concentrations of uric acid after adsorption, respectively; *V* is the volume of solution; *m* is the mass of absorbent PEI/SiO₂; Γ (mg g⁻¹) is the adsorption quantity of uric acid on PEI/SiO₂.

2.5. Measuring adsorption isotherm with static method

Numbers of uric acid solutions of 200 mL with the concentrations in a range of 0.1–1.0 mg/mL were taken respectively and their pH values were adjusted, about 0.2 g of PEI/SiO₂ weighed accurately were added, these suspensions were shaken on shaker at a constant temperature for 2.5 h, and the sorption was made to reach equilibrium. After centrifugal separation, the equilibrium concentrations C_e of uric acid were determined, the equilibrium adsorption quantity Γ_e of uric acid

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