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## Preparation of magnetically recoverable mesoporous silica nanocomposites for effective adsorption of urea in simulated serum

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## ABSTRACT

In this study, SBA-15/Fe<sub>3</sub>O<sub>4</sub> nanocomposites were prepared by conjugating Fe<sub>3</sub>O<sub>4</sub> nanoparticles to mesoporous silica SBA-15 particles as magnetically recoverable adsorbents. The physicochemical and magnetic properties of bare and as-prepared samples were characterized using the X-ray diffractometer, transmission electron microscope, Fourier transform infrared spectroscope, porosimeter, and vibrating sample magnetometer. The performance of SBA-15, Fe<sub>3</sub>O<sub>4</sub>, and SBA-15/Fe<sub>3</sub>O<sub>4</sub> nanocomposites for adsorption removal of uremic toxin urea in simulated serum was compared at 25 and 37 °C. Except for Fe<sub>3</sub>O<sub>4</sub>, both SBA-15 and SBA-15/Fe<sub>3</sub>O<sub>4</sub> showed a high and equivalent adsorption ability for urea. Although the surface area of SBA-15/Fe<sub>3</sub>O<sub>4</sub> nanocomposites decreased from 1167 m<sup>2</sup>/g (bare SBA-15) to 311 m<sup>2</sup>/g, the pore volume of SBA-15/Fe<sub>3</sub>O<sub>4</sub> was not greatly decreased because most of Fe<sub>3</sub>O<sub>4</sub> nanoparticles were coated onto the surface of SBA-15 particles. The adsorption mechanism of urea on SBA-15/Fe<sub>3</sub>O<sub>4</sub> was proposed. Finally, the cytotoxicity of SBA-15/Fe<sub>3</sub>O<sub>4</sub> nanocomposites before and after urea adsorption for fibroblast L929 cells was studied.

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## 1. Introduction

The incidence of chronic kidney disease (CKD) is continuously rising to an alarming region in the developing countries, presumably because modern people often overwork, stay up all night, withstand large pressure of life, and eat high salt and oil food as well as are countered many risk factors including hypertension, diabetes, aging, and the abuse of drugs that are toxic to kidneys [1,2]. CKD frequently associates with an increased risk of hospital admission, morbidity, and death due to the cardiovascular disease [3] and the progressive loss of kidney function. One of the main reasons to cause CKD is the abnormally high levels of low molar-mass uremic toxins in human body including urea, creatinine, and *p*-cresol [4,5]. Regarding the long list of such uremic toxins, the level of urea is the highest [5]. The concentration of urea in a healthy adult is less than 6.7 mM, whereas is 38 ± 18 mM for a kidney failure patient which is more than the level that kidneys can metabolism [5].

Kidney damage is permanent and the body is unable to repair necrosis kidney cell [6]. To relieve uncomfortableness resulting

from high levels of uremic toxins, the kidney disease patients must perform hemodialysis several times a week. At this stage, urea can be partly removed by low- or high-flux hemodialyzer; however, it must spend 3–4 h in a hemodialysis course. After hemodialysis, patients need to rest 3–4 h to regain physical strength, because in addition to urea, the nutrients such as amino acids, water-soluble vitamins, proteins, and iron are also cleared during hemodialysis [7–9]. Moreover, frequent use of hemodialyzer and hospitalization may increase the likelihood of developing infections such as hepatitis B and AIDS. If the frequency and duration of hemodialysis is reduced, most of the above drawbacks can be avoided. It has been accepted that the combination of hemodialysis and hemoadsorption offers a potential and promising method for this purpose [10].

Literature survey indicated that the widely-used materials for hemoadsorption include activated carbons [4], zeolites [10], and mesoporous silica [11–13]. Activated carbons have a high surface area and are easily available; however, their affinity toward urea is rather low. The amount of activated carbons required to clear urea is much larger than other uremic toxins such as creatinine and *p*-cresol [5,14]. Apart from traditional activated carbons, based on high surface area alone, zeolites and mesoporous silica are the possible candidates for urea adsorption [4,14]. The pore size of mesoporous silica can be readily varied compared to zeolites [15]. Moreover, various surface functional groups can be easily

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introduced on mesoporous silica to enhance adsorbent–adsorbate interactions, thereby improving its adsorption ability. Common variants of mesoporous silica include MCM-41 [12], SBA-15 [13], and FSM-16 [16]. Although MCM-41 and FSM-16 possess higher surface area, their pore size is smaller than the size of urea molecule. That is, urea cannot be absorbed into the pore surface of MCM-41 and FSM-16 [12,13,16]. Therefore, SBA-15 was selected in this work due to its larger pore diameter and easy surface functionalization without risking the closure of pores [17,18].

As expected, one important problem is encountered in the case of extracorporeal hemoadsorption operation. After adsorbing urea, the SBA-15 particles still flow and mix with blood in the vessel and cannot be easily separated from the blood because silica has no polarity. In this regard, the combination of mesoporous silica and magnetic nanoparticles can be one solution [19,20]. In many kinds of magnetic nanoparticles, the most commonly used is iron oxide such as  $\text{Fe}_3\text{O}_4$  and  $\gamma\text{-Fe}_2\text{O}_3$ . They both have strong magnetic properties, susceptible to magnetic field to control the movement. Also, iron oxide is inherently biodegradable [21]. United States Food and Drug Administration Association (FDA) has certified that iron oxides ( $\text{Fe}_3\text{O}_4$  and  $\gamma\text{-Fe}_2\text{O}_3$ ) is harmless to human body [22]. The movement of magnetic materials is easily controlled by magnetic field; therefore, they are widely used in biomedical fields.

In this work, the adsorption removal of urea from simulated serum was studied using magnetically recoverable adsorbents derived from  $\text{Fe}_3\text{O}_4$  nanoparticles and mesoporous SBA-15. Our results indicated that  $\text{Fe}_3\text{O}_4$  nanoparticles could be coated onto SBA-15 particles. The concentration of urea was reduced from 38 mM to 12 mM by adsorption for 100 min. It is expected that the combination of hemoadsorption and magnetic materials could aid and improve the efficiency of hemodialysis in the future, enabling the patients to not only sustain their life and but also improve life quality.

## 2. Experimental

### 2.1. Materials

The chemicals including urea (Mw, 60.1 g/mol), tetraethyl orthosilicate (TEOS), HCl, NaCl, KCl,  $\text{NaH}_2\text{PO}_4$ ,  $\text{Na}_2\text{HPO}_4$ , and  $\text{NH}_3$  solution were purchased from Sigma-Aldrich Co. (St. Louis, MO, USA). The non-ionic surfactant triblock copolymer poly(ethylene oxide)-poly(propylene oxide)-poly(ethylene oxide) (pluronic P123,  $\text{EO}_{20}\text{PO}_{70}\text{EO}_{20}$ , MW 5,800), which used as a structure-directing agent or soft template, was supplied by Sigma-Aldrich Co. The deionized (DI) water was used for preparing the solutions throughout this work. Salts  $\text{FeCl}_2\cdot 4\text{H}_2\text{O}$  and  $\text{FeCl}_3\cdot 6\text{H}_2\text{O}$ , the precursors of  $\text{Fe}_3\text{O}_4$ , were supplied by Riedel-de Haën Co. (Morristown, NJ, USA). All chemicals were analytical grade.

The L929 fibroblast cells (RM60091) were offered from the Bioresource Collection and Research Center (BCRC), Taiwan. The culture media of Dulbecco's modified Eagle's medium (DMEM) and fetal bovine serum (FBS) were supplied by Sigma-Aldrich Co. (USA). The cell proliferation reagent WST (water-soluble tetrazolium salts)–1, purchased from Takara Bio, Inc. (Japan), was used for nonradioactive and spectrophotometric quantification of cell proliferation and viability in cell populations.

### 2.2. Synthesis of mesoporous silica SBA-15 [23,24]

In a standard synthesis procedure, 4 g of P123 was mixed with 30 mL of DI water and 120 mL of 2 M HCl at room temperature with constant stirring. After the mixture was heated increased to 40 °C in water bath, TEOS (8.5 g) was added dropwise into the solution under vigorous mixing. Once the TEOS was dripped completed, the solution was agitated at 120 rpm for 20 h. Next, the

suspension was transferred into a Teflon bottle and hydrothermally aged at 100 °C for 24 h in an oven. After aging process, the solution was cooled to room temperature and the resulting solid was recovered by centrifugation. The solid product was washed and dried overnight at 100 °C. The dried powder was calcined at 500 °C for 5 h to burn off P123 template to facilitate the formation of pores.

### 2.3. Synthesis of magnetic $\text{Fe}_3\text{O}_4$ nanoparticles [25]

The synthesis consisted of dissolving 8.46 g of  $\text{FeCl}_2\cdot 4\text{H}_2\text{O}$  and 22.95 g of  $\text{FeCl}_3\cdot 6\text{H}_2\text{O}$  in 500 mL of DI water and degassing the solution with  $\text{N}_2$  gas. The reaction mixture was placed in an oil bath at 80 °C, to which 20 mL of aqueous  $\text{NH}_3$  solution (14.7 mM) was then added dropwise. The reaction was allowed proceeding for 1 h at 80 °C. Black reaction product was washed several times with DI water until the pH of the washing water reached 7.0. The product was then dried at 60 °C overnight. To prevent the transformation of magnetite to maghemite in air, the whole synthetic processes were kept in  $\text{N}_2$  atmosphere.

### 2.4. Conjugation of $\text{Fe}_3\text{O}_4$ nanoparticles onto SBA-15 to form SBA-15/ $\text{Fe}_3\text{O}_4$ nanocomposites

One gram of SBA-15 was dispersed in 30 mL of DI water, to which 0.63 g of  $\text{FeCl}_2\cdot 4\text{H}_2\text{O}$  and 1.73 g of  $\text{FeCl}_3\cdot 6\text{H}_2\text{O}$  were added. An amount of aqueous  $\text{NH}_3$  solution (35 mL, 14.7 mM) was then added dropwise to the solution. The mixture was vigorously stirred and degassed with  $\text{N}_2$  gas at 80 °C. The solution became black immediately under continuous stirring for another 5 h. After that, the mixture was washed with equal volume of DI water, and was centrifuged repeatedly until the residue solution became neutral. The synthesized SBA-15/ $\text{Fe}_3\text{O}_4$  nanocomposites were finally dried in a vacuum desiccator at 80 °C for 12 h.

### 2.5. Conjugation of SBA-15 onto $\text{Fe}_3\text{O}_4$ nanoparticles to form $\text{Fe}_3\text{O}_4$ /SBA-15 nanocomposites

First, 0.4 g of  $\text{FeCl}_2\cdot 4\text{H}_2\text{O}$  and 0.6 g of  $\text{FeCl}_3\cdot 6\text{H}_2\text{O}$  were dispersed in 200 mL of DI water. The aqueous  $\text{NH}_3$  solution (10 mL, 14.7 mM) was then added dropwise to the solution over 1 h and the mixture was degassed with  $\text{N}_2$  gas. The black reaction product was repeatedly washed several times with DI water until the pH of the suspension became nearly 7.0. Then, 1 g of SBA-15 was added to the previous solution. The black material was dried in a vacuum desiccator at 80 °C for 12 h and it was designated  $\text{Fe}_3\text{O}_4$ /SBA-15 nanocomposites.

### 2.6. Characterization of the prepared samples

The magnetic properties of as-prepared samples were measured by the vibrating sample magnetometer, VSM (DMS model 1660, ADE Technologies Inc., MA, USA). Before the measurements, the magnetic powders were compacted into a hollow plastic cylinder box. The samples were measured at room temperature with a maximum magnetic applied field of 1.0 Tesla. Surface functional groups of the as-synthesized samples were determined by the Fourier-transform infrared (FTIR) spectroscopy (PerkinElmer Spectrum 100, PerkinElmer, MA, USA) using KBr pellet method. Samples were compactly flatted on a glass slides for XRD characterization. Crystal structures of the samples were analyzed by a D2 PHASER X-ray diffractometer, XRD (Bruker, Germany) with a scanning region of  $2\theta$  from 10° to 70°. The diffraction patterns of small angle X-ray scattering (SAXS) (PSAXS-USH- WAXS-002, Osmic Inc., Cleveland, OH, USA) were recorded at 40 kV voltage and 40 mA current. The

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