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Highly efficient capture of naphthalene by nonionic surfactants in hydrogel capsules

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ABSTRACT

The impregnation of Triton X-100 (TX100) in chitosan capsules (TCC) via an anionic surfactant gelation method was investigated for the sorption of naphthalene (NAP) from aqueous solutions. Impregnation with TX100 enhanced NAP sorption at all tested concentrations of TX100, ranging from 0.1 to 10 g/l. The highest NAP sorption capacity (68.7 mg/g) was obtained at 2 g/l of TX100, a capacity 5.1-fold higher than that of chitosan capsules without TX100 impregnation (CC) (13.4 mg/g). The highest molar sorption ratio of capsule (MSR_C) was 12.23 mol NAP/mol TX100 at 0.1 g/l, which was much higher than the MSR_B for beads (0.70) and the MSR_W for the aqueous phase (0.11). Thus, when a nonionic surfactant is present in a hydrogel capsule, the ability of the surfactant to solubilize organic chemicals can be significantly increased compared to the surfactant in water or in hydrogel beads.

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1. Introduction

With the widespread occurrence of hazardous polycyclic aromatic hydrocarbons (PAHs) in environments due to global industrialization, intensive studies have been undertaken to better understand the mobility and fate of these contaminants in subsurface environments and to find methods for their remediation [1,2]. PAHs, which many countries consider priority contaminants to address, are carcinogenic and toxic in nature [3]. These contaminants can exist and accumulate in the air, in bodies of water, in soil and in food for long timescales due to their refractory nature and difficulties associated with their biological degradation [4]. PAHs are composed of only carbon and hydrogen, and most PAHs are hydrophobic with low aqueous solubility [5,6]. A potential technology for rapid removal of PAHs in soil is the washing of soil with a surfactant solution [7]. Due to their hydrophobicity, surfactants significantly enhance the solubility of these compounds [8].

The adsorption process is widely applied to PAH removal from wastewater with high removal efficiency [9]. The most commonly used adsorbent for the removal of PAHs from wastewater or groundwater is activated carbon. Although activated carbon is the most commonly used adsorbent for the removal of PAHs from water, it still has some limitations in its application for multiple rea-

sons, such as its relatively high cost, difficulty of regeneration, and reduction of adsorption capacity [10]. Recently, for the removal of PAHs from aqueous environments, considerable attention has been given to the development of adsorbents from naturally occurring materials, such as zeolite [11], talc [12], various nanoporous carbon adsorbents [13], carbon adsorbent from waste ion exchange resin [14], and agricultural byproducts, such as bean pods [15], and biochars [16].

Chitosan (CS) is a linear biopolymer of glucosamine [17], and this biomaterial exhibits a high adsorption capacity for dyes [18], metal ions [19], and other ionic compounds [20] because of its multiple functional groups (amine and hydroxyl groups). CS can normally form a hydrogel bead by gelation in an alkaline solution through neutralization. However, the density can be dramatically increased by using an anionic surfactant as the gelling chemical instead of alkaline conditions, with the resulting formation of a capsule that has a membrane-like shell with high gel density [18]. This shrunken gel structure has many unique but advantageous properties, such as high density, high strength, and different adsorption characteristics [18]. When this new gelation method is used, the resultant chitosan capsules (CC) show higher adsorption capacities for various dyes in solution compared to beads formed via normal alkali gelation [18].

Theoretically, CS hydrogel beads are not assumed to be a good alternative for the adsorption of hydrophobic compounds, due to the many ionic moieties present in CS. For this reason, there have

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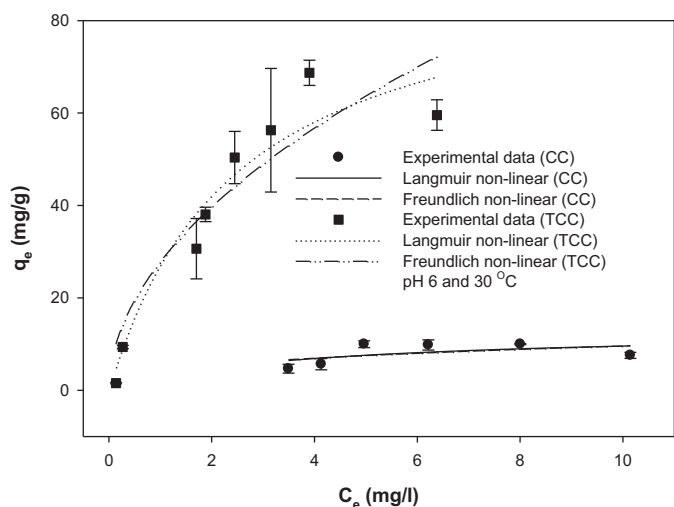


Fig. 1. Plots of q_e vs. C_e for the adsorption of NAP to CC and TCC (2 g/l TX100) prepared by 5 g/l SDS gelation at pH 6 and 30 °C.

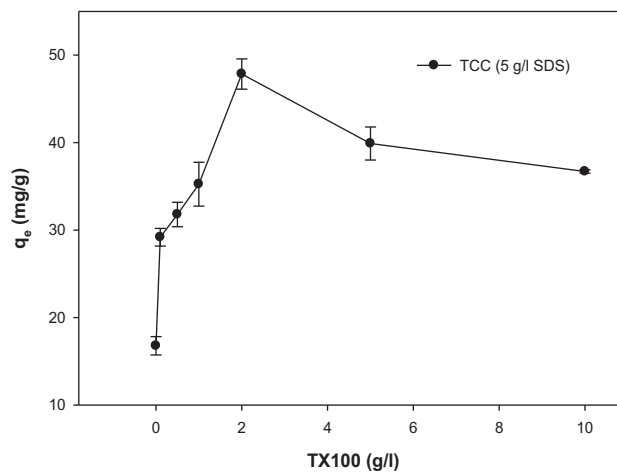
been no reports on PAH adsorption by CS-containing materials, except for a single report from our own previous study [21]. In that report, various types of surfactants (cationic, anionic, and nonionic) were impregnated in CS hydrogel beads (SICB) formed by alkali gelation to use as sorbents for the removal of naphthalene (NAP) from aqueous solutions [21]. CS beads (CB) that had not been impregnated with surfactant were not effective for NAP sorption, as expected, but the sorption capacity of beads (SICB) was enhanced after surfactant impregnation. More interestingly, the amount of sorbed NAP was much greater than that expected from the solubilizing capability of the surfactant, normally represented as the molar solubilization ratio (MSR). A new conceptual model was introduced in the study by considering impregnated surfactant in the beads as being in a separate phase, and its contribution was measured as a molar sorption ratio (MSR_B). The MSR_B of surfactants in SICBs were compared to intrinsic MSR_W values of surfactants within the same concentration range. It was found that the MSR_B for surfactants impregnated in the beads were higher than their MSR_W (e.g., 6.4–12.9 times higher) [21].

In this study, CS capsules (CC) formed by anionic surfactant gelation were used for the first time as a sorbent for the removal of NAP from aqueous solution, and surfactant-impregnated CC were investigated using Triton X-100 (TX100) as a model nonionic surfactant to impregnate capsules (TCC). The hydrogel capsules have a different volume structure compared to hydrogel beads that are completely filled inside. Therefore, a similar but different MSR term (MSR_C) was newly introduced. The true effectiveness of the surfactant for the removal of NAP was compared among surfactant present in capsules, hydrogel beads, or the liquid phase with different types of capsules formed by different gelling conditions and TX100 concentrations.

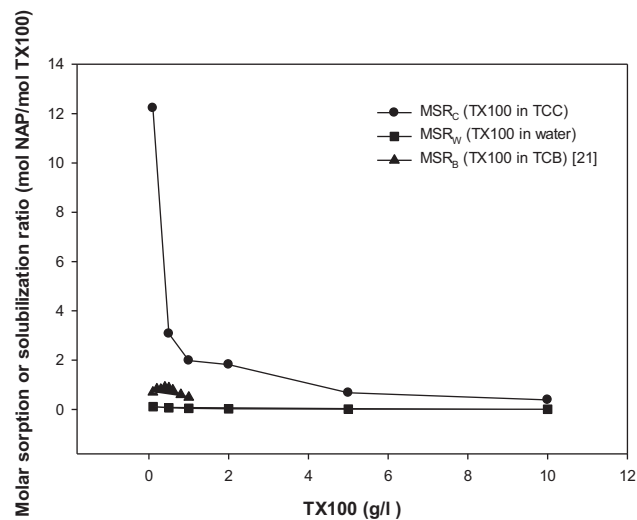
2. Materials and methods

2.1. Formation of hydrogel capsules

The formation of TCC involved multiple steps, including the addition of the desired amount TX100 solution from a stock solution of TX100 (10 w/v %) to a CS-acetic acid solution (1 w/v % CS in 2 v/v % acetic acid solution), followed by dropwise addition to sodium dodecyl sulfate (SDS) solution of the required concentration (5, 10, 20, and 50 g/l) that was used as anionic surfactant for the formation of capsules. TX100 concentrations were varied from 0 to 10.0 g/l in the 100-ml solution of CS. For the preparation of 20



(A)



(B)

Fig. 2. The effect of the concentration of impregnated TX100 in TCC on NAP sorption from aqueous solutions (A) and the molar sorption ratio (MSR_C) of TX100 in TCC (B). The MSR_W of TX100 in water for NAP was obtained from calculations using a partitioning model of a pure liquid system. The molar sorption ratio (MSR_B) of TX100 in TCB was obtained in ref [21].

capsules of any variety, 10 ml SDS solution of the desired concentration was used. Capsules formed in SDS solution were collected after 3 h, followed by repeated washing with deionized water to remove surfactant on capsule surface, and preserved in deionized water. The formation of CC followed the same method with TCC without the addition of TX100. Fig. S1 in the supplementary material of the manuscript depicts the formation of hydrogel capsules (TCC and CC). TX100 impregnated CS hydrogel beads formed by alkali gelation were designated as TCB, and in this case CS-TX100 solution was dropped into alkaline coagulating solution having composition of NaOH: water: methanol (1:4:5; w/w) to form TCB [21]. TX100 concentration was fixed at 1.0 g/l in the CS solution for TCB, and the CS hydrogel beads without TX100 impregnation was named as CB [21]. Thereby, anionic surfactant gelation method was applied to form CC and TCC [18], and CB and TCB were formed by alkali gelation method [21].

2.2. Batch sorption experiments

Batch sorption experiments using CC and TCC were performed to study the sorption of NAP from aqueous solutions. The batch sorption experiments were carried out by adding 0.2 g of wet CC

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