

Rapid removal of cationic dyes from water by coprecipitation with aluminum hydroxide and sodium dodecyl sulfate

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ABSTRACT

A coprecipitation method using sodium dodecyl sulfate (SDS) and aluminum hydroxide was designed for the efficient removal of three cationic dyes: methylene blue (MB), malachite green (MG), and crystal violet (CV), from water. The coprecipitation occurred by the hydrolysis of Al(III) ions in the presence of SDS far below the critical micelle concentration. Microscopic observation and spectrometric analyses clearly indicated the sorption of SDS into the precipitate. A molecular probe study using 1-phenyl-*N*-naphthylamine suggested that the precipitate provided hydrophobic regions suitable for incorporating hydrophobic ion-pairs of cationic dyes with dodecyl sulfate ions. In the pH region from 7.9 to 8.6, the use of 100 mg L⁻¹ each of SDS and Al(III) allowed nearly complete (>99%) removal of three cationic dyes that had hardly (<4%) been removed by the conventional coagulation method using aluminum hydroxide. The proposed method was successfully applied to the treatment of different wastewater samples containing dissolved organic substances and superior to the admicellar sorption method using SDS-coated aluminum hydroxide.

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Introduction

Methylene blue (MB), malachite green (MG), and crystal violet (CV) are popular cationic dyes used in a wide area of including textiles, rubbers, papers, and plastics. They have also been employed as biological stains for microscopic analysis of cells or tissues and as fungicides for aquariums or culture media. Because of the high toxicity to aquatic organisms and potential carcinogenicity to human being, these cationic dyes have to be eliminated from wastewater. Although a number of methods have been studied and successfully employed for the removal of synthetic dyes [1–8], rapid and complete removal of these cationic dyes from wastewaters still remains an important problem.

Recently, attention has been paid to surfactant-mediated separation techniques such as micellar enhanced ultrafiltration [9–11] and cloud point extraction [12–15]. Cationic dyes distributing into the surfactant micelles can readily be separated from water by the ultrafiltration of micelles or the temperature-induced phase separation of aqueous micellar solutions. However, time-consuming processes are required for the phase separation or ultrafiltration of viscous aqueous micellar solution. Estrogenic property of alkylphenyl-type nonionic surfactants and their

decomposed products [16] is another reason for eliminating the practical use of cloud point extraction. The admicellar sorption method using surfactant-coated alumina seems an attractive alternative, because of easy preparation of admicelles, and rapid removal of selected cationic dyes from water [17,18]. However, large amount (e.g. more than 10 g L⁻¹) of alumina was required for the sufficient removal of organic pollutants. The difficulty in mechanical shaking of the alumina suspension may be also reasons for hesitating to use alumina-based admicelles.

Previously, we designed a novel admicellar sorption method using surfactant-coated aluminum hydroxide and applied to the removal of hydrophobic organic pollutants [19]. The admicelles were formed by the adsorption of anionic surfactant on positively charged aluminum hydroxide that had been prepared by the hydrolysis of Al(III) ions in the neutral pH regions. Hydrophobic organic pollutants such as alkylphenols, polycyclic aromatic hydrocarbons, estrogens, chlorinated antifungals, and pesticides were collected to the surfactant-coated aluminum hydroxide and efficiently removed from water. In our preliminary study, the newly designed admicellar sorption method was also applicable to the removal of MB but the yield significantly decreased in the application to wastewaters containing dissolved organic substances.

In the present study, we examined the removal of cationic dyes from water by coprecipitation with aluminum hydroxide and anionic surfactant. Different from the admicellar sorption method, the surfactant sorption occurs during the formation of aluminum

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hydroxide by the hydrolysis of Al(III) ions (Fig. 1). Because of the combination of hydrophobic and electrostatic interactions with anionic surfactant, cationic dyes may be efficiently incorporated into the coprecipitate. Sodium dodecyl sulfate (SDS) was selected as an anionic surfactant because of the extensive use for admicellar sorption method [17,18,20–22] as well as its high biodegradability [23]. The effectiveness of the proposed method for dye removal and the factors influencing the yields were investigated in detail. The removal of MG was compared with the results obtained by using other sorbents such as bentonite, silica gel, zeolite, and activated carbons because of their extensive study or practical use [6,24–28]. The applicability to wastewater treatment was tested by using different wastewater samples and the results were compared with those of admicellar sorption method. The interference of fulvic acid and proteins was also studied, because they are possible organic substances omnipresent in wastewaters [29,30].

Materials and methods

Apparatus

Scanning electron microscope (SEM) images, transmission electron microscope (TEM) images, energy dispersive X-ray spectra (EDS), and X-ray diffraction (XRD) spectra were obtained by using a JEOL JEM-2500SE analytical transmission electron microscope (Akishima, Japan). A Jasco 4200 FT-IR spectrometer (Hachioji, Japan) was used for measuring the FT-IR spectra of the precipitate. Zeta-potential of aluminum hydroxide was measured with a Microtec Niton Zeecom ZC2000 zeta-potential analyzer (Funabashi, Japan). A PerkinElmer LS-50B luminescence spectrometer (Waltham, MA, USA) with a 1-cm quartz cell was used for the fluorescence probe study. An isocratic HPLC 2000 system (Hachioji, Japan) with an Inertsil ODS-3 column (length: 150 mm, inner diameter: 3.0 mm, GL Sciences, Tokyo, Japan) was used for the

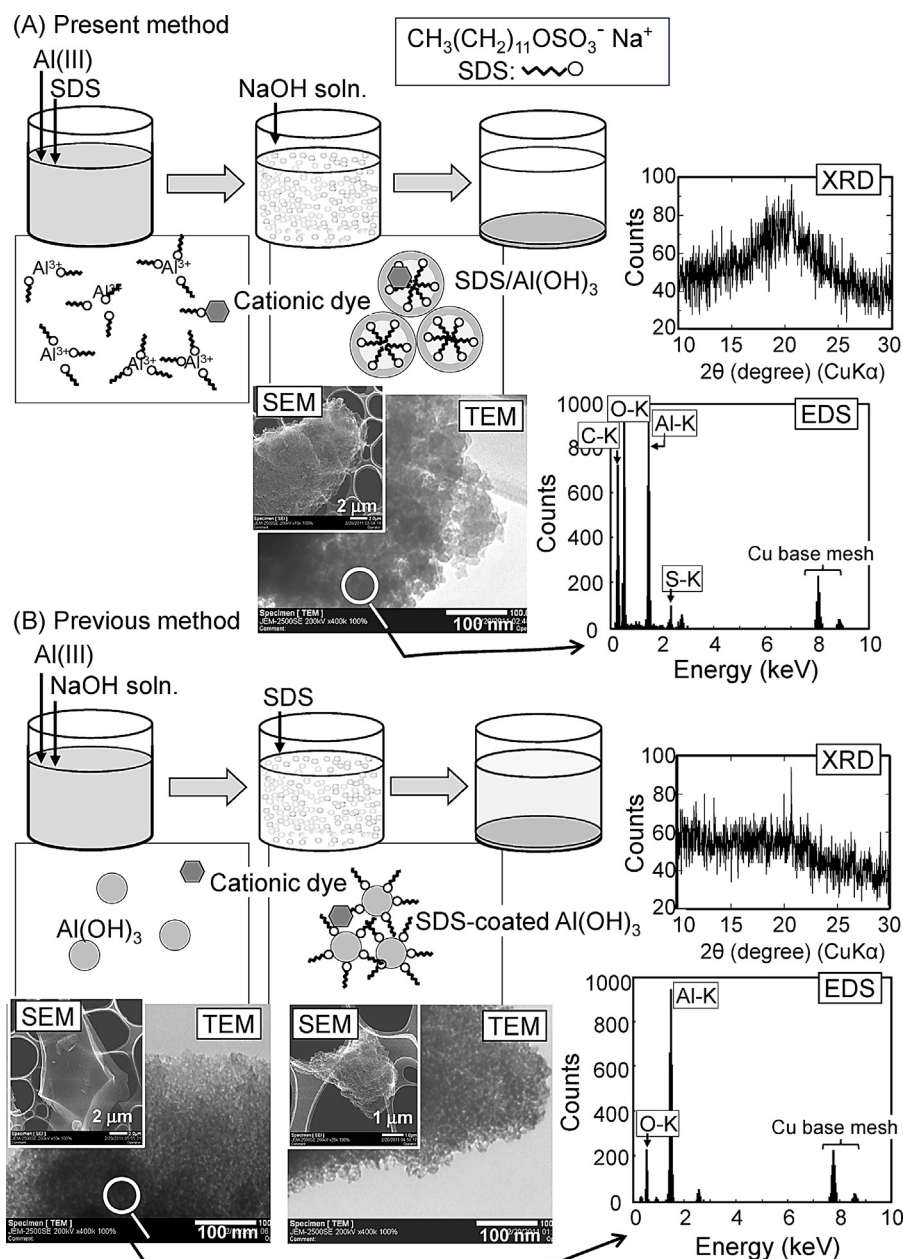


Fig. 1. Schematic procedures and assumed mechanisms of the present (coprecipitation, A) and previous (admicellar sorption, B) methods. SEM and TEM images of formed precipitate as well as EDS and XRD are also shown.

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