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Influence of stabilizing agents on the microstructure of Co-nanoparticles for removal of Congo red☆

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HIGHLIGHTS

- α-cobalt nanomaterials synthesized in presence and absence of stabilizers.
- Surfactants (CTAB) and polymer (PVA) has significant impacts on the morphology.
- The efficiency of cobalt nanomaterials improved in presence of $NaBH_4$ for the reduction of Congo red.

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ABSTRACT

Removal of Congo red is a serious environmental problem. Metal nanoparticles are emerging as a efficient catalysts and/or adsorbents to the degradation of dyes due to the large surface area. UV–visible absorption spectroscopy was used to quantify the decolorization of Congo red by Co-nanoparticles with and with out sun light at different time intervals. The experimental results show that the 100% Congo red (2.1×10^{-4} mol dm^{-3}) can be mineralized after ca. 1 min with Co-nanoparticles (3.2×10^{-3} mol dm^{-3}) in presence of sun light irradiation. The degradation pathway of Congo red under the optimal experimental conditions is also proposed and discussed. In this paper, a simple and nonexpensive method was used to the synthesis of Co-nanoparticles in absence and presence of stabilizers. The morphology, stability and color of cobalt sols strongly depends on the nature and/or presence of stabilizers. Transmission electron microscopic data revealed that the Co-nanoparticles possesses multi-layered aggregated sheet, and dumbly shaped morphology with different stabilizers.

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1. Introduction

Molecular structure of dyes contained mutagenic and carcinogenic aromatic coal tar-based hydrocarbon(s). Industrial wastes contained dyestuffs and other coloring materials, which can be mixed with surface water and then they may bring a chief threat to human health. Therefore, it is necessary to prepare a suitable adsorbent for the removal of the dye pollutions from our water resources. Synthesis, and structural evaluation of cost effective, environmental friendly and higher efficient advanced nanomaterials for the removal of toxic industrial wastes, especially organic dyestuffs have been the subject of

☆ Capsule: Cobalt nanoparticles synthesized with and without stabilizers, and used as a adsorbent to the removal of Congo red in absence and presence of sun light.

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various investigations (Chiou et al., 2004; Gong et al., 2005; Mittal and Gupta, 2009; Cheng et al., 2011; Gupta et al., 2013). Size-dependent reactivity and large surface area of nanomaterials lead to their use as efficient catalysts (Pal et al., 1998). The methods of preparations, and presence of stabilizers have significant impact on the morphology (nanocones, nanoplates, nanodiscs, nanorods, nanosheets, nanoneedles, nanowires, nanoflake, and butterfly-like structures) of cobalt nanomaterials (Jeevanandam et al., 2000; Hosono et al., 2005; Yang et al., 2010; Yan et al., 2011; Wang et al., 2011b) which also have potential applications in different technologies, such as catalysts (Meyn et al., 1990), adsorbents (Nedez et al., 1996), composite materials (Xue et al., 2003), ceramics (Philipse et al., 1994), rechargeable batteries (Faure et al., 1991), gas sensing (Frost and Wain, 2008), ionic exchangers (Liu et al., 2006), magnetic materials (Zhang et al., 2008). It has been established that cobalt hydroxides exist in α-, and β- polymorphic forms (hydrotalcite-, and brucite-like morphologies, respectively) and have a hexagonal layered structure (Liu et al, 2005; Xu and Zeng, 1999; Kamat et al., 1997; Zhu et al., 2002; Kobayashi et al., 2003; Sahiner et al., 2010).

The chemical literature contains abundant reports aimed towards understanding the role of stabilizers in the synthesis and characterization of CONPs and/or its alloy with and with out doped ($CoFe_2O_4$, $CoFe_{2-x}Gd_xO_4$, and $Co_xCu_{1-X}TiO_3$) having different morphologies under various experimental conditions (Pouretedal et al., 2010; Liang and Zhao, 2012; Zhao et al., 2014; Hashemian and Foroghimoqhadam, 2014; Ding et al., 2015). These investigators also used Co-nonmaterials for the degradation of congo red with and without sunlight irradiations. The polyhedral Cu_2O nanoparticles has been used for the adsorption removal of Congo red from aqueous solution (Shu et al., 2015).

Tri cobalt tetraoxide nanocubes have been prepared by a simple hydrothermal reaction under external magnetic fields (Wang et al., 2011a). Synthesis of uniform cobalt nanoparticles by the reduction of $CoCl_2$ with $NaBH_4$ inside the reverse micelles of didodecyldimethylammonium bromide has been reported (Chen et al., 1994). Synthesis of trioctylphosphine-coated 2D superlattices of magnetic cobalt nanomaterials was discussed (Petit et al., 1998, 1999). They used stoichiometric ratio (1:2) of cobalt bis(2-ethylhexyl)sulfosuccinate) and $NaBH_4$in two micellar solution of same surfactant having the 0.25 mol dm^{-3} diameter, sodium bis(2-ethylhexyl) sulfosuccinate and discussed their self-organization predisposition, which converted nanoparticles into 2D superlattices. Generally, Co^{2+} - $NaBH_4$ redox system with and without stabilizers (organic solvents, mixture of surfactants, silica) was used to the synthesis of stable cobalt nanoparticles (Kobayashi et al., 2003; Sahiner et al., 2010) . However, the published articles on the effect of individual surfactant and PVA on the nucleation and growth of cobalt nanoparticles are rather limited.

It has been established that the morphology of the nanomaterials can be easily controlled by using different kinds of stabilizers, such as surfactants, polymers, proteins, phospholipids, etc. (Bakshi, 2016, 2011). In this work, Co^{2+}-$NaBH_4$ redox system was used to the preparation of CoNPs in presence of two stabilizers, namely, CTAB, and PVA. The degradation of congo red (first synthetic anionic diazo dye, which is capable of dying cotton directly. It is prepared by coupling tetrazotised benzidine with two molecules of napthionic acid. Congo red containing effluents are generated from textiles, printing and dyeing, paper, rubber and plastics industries. Due to its structural stability, it is difficult to biodegrade) was preformed in presence of CTAB-capped CoNPs under $NaBH_4$ with and without sun light.

2. Experimental

2.1. Materials

Double-distilled deionized water was used as a solvent to the preparation and dilution of all reagent solutions. Cobalt nitrate ($Co(NO_3)_2$; oxident), sodium borohydride ($NaBH_4$; reductant), stabilizers (cetyltrimethylammonium bromide, $C_{19}H_{42}BrN$ and poly(vinyl)alcohol, 99%–100% hydrolyzed), and Congo red ($C_{32}H_{22}N_6Na_2O_6S_2$) were used as received from Merck India products (purity $\geq$ 99%). Stock PVA solutions were prepared by slow stepwise addition of PVA to solvent, water, whilst rapidly stirring to avoid their aggregation. Due to the instability and/or hydrolysis of $NaBH_4$ in water, its aqueous solution contains certain amount of NaOH (Eq. (1)). Therefore, freshly prepared solutions were used to the synthesis of CoNPs (Cloutier et al., 2007).

$$NaBH_4 + 4H_2O \longrightarrow NaOH + H_3BO_3 + 4H_2\uparrow \quad (1)$$

2.2. Preparation of CoNPs

In a typical experiment, the required $NaBH_4$ solution was added into the reaction mixture containing $Co(NO_3)_2$ and stabilizer (if necessary). The appearance of gray turbidity, blue color, and light green color, indicating the formation of CoNPs having different morphologies, and light pink color of Co^{2+} ions has been disappeared completely (Guella et al., 2006). The as prepared CoNPs were collected with a magnet and washed them with deionized water and ethanol several times. The transparent sols were centrifuged (10000 rpm for 30 min). Aqueous solutions were decanted from the centrifuge tubes, and CoNPs were dispersed in water, filtered and washed three times, and dried under vacuum for 3 to 4 h. The formation of

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