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Sustainable super fast adsorptive removal of Congo red dye from water by a novel technique based on microwave-enforced sorption process

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ABSTRACT

Removal of Congo red was completed in 5–20 s using microwave-enforced sorption (MES) as a sustainable technique. Nano-γ-Al_2O_3 was covalently bonded with 3-chloropropyltrimethoxysilane to produce γ-Al_2O_3-SiCl nanosorbent via microwave-assisted approach. Congo red extraction by γ-Al_2O_3 and γ-Al_2O_3-SiCl were found 58.42% and 69.41% and increased to 66.13% and 81.47%, respectively using 5 and 20 s of microwave heating. The percentage extraction of Congo red (30 mg L^{-1}) by γ-Al_2O_3-SiCl was increased from 46.21% to 83.61% upon increasing the pH from 1.0 to 10.0. Other controlling conditions were investigated and optimized. Excellent recovery of Congo red from wastewater was established in 20 s.

Introduction

One of the most important classes of organic pollutants is mainly based on dyes and their derivatives. These pollutants are well characterized by their high toxicity as well as coloring of wastewater to represent well documented environmental problems [1–10]. Dyes are aromatic compounds with two groups viz, chromophores and auxochrome which are responsible for color and intensity of color, respectively [11–18]. According to the color index (C.I.), more than 10,000 types of synthesized dyes are commonly known. Dyes can be classified according to the application methods where three categories of dyes are categorized according to their dissolution in aqueous medium including anionic in the form of direct, acid and reactive dyes with completely negative charge due to (SO_3^{-3}) groups, cationic in the form of basic dyes with positive charge due to the presence of protonated amine group and nonionic in the form of disperse dyes [19,20].

To minimize their environmental impact and pollution, a wide range of methods has been implemented for wastewater treatment and removal of dyes. These technologies are mainly based on physicochemical methods such as adsorption using different adsorbents, coagulation and precipitation, membrane filtration, photocatalysis, oxidation processes, electrochemical oxidation, decomposition by microbiological or enzymatic components [21–26]. Congo red is an aromatic reactive dye characterized by a complex chemical structure due to the presence of various reactive center and functional groups. Congo red dye is also known to exhibit high solubility in aqueous solutions and therefore, it is widely used in textiles, printing and dying process, paper, rubber and plastics industries [27–29]. As a result of these industrial activities, contaminated industrial wastewater with dyes such as Congo red must be removed before discharge into the environment where various removal techniques of Congo red have been successfully developed. For example, nanoparticles of $CoFe_{2-x}Gd_xO_4$ (x = 0, 0.03, 0.05, 0.07, 0.1) were prepared by hydrothermal method and used to enhance the adsorption capability for Congo red from aqueous solution [30]. Removal of Congo red dye from aqueous solution using natural coagulants such as Surjana seed, Maize seed and chitosan was reported [31]. Bamboo sawdust was treated to produce twelve hydrochars and studied for adsorption of Congo red and 2-naphthol [32]. Mg–Al-layered double hydroxide was prepared using coprecipitation method of Mg^{2+}/Al^{3+} with molar ratio of 2 at constant pH 9 and the adsorbent was examined for removal of Congo red from aqueous solutions [33]. Cetyltrimethylammonium bromide and sodium dodecyl sulfate (SDS) were used with Ag-nanoparticles (Ag-NPs) as catalysts for removal of Congo red from industrial wastewater [34].

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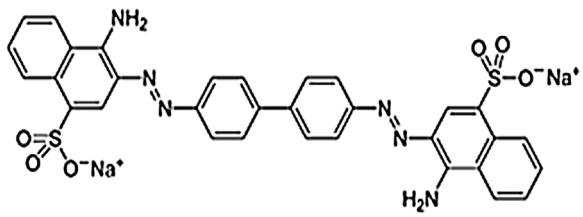
Radiation or photo induced degradation technique is another well established approach to treat and remove industrial wastewater containing dyes [35]. The photo induced decoloration of some dyes using a hybrid catalyst V_2O_5/TiO_2 was investigated under different operational conditions [36]. ZnO nanoparticles were prepared using different solvents and the photocatalytic activities of these materials were studied and reported for removal of Congo red from aqueous solution under solar radiation [37]. A study was performed to evaluate the sunlight degradation of Congo red using sono-intercalation of CdS nanoparticles into titanate layers [38]. The photocatalytic oxidation of Congo red under visible light irradiation in presence of $Bi_{3.84}W_{0.16}O_{6.24}$ nanoparticles was reported [39]. A facile integrated biological-cum-photo-assisted electro-oxidation process for degradation of Congo red in textile wastewater was investigated and reported [40].

The present study reports a novel super fast methodology to remove Congo red, as an example of reactive dyes from aqueous solution and real water samples. The proposed method is mainly based on the implementation of microwave-enforced sorption (MES) technique for extraction and removal of Congo red from aqueous solutions and real water samples. This technique is suggested to operate by migration of the adsorbate from solution and adsorption on the sorbent surface in few seconds under the influence of microwave heating irradiation to establish an equilibrium condition [41,42]. This technique is working in the reverse direction of the microwave-assisted extraction (MAE) process [43]. A microwave-assisted synthesis approach was used to synthesize and covalent bonding of 3-chloropropyltrimethoxysilane on the surface of γ-Al_2O_3 nano particles to produce γ-Al_2O_3-SiCl nanosorbent. The designed nanosorbents were characterized by the FT-IR, SEM, TEM, TGA and XRD. The novelty in this work is mainly focused on the implementation of a super fast removal process of Congo red dye from aqueous solutions and real water samples using a novel microwave-enforced sorption (MES) technique. The MES-technique of Congo red dye has been additionally explored, confirmed and optimized under different controlling conditions such as pH of contact solution, microwave heating time, nanosorbent dose, initial dye concentration, and interfering ions.

Experimental

Instrumentations

Absorption spectroscopic measurements were made using a Unico UV–Vis-7200 spectrophotometer in a 1 cm cell. A pH meter-Adwa was calibrated by standard buffer solutions 4.01, 7.00 and 9.21 and used to measure the pH value of dye solutions. Surface area (BET) of the nanosorbents were made by using Nova 3200 Nitrogen Physisortion Apparatus, USA. To image the nanosorbents, high resolution transmission electron microscopy (HR-TEM) model JEOL-JEM 2100, Japan was used to acquire the images at 200 kV. Fourier transform infrared (FT-IR) spectra of nanosorbents were recorded using Bruker Vertex70. Scanning electron microscope (JSM-6360LA, JEOL Ltd.)-(JSM-5300, JEOL Ltd.) and an ion sputtering coating device (JEOL-JFC-1100E) were employed to examine the images and particle size of the nanosorbents. Thermal gravimetric analysis (TGA) and thermoanalytical curves were collected by PerkinElmer TGA7 thermo balance. The selected operating conditions were a temperature heating range at 10–600 °C, a heating rate is 10 °C min^{-1}, a flow rate is 20 mL min^{-1} and the sample mass was taken in the range of 5.0–6.0 mg. The X-ray diffraction (XRD) analysis was investigated by Adrukru D8 ADUANCE, X-ray diffractometer. The microwave apparatus is a household oven (Smart, Model: SM-3025MS) and operates with 900-W output power at a frequency of 2.45 GHz.



Scheme 1. Structure of Congo red dye.

Chemicals

Aluminum nitrate nonahydrate ($Al_2(NO_3)_3{\cdot}9H_2O$, FW 375.13 and assay 98.0%), ethandiol (ethylene glycol), ($C_2H_6O_2$, FW 62.07 and assay 99.0%), oleic acid ($C_{17}H_{33}COOH$, FW 282.47 and assay 65–70%) were all purchased from oxford, India. The chemical structure of Congo red ($C_{32}H_{22}N_6Na_2O_6S_2$, FW 696.65) is shown in Scheme 1, purchased from oxford, India and used without further purification. 3-Chloropropyltrimethoxysilane ($Cl(CH_3)_3Si(OCH_3)_3$, FW 198.72 and assay 97.0%) was purchased from Aldrich Chemical Company, Inc., USA.

Synthesis of γ-Al_2O_3 nanoparticles [44]

A 0.0974 mol (36.55 g) of aluminum nitrate nonahydrate ($Al(NO_3)_3.9H_2O$) was added to 0.0974 mol (27.51 g) of oleic acid ($C_{17}H_{33}COOH$) and 120 mL of ethylene glycol in a beaker and stirred at room temperature to complete dissolution of the solid materials. The reaction mixture was then stirred for two more hours using magnetic stirrer, left inside an oven at 180 °C for five hours and allowed to cool at room temperature. The produced yellow precipitate was filtered, washed several time with methanol (CH_3OH). The yellow product was then dried in oven at 80 °C and calcinated at 900 °C for two hours to obtain the γ-Al_2O_3 nanoparticles.

Microwave-assisted synthesis of γ-Al_2O_3-SiCl nanosorbent

Typically, 0.0784 mol (8.0 g) of γ-Al_2O_3 nanoparticles were allowed to react with 16.0 mL of 3-chloropropyltrimethoxysilane in a microwave oven for 15 min and allowed to cool at room temperature. The produced nanosorbent, γ-Al_2O_3-SiCl was filtered, washed with diethylether and dried in an oven at 60 °C for 6 h.

Sorption of Congo red dye by nanosorbents

A stock solution (1000 mg L^{-1}) of Congo red dye was prepared in distilled water and further diluted to obtain the desired concentrations of dye (30, 50, 70 mg L^{-1}). The full curve of Congo red was measured using Unico UV–Vis-7200 spectrophotometer to identify λ_{max} of absorption. A calibration curve for Congo red dye was also established as shown in Fig. 1.

The sorption process of Congo red dye by γ-Al_2O_3 and γ-Al_2O_3-SiCl nanosorbents from aqueous solutions was studied by the MES-technique in presence of different experimental conditions. The effect of pH on color removal of Congo red dye using γ-Al_2O_3 and γ-Al_2O_3-SiCl nanoparticles was studied by using 20 mL of dye solutions (30, 50 and 70 mg L^{-1}) already adjusted to the pH 1.0–12.0 using 1.0 mol L^{-1} of either HCl or NaOH. The MES of Congo red dye using γ-Al_2O_3 and γ-Al_2O_3-SiCl nanoparticles was performed by addition of 20 ± 1 mg of nanosorbent to the dye solutions and heating this mixture in the microwave oven for 20 s. The solid material was then filtered and the remaining dye was detected by measuring the absorbance at λ_{max} = 490 nm.

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