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Regular Article

Untapped conversion of plastic waste char into carbon-metal LDOs for the adsorption of Congo red

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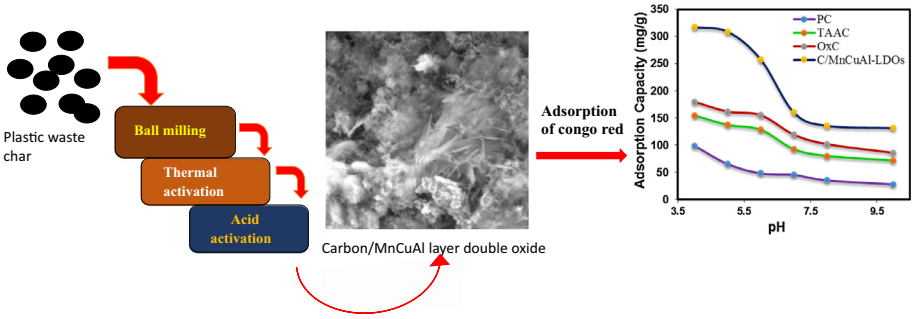
GRAPHICAL ABSTRACT



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ABSTRACT

A low-cost novel carbon-metal double layered oxides (C/MnCuAl-LDOs) nano-adsorbent was synthesized by co-precipitation, for the adsorption of Congo red (CR), using modified carbon derived from pyrolysis of polystyrene (PS) plastic waste. The synthesized C/MnCuAl-LDOs has a crystalline structure with a high surface area of 60.43 m^2/g and pore size of 99.85 Å. Adsorption of CR using all prepared adsorbents from aqueous solution under equilibrium and kinetic conditions were evaluated against different values of the pH (4–10), initial CR concentrations (25–250 mg/g), contact time (0–310 min) and temperature (30–50 °C). The obtained results revealed that C/MnCuAl-LDOs showed maximum adsorption capacity for CR among all the used adsorbents. The optimum equilibrium time was 180 min, whereas acidic medium (pH 4.5) favored the maximum adsorption of CR up to 317.2 mg/g on C/MnCuAl-LDOs. The adsorption kinetics followed the pseudo-second-order model, whereas Freundlich adsorption isotherm fitted best to obtained data in comparison to Langmuir adsorption isotherm. The results suggested that C/MnCuAl-LDOs is an efficient material for the removal of organic pollutants from the wastewater.

1. Introduction

Water pollution has become one the major threat to the environment and human being since last few decades [1]. Industrial expansion has increased the concerns of water pollution due to a massive release of organic and inorganic pollutants to the water bodies [2]. As a consequence, the synthetic dyes such as Congo red (CR) is found in the aquatic system that is one of the highly carcinogenic and non-biodegradable organic pollutant [3]. CR is easily metabolized to benzidine that causes carcinogenic effects to human beings [2]. In addition, CR causes vomiting, difficulties in breathing, nausea, and diarrhea [4].

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Various methods have been used such as membrane separation [1], photocatalysis [5,6], electrochemical method [7], adsorption [8] for removal of CR. However, adsorption is considered as a promising method due to its simplicity, easy to handle, effectiveness and economically feasible [4]. The performance of the adsorbent can be increased by using a material with low cost, non-toxicity, easily available, smaller particle size and high BET surface area [2]. Layered double hydroxides (LDHs) have gained significant attention in recent years due to their excellent adsorption capacities and potential applications, including ion exchange host, catalysts and decolorizing agent [4].

LDHs have a hierarchical structure that makes them suitable for treating the wastewater [9,10]. The LDH can be represented by the simple formula: $M_a^{2+}M_b^{3+}(OH)_{2a+2b-}(X^-)\cdot xH_2O$, where M^{2+} may be referred to Fe^{2+}, Mg^{2+}, Co^{2+}, Zn^{2+}, Ni^{2+} and M^{3+} is Cr^{3+}, Fe^{3+}, Al^{3+} [11,12]. In addition, it is known as hydrotalcite anionic clay materials that are rare in nature and can be synthesized [9,13]. It is composed of hydroxide layer with positive charge and anions together with H_2O molecules present between the layers, where interlayer anions can be exchanged [4]. LDHs can be synthesized by following various routes, including calcination rehydration, precipitation, mechanochemical approach, the sol-gel method, and induced hydrolysis method [14,15]. LDH can be easily transformed into the corresponding metal oxides by simply heating at a certain temperature [9]. The unique physiochemical properties and various application made the mixed oxides an important class of material [16]. A well-known potential field for such oxides is adsorption [17].

Various researchers have reported that calcined LDHs have high adsorption capacity as compared to un-calcined LDHs [16]. In addition, calcination of LDHs may increase its BET surface area, dispersion of M2+ and M3+ and synergetic effects between the elements [18,19]. Previously, many researchers reported the potential of mixed metal oxides such as ZnAl–CO3 [20], Mg-Al [21], biomorphic Mg-Al [16]. However, the activation and modification of char produced from pyrolysis of polystyrene (PS) plastic waste for the synthesis of carbon-metallayered double oxides (LDOs) and the adsorption of pollutants from wastewater are not published previously in the scientific community, which is the focus of this study.

This study aims to optimize the conversion of PS plastic waste char into carbon-metalLDOs for the adsorption of CR. The char was produced from pyrolysis of PS plastic waste at 450 °C and 75 min retention time using a small pilot scale reactor [22]. The obtained char was pretreated thermally (550 °C) and chemically (H_2SO_4 and HNO_3) and then it's composite with metal layered double oxides (C/MnCuAl-LDOs) was prepared. The batch experiments were carried out to compare the CR adsorption capacity of pure carbon (PC), thermally activated carbon (TAAC), oxidized carbon (Ox-C) and C/MnCuAl-LDOs. Furthermore, the kinetic and adsorption isotherms of CR on these adsorbents were examined.

2. Material and methods

2.1. Materials and preparation of adsorbents

$MnSO_4{\cdot}4H_2O$, $CuSO_4{\cdot}5H_2O$ and $Al(NO_3).9H_2O$ were purchased from BDH Chemical Ltd, Poole, England and Hopkin and Williams Private Ltd, England, respectively. Urea, HNO_3, and H_2SO_4 were purchased from Panreac Co. Ltd. The Congo red (CR) was purchased from Techno Pharmchem Haryana, India. A 1000 mg/L stock solution of CR ($C_{32}H_{22}N_6Na_2O_6S_2$) was prepared by dissolving 1 g of CR in 1000 ml of deionized water.

Thermal pyrolysis of PS plastic was carried out in a small-scale pilot reactor in the absence of oxygen. The experiment was conducted at the temperature of 450 °C with 75 min of retention time [22]. The main product of pyrolysis process was liquid oil, whereas char and gases were the by-products. The char was crushed in ball milling machine for 3 h with a frequency of 20 Hz/sec. The obtained nano char was washed with deionized water and acetone and dried in an oven at 105 °C for 12 h. The dried char was thermally activated in a muffle furnace at 550 °C for 3 h at the heating rate 5 °C/min. Thereafter, the chemical activation of char (0.5 g) was performed using 20 ml of each H_2SO_4 and HNO_3 while sonicatsonicatored for 1 h followed by 5 h on a magnetic stirrer. The chemical activated char was centrifuged and washed with deionized water for several times until normal pH was obtained and dried in an oven at 105 °C for 24 h.

A solution was prepared by dissolving 0.082 g $MnSO_4{\cdot}4H_2O$, 0.078 g $CuSO_4{\cdot}5H_2O$, and 2.6 g $Al(NO_3).9H_2O$ in 150 ml of deionized water in round bottom flask. A 1.35 g of activated char was added to the solution, while later on 12 g of urea was mixed with solution and flask was reflux at 130 °C for 10 h. The prepared sample was centrifuged, thoroughly washed several times with deionized water and finally dried in an oven at 80 °C. The final product was calcined in the muffle furnace at 450 °C for 3 h at a heating rate of 5 °C/min to achieve the final product; the C/MnCuAl-LDOs composite.

2.2. Characterization of the materials

The XRD pattern of materials was recorded on Siemens D5005 X-ray diffractometer system with Cu Kαradiation (λ = 1.5406 Å). A Gemini 1525 FEGSEM was used to study the morphology of the pure and nanocomposite materials. In addition, to find out the presence of different functional groups on the surface of adsorbents, FT-IR spectra were recorded on FT-IR, Perkin Elmer's, UK. A Micromeritics Tristar 3000 was used for nitrogen adsorption and desorption. The Brunauer-Emmett-Teller (BET) was used for the measurement of surface area, whereas pore size distribution was examined by following the Barrett-Joyner-Halanda (BJH) method. X-ray photoelectron spectroscopy (XPS) was used to examine surface composition using SPECS GmbH, spectrometer, Germany (step size = 1 eV, base pressure = 10^{-10} mbar, X-ray = non-monochromatic Mg-Kα 1253.6 eV; 100 W; 13.5 kV).

2.3. Adsorption studies

The batch experiment was conducted in a 100 ml conical flask to observe the adsorption potential of CR on prepared materials in a water bath shaker (Julabo SW22). The experimental solution was obtained by diluting the stock solution to get the desired concentration solution. In each experiment, 0.02 g of adsorbent was mixed with 50 ml of CR solution of known initial dye concentration at fixed solution pH, temperature and time under continuous agitation at 200 rpm. After equilibrium, the solution was filtered with 0.22 μm filter membrane. The experiment was carried out at various pH ranges from 4 to 10 to examine the effect of pH, whereas 0.1 M HCl and NaOH solution were used to adjust the desired solution pH. The remaining concentration of CR in solution after adsorption was measured at 495 nm using UV–visible spectrophotometer (DR-6000, LANGE Germany) and following equation was used to determine the adsorption capacity (q) of the adsorbent.

$$q = (C_0 - C_f)V/m \quad (1)$$

where q is the adsorption capacity (mg/g), C_0 and C_f are the CR (mg/l) initial and equilibrium concentration respectively. Whereas m is the mass of adsorbent (g) and V is the volume of dye solution (L).

2.4. Adsorption kinetics and isotherm

In the present study two most frequently used kinetic models were selected namely pseudo-first and pseudo-second-order to