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FULL LENGTH ARTICLE

Adsorption separation of condensate oil from produced water using ACTF prepared of oil palm leaves by batch and fixed bed techniques ☆

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ABSTRACT

A novel method is applied to produce amorphous carbon thin film (ACTF) from oil palm leaves. The novel prepared ACTF is in the form of thin films like graphene sheets having winding surface. ACTF was characterized by different methods of characterization: FTIR, BET, SEM, EDX, TEM, and Raman. ACTF employed as an adsorbent to separate emulsified condensate oil from synthetic produced water as a treatment process before reinjection in oil reservoirs. The adsorption performance of batch and fixed bed adsorption systems were investigated. Contact time, initial concentration of condensate oil (C_o = 100–2500 mg/l) and temperature were studied by batch experiments. The obtained results indicated that the adsorption capacity and the removal efficiency increased with time up to 132.77 mg condensate/g adsorbent and 66.38% respectively, within 6 h equilibrium time at 308 K. The thermodynamic adsorption experiments conducted at 288, 308 and 318 K, referring exothermic nature of the adsorption process.

The performance study of fixed bed adsorption described through the breakthrough curves concept with two parameters: column bed heights (5, 10 and 15 mm) and flow rate (2.2, 5 and 8.4 ml/min). Two models (Thomas and Yoon-Nelson models) were applied to expect different parameters of fixed bed as adsorption capacity and time need for 50% breakthrough. The results exhibited that 2.2 ml /min feed flowrate and 5 mm bed height at 1000 mg/l initial oil condensate concentration were the optimum conditions for the ACTF column. The experimental breakthrough curves showed acceptable fit with the calculated breakthrough profiles obtained by Thomas model.

1. Introduction

Raw produced water is commonly regarded as a high-volume toxic waste but can be beneficial to humans if properly managed. The treatment of produced water is very important due to legislation and environmental concerns. All along more stringent environmental regulations require varied produced water treatment from oil and gas productions before discharge and before injection into reservoirs to reduce formation damage [1]. Currently, properly treated water can be recycled and used for water flooding (produced water re-injection (PWRI)) [2,3]. These beneficial reuses directly decrease the withdrawal of potable water, a highly valuable commodity in many regions of the world [4–8]. The general objectives for operators treating produced water are: de-oiling (removal of dispersed oil and dissolved grease), desalination, removal of suspended particles and sand, removal of soluble organics, removal of dissolved gases, removal of naturally occurring radioactive materials (NORM), disinfection and softening (to remove excess water hardness) [9]. To meet up with these objectives, operators have applied many standalone and combined physical, biological and chemical treatment processes for produced water management [10]. Among several chemical and physical methods, adsorption process is one of the effective methods widely used in wastewater systems.

A wide variety of agricultural by-products and wastes has been investigated as cellulosic precursors for the production of activated carbon [11]. These precursors include coconut shell and wood [12], Olive stones [13,14], sugarcane bagasse [15], pecan shells [16,17], palm seed [18], apple pulp [19], rubber seed and palm seed coat [20], and molasses. [21]. Recently, activated sludge has been

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produced as a result of wastewater treatment activities and has emerged as an interesting option for the production of activated carbon [22–25].

Adsorption onto has been found to be better than other methods of wastewater treatment because of its capability for adsorbing a broad range of different types of adsorbents efficiently, and its simplicity of design [26–28]. Maghrabi, et al., [29] prepared mesoporous silica (MCM-41) for oil adsorption from produced water injected in water injection projects.

Amorphous carbon thin film (ACTF) is a novel form of carbon materials which consists of graphitic-like monolayer of carbon. Fathy et al., [30] synthesized Amorphous Carbon Thin Film (ACTF) from Rice Straw and studied its structure and properties as new adsorbents of sodium ions from synthetic water. Also Fathy et al., [31] synthesized waste polystyrene/amorphous carbon thin film composite resin (WPS/ACTF) beads by grinding and mixing the unused waste polystyrene bottles or others with carbon nanostructure. According to El-Sayed et al., [32] amorphous carbon thin film prepared from wood sawdust was effectively utilized for oil removal with high adsorptive capacity.

The first aim of this work is to prepare and characterize a new carbon material which is amorphous carbon thin film (ACTF) using a suitable agricultural wastes (oil palm leaves). The second aim is to investigate the adsorption behavior of ACTF in removing condensate oil from synthetic produced water through batch adsorption and a fixed bed column technique.

2. Material and experimental

2.1. Preparation of amorphous carbon thin film (ACTF)

The following described method of preparing amorphous carbon thin film is a novel one developed by Fathy et al., [30]. The method goes through four phases. Firstly the Oil palm leaves (OPL) leaves washed with hot distilled water, dried overnight at 60 °C, grinded to sizes (0.07–0.1 mm) and stored in a tight bottle until used. Second phase is the chemical exfoliation of cellulose. Hence, the dried precursor was hydrolyzed for 60 min at 120 °C, using 1% (wt/wt) sulphuric acid. 5 g of precursor were added in the presence of 0.1 g silica to 5 ml concentrated sulfuric acid, stirred for 10 min, filtrated and washed with hot deionized water till pH 7. The following phase is the drying at 40 °C for 6 h. Finally the carbonization process is accomplished, where the semi carbonized sample mixed with 0.01 g prepared cobalt silicate nanoparticle [33], heated up to 40 °C for 30 min. Finally the prepared amorphous carbon thin film (ACTF) left to cool for 1 h, then dried in a vacuum oven for 24 h at 50–70 °C [34].

2.2. Preparation of synthetic oil/water emulsions

Samples of oil/produced water emulsions were prepared using Egyptian condensate oil (of density 0.7928 mg/l) and synthesized produced water. The synthetic produced water made up based on the chemical characteristics of natural field samples collected from Egyptian oilfields (Chemical composition: NaCl, 53.4 g; KCl,1.196 g; $MgCl_2$, 9.69 g; Na_2SO_4, 10.44 g; $CaCl_2$, 5.09 g and $NaHCO_3$, 0.306 g in 1000 ml of deionized water).

Stock solution of oil/water was prepared by mixing 1 g of condensate oil with 1000 mL of synthetic produced water and 0.1% of an anionic surfactant (Sodium dodycle sulfate) as an emulsifier. The mixture was then stabilized speed of 500 rpm for 60 min. It was later diluted to different concentrations. Synthetic oil/water samples was prepared at desired concentrations. The prepared ACTF shows high ability to disperse in both polar and non-polar solvents which noticed by a simple visual testing. Fig. 1 is an image shows stable aqueous and non-aqueous emulsions of ACTF in different types of solvents (from left to right: water, Hexane, Ethyl alcohol; n-Heptane, and Propanol). That reflects the wide potential of ACTF applications.

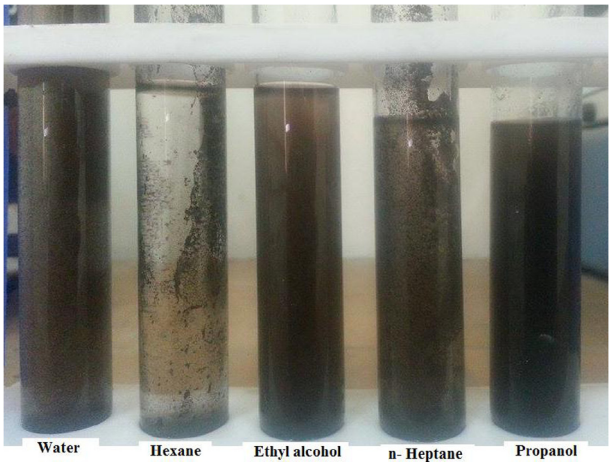

Fig. 1. ACTF emulsions stabilized without additives in different solvents (from left to right water, hexane, Ethyl alcohol, n-heptane and propanol).

2.3. Characterization of prepared amorphous carbon thin film (ACTF)

The prepared amorphous carbon thin film (ACTF) was characterized by many techniques in order to investigate its physical and chemical properties related to the adsorption behavior. BET technique was applied to determine total pore volume and pore size distribution by N_2 adsorption at 77°K using an Autosorb (Quantachrome Corp., USA Nova 3200). KBr disk (FTIR, model Spectrum One, Perkin Elmer, USA) used to study the FTIR spectra of the samples. Microstructure and surface morphology of ACTF adsorbent before and after oil adsorption was examined using scanning electron microscope (SEM, JEOL JSM 671°F). TEM analysis was done using high resolution transmission electron microscope (HRTEM, model JEOL JEM 2200FS TEM with a field emission gun operating at 200 kV). The Raman spectrum was measured using (Model Sentera, Bruker, Germany) equipment, at laser wave length 532 nm [doubled Nd:YAG laser (neodymium-doped yttrium aluminum garnet)] and power 10 mW. The equipment was fitted with an Olympus metallurgical microscope and the sample was investigated on a microscope slide with an X 80 lens. The spectrum was obtained at room temperature in the spectral range of 1200–100 cm^{-1}.

2.4. Batch adsorption experiments

The batch adsorption experiments performed in 100 ml conical glass flask containing 25 ml of oil/water emulsion contacting 5 g of the adsorbent. The flask put in an orbital shaker device set under constant stirring rate (150 rpm). In order to follow up the remaining oil concentrations, withdrawn samples were centrifuged at 4000 rpm for 15 min, the supernatant was repeatedly extracted using cyclohexane to extract the remaining oil. The oil concentrations were evaluated based on ASTM D7678-11(2011) [35]. Contact time effect was studied over time range (0.5–24 h), with initial concentration (C_o = 1000 mg/l) at 308 K. adsorption isotherm performed using different initial concentrations of oil (C_o = 100–2500 mg/l) at 308 K. Thermodynamic adsorption experiments were conducted over a temperature range (288 to 318 K) at initial oil concentration of 1000 mg/l.

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