



Effect of adsorbent addition on floc formation and clarification



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ABSTRACT

Adding adsorbent into the coagulation process is an emerging treatment solution for targeting hard-to-remove dissolved organic compounds from both drinking water and industrial wastewater. The impact of adding powdered activated carbon (PAC) or organoclay (OC) adsorbents with ferric chloride (FeCl_3) coagulant was investigated in terms of potential changes to the coagulated flocs formed with respect to size, structure, and breakage and regrowth properties. The ability of dissolved air flotation (DAF) and sedimentation (SED) clarification processes to remove hybrid adsorbent-coagulant flocs was also evaluated through clarified water quality analysis of samples collected in bench-scale jar test experiments. The jar tests were conducted using both a synthetic fresh water and oily wastewater test water spiked with dissolved aromatic compounds phenol and naphthalene. Results of the study demonstrated that addition of adsorbent reduced the median coagulated floc size by up to 50% but did not affect floc strength or regrowth potential after application of high shear. Experimental results in fresh water demonstrated that sedimentation was more effective than DAF for clarification of both FeCl_3 -PAC and FeCl_3 -OC floc aggregates. However, experimental tests performed on the synthetic oily wastewater showed that coagulant-adsorbent floc aggregates were effectively removed with both DAF and sedimentation treatment, with lower residual turbidity achieved in clarified water samples than with coagulation treatment alone. Addition of OC or PAC into the coagulation process resulted in removals of over half, or nearly all of the dissolved aromatics, respectively.

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1. Introduction

The removal of dissolved organic contaminants presents a challenge for both the drinking water and industrial wastewater treatment industries. While coagulation is a commonly used water treatment technology, it has long been understood to be limited in its ability to remove certain types of dissolved organic compounds from water (Edwards, 1997), particularly compounds with low molecular weight or high charge density (Collins et al., 1986). In petroleum industry wastewaters, such as produced water or refinery wastewater, coagulation with aluminum or ferric-based metal salts has been shown to be highly effective for the removal of emulsified, or dispersed, oils from wastewater (Rios et al., 1998; Zouboulis and Avranas, 2000), but is largely ineffective for the removal of the dissolved organic compounds, such as phenol, typically present in these waste streams (Younker and Walsh, 2014a). Previous work by the authors demonstrated that an

integrated coagulation-adsorption treatment for produced water can effectively remove both the dispersed and dissolved organic fractions (Younker and Walsh, 2014b).

Several other studies have evaluated the combination of chemical coagulation and adsorption into a single treatment step to enhance removal of target contaminants in drinking water and municipal wastewater systems. Integrating powdered activated carbon (PAC) adsorption with coagulation has been examined in drinking water treatment systems to (1) enhance removal of dissolved organic carbon (DOC) and UV254 (Kristiana et al., 2011; Szlachta and Adamski, 2009; Uyak et al., 2007), (2) target the removal of the taste and odour-causing compounds (i.e., 2-methylisoborneol (MIB) and gesomin) (Cook et al., 2001) and (3) simultaneously remove both humic acid and phenol (Tomaszewska et al., 2004). In those studies, the coagulation process was used to target the larger, more negatively charged fraction of natural organic matter (NOM), while PAC adsorption was used to target the smaller, neutral-charged and/or aromatic organic fractions. Organoclay (OC) adsorbent coupled with coagulation has also been investigated for simultaneous removal of phosphate and phenanthrene as a tertiary treatment for municipal wastewater (Ma et al.,

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2010).

In addition to enhancing the finished water quality, the introduction of adsorbent material into the coagulation-flocculation process may have an impact on floc properties that could impact removal in subsequent clarification steps. In ballasted flocculation processes, microsand is added to the flocculation tank to deliberately change floc properties in order to enhance their settling rate (Desjardins et al., 2002; Ghanem et al., 2007; Young and Edwards, 2003). Addition of microsand to flocs was found to increase floc strength (Dianous and Dernaucourt, 1991) and floc density (Ghanem et al., 2007). Several adsorbent materials have also been investigated as ballasting agents. Demirci et al. (1998) investigated the use of clay as a ballasting agent to enhance turbidity removal by sedimentation in petroleum refinery wastewater treated with alum coagulation. In that study clay addition reduced settled water turbidity by 80% compared to alum treatment alone, and also improved removal of organic compounds as measured by chemical oxygen demand (COD). Aguilar et al. (2003) examined PAC as a coagulant aid for slaughterhouse wastewater treatment, and found increased particle removal (i.e., 95%) with PAC addition as compared to ferric sulfate coagulation alone (i.e., 87%). However, there is a paucity of research that has investigated the impacts of adding adsorbents and coagulants simultaneously on floc structure and DAF clarification.

The purpose of this study was to investigate the impact of PAC and organoclay (OC) addition on floc properties and downstream DAF and sedimentation clarification efficacy in both fresh water and synthetic oily wastewater.

2. Materials and methods

2.1. Adsorbents

Test adsorbents used in this study were powdered activated carbon (PAC) (Sigma-Aldrich) and a lab-synthesized organoclay (OC). OC was prepared following the procedure described by Shen (2004) using sodium montmorillonite clay from the Source Clays Repository (Clay Minerals Society, Columbia, MO). The clay was swelled in DI water overnight by mixing with a magnetic stirrer, after which benzyltrimethyl ammonium chloride solution (Sigma Aldrich) equal to 100% of the clay's cation exchange capacity was added and stirred with a mechanical mixer for six hours. The resulting organoclay was vacuum filtered and rinsed repeatedly with DI water, dried overnight in a 100 °C oven, crushed and sieved through a 200 mesh sieve.

The specific gravity of OC was calculated as 2.2 ± 0.005 by measuring the volume of a known mass sample by helium displacement in a stereopycnometer (Quantachrome, Boyton Beach, Fla, USA). Wetted PAC has been reported to have a specific gravity of 1.3 (Hendricks, 2006).

2.2. Synthetic wastewater

Fresh water trials were conducted using DI water with 5 mg/L of NaHCO_3 added for alkalinity. A stock solution of synthetic produced water with 100 mg/L of emulsified crude oil, 5 mg/L phenol (Sigma Aldrich), 1 mg/L naphthalene (Fisher Scientific) and 32 g/L sea salt was generated according to procedure described by Younker and Walsh (2014a,b). Prior to testing, both fresh water and wastewater were adjusted to a pH level of 8 using pre-determined volumes of 1 M NaOH solution, in order to maintain the water conditions near the pH of minimum solubility for FeCl_3 .

2.3. Jar test procedure

Jar tests were performed in a 1L DAF jar tester (EC Engineering, Edmonton, Canada) under conditions similar to the procedure outlined by Younker and Walsh (2014b) for integrated adsorption-coagulation treatment of synthetic produced water. For coagulation tests, 50 mg/L of FeCl_3 (in a 10 g/L solution) was added to the water and rapid mixed at 200 rpm ($G\text{-value} = 550 \text{ s}^{-1}$) for 1 min. The test water was then mixed for 5 min at 50 rpm ($G\text{-value} = 55 \text{ s}^{-1}$) to simulate the flocculation stage, followed by clarification. For adsorbent-only tests, 100 mg/L of PAC or OC were added to the jars and fully dispersed into the water by mixing at a high speed of 300 rpm ($G\text{-value} = 1000 \text{ s}^{-1}$), then mixed at 50 rpm ($G\text{-value} = 55 \text{ s}^{-1}$) for 5 min, followed by clarification. For combined adsorbent-coagulation trials, PAC and OC adsorbents, at either a low dose of 100 mg/L or a high dose of 1000 mg/L, were added to the jars in separate experiments and fully dispersed into the water by mixing at a high speed of 300 rpm ($G\text{-value} = 1000 \text{ s}^{-1}$) for 1 min, followed by coagulant addition and rapid mixing at 200 rpm ($G\text{-value} = 550 \text{ s}^{-1}$), then a 5 min flocculation stage at 50 rpm ($G\text{-value} = 55 \text{ s}^{-1}$), followed by clarification. DAF clarification was performed using a 10% recycle rate with DI water saturated with air at 75 psi (i.e., 517 kPa). In the sedimentation trials, the jar test apparatus mixers were turned off to allow for floc settling. Grab samples of approximately 30 mL of DAF and SED clarified water were taken at 0, 5, 10, and 30 min of clarification, through the sample port for analysis.

For floc characterization tests, the flocculation stage as described in jar test experiments was followed by a 5 min floc breakage phase at a high shear of 200 rpm ($G\text{-value} = 550 \text{ s}^{-1}$) followed by a 5 min floc regrowth phase where $G\text{-values}$ were returned to the flocculation stage value of 50 rpm ($G\text{-value} = 55 \text{ s}^{-1}$).

2.4. Analytical methods

Particle size distribution (PSD) in flocculated water was measured using a Malvern Mastersizer (Malvern Instruments, Worcestershire, UK). Flocculated water was pumped directly from the jar tester to the sample cell by a peristaltic pump located after the sample cell, and recirculated back to the jar tester. Photographic images of flocs were taken using a Zeiss microscope with a digital camera attachment (Carl Zeiss, Jena, Germany). Microscope slides were prepared by drawing water through a wide-mouth glass pipette approximately 2 inches below the water surface, depositing 1 mL of flocculated water on the slides, and allowing slides to dry before analysis. Zeta potential was measured in both fresh water and synthetic wastewater using a Malvern Zetasizer (Malvern Instruments, Worcestershire, UK). Turbidity measurements were performed using a Hach Turbidimeter (Hach Company, Loveland, USA).

Water quality tests were performed on synthetic wastewater samples. Total oil and grease concentrations were determined by IR spectroscopy (Bruker Optics, Ettlingen, Germany) using Standard Method 5520C (APHA, AWWA, and WEF, 2005), with tetrachloroethylene substituted as the extraction solvent, as described by Farmaki et al. (2010). Phenols concentrations were analyzed by 4-aminoantipyrine indicator method and UV-VIS spectroscopy (Hach Company, Loveland, USA) using Standard Method 5530 (APHA, AWWA, and WEF, 2005). Naphthalene concentration was determined by headspace analysis of a 0.8 mL sample by adsorption onto and desorption from solid phase micro extraction polydimethylsiloxane fibres using a 3800 Varian gas chromatograph (Agilent Technologies, Santa Clara, USA) with flame ionizing detection.

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