



Retention–oxidation–adsorption process for emergent treatment of organic liquid spills

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ABSTRACT

The feasibility and effectiveness of retention–oxidation–adsorption process (ROA) for the elimination of organic contaminants induced by chemical accidents were investigated in this study. Organobentonites (DTMA-, TTA-, CTMA- and OTMA-bentonite), potassium ferrate (Fe(VI)), ozone and granular activated carbon (GAC) were used as rapid and efficient materials in the treatment and recovery of organic liquid spills. Results indicated that the retention capacities of organobentonites (especially CTMA-bentonite) were much higher than that of natural bentonite towards the chosen organic compounds. Additionally, pH, oxidant dosage, initial concentration of contaminant and chemical structure had significant influences on the effectiveness of the oxidation process. In a pilot-scale experiment, the ferrate/GAC (F/G) and ozone/GAC (O/G) processes made a comparatively good performance in the treatment of wastewater containing aniline or nitrobenzene, with the removal efficiencies of the contaminants greater than 80%. Overall, the ROA process showed a high efficiency and steady operation in the removal of hazardous organic liquids and subsequent clean up of the contaminated site.

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1. Introduction

In recent years, there has been growing concern in China about the widespread occurrence of chemical incidents in industry and road transport [1]. Every year, sudden and unexpected contamination events (accidental leakage, spills, explosions, etc.) put large numbers of the public in the danger of exposure to hazardous organic pollutants (benzene, nitrobenzene, petroleum, etc.), which account for about 34% of the total involved pollutants [2]. Since the accident scenes and contaminants varied, the systematic study of remediation technologies and processes seems to be very difficult and necessary, especially in China.

When chemical incidents take place accidentally, response actions must be quick and accurate to minimize the impact of the contaminants [3]. The method of retention is considered to be one of the most common processes for the removal of hazardous organic liquids and subsequent cleanup of the contaminated site, and many types of sorbents are developed accordingly. Among them, organobentonites, normally synthesized by cationic surfactants and bentonite with cationic exchanging, are widely used as effective sorbents for organic contaminants and numerous stud-

ies have investigated their mechanisms in the past decades [4–6]. Ozonation [7–10] and ferrate oxidation [11–16] have also shown their own advantages in removal of a wide range of organic pollutants both in wastewater and drinking water. Meanwhile, activated carbon has proven to be one of the most effective adsorbent materials, which is widely used in the elimination and recovery of hazardous organic pollutants [17–19]. However, more information remains to be gathered to evaluate their application in organic contamination treatment.

Considering the ideas addressed above, this research chose eight organic compounds as representative pollutants on the basis of their predominant occurrence in chemical accidents, including aromatics (benzene, toluene, *o*-xylene, nitrobenzene, chlorobenzene, aniline) and chlorophenols (2,4-dichlorophenol, 2,4,6-trichlorophenol), and introduced an effective retention–oxidation–adsorption process for the emergent treatment of organic liquid spills induced by chemical accidents. The present study was conducted to investigate the effectiveness and the influence factors (sorbent material, solution pH, oxidant dosage, initial concentration of contaminant, chemical structure, etc.) of retention–oxidation–adsorption process, and to try to develop a mobile treatment system based on the ROA process for environmental emergency management departments, corporations that deal with organic chemicals, etc., which could be applied for the treatment of chemical incidents that occur in industry and road transport.

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2. Experimental

2.1. Chemicals

Benzene, toluene, *o*-xylene, nitrobenzene, chlorobenzene, aniline, 2,4-dichlorophenol, 2,4,6-trichlorophenol were purchased from Sinopharm Chemical Reagent Co. (China) and were of 99% purity or higher. Dodecyltrimethylammonium bromide (DTMAB), tetradecyltrimethylammonium bromide (TTAB), cetyltrimethylammonium bromide (CTMAB) and octadecyltrimethylammonium bromide (OTMAB) of analytical grade were obtained from Shanghai Chemical Co., China.

The natural bentonite composed primarily of Ca^{2+} -montmorillonite was obtained from Inner Mongolia, China. The bentonite BET- N_2 surface area, organic carbon content, and cation exchanged capacity (CEC) was $73.3 \text{ m}^2 \text{ g}^{-1}$, 0.17%, and 108 cmol kg^{-1} , respectively.

Potassium ferrate (97%) was purchased from Sigma–Aldrich Co., USA. Ozone was generated from oxygen by an ozone generator (NPL5W, Green Continent Co., China) with a maximum ozone production rate of 5 g h^{-1} . All other chemicals were of reagent grade, and used as received. Doubly distilled water was used throughout the study except in the pilot-scale experiment.

2.2. Batch retention experiment

In this investigation, DTMA-bentonite, TTA-bentonite, CTMA-bentonite, OTMA-bentonite and natural bentonite were chosen as sorbents. The organobentonites were synthesized with the following procedure [20]: 30.0 g natural bentonite was mixed with 500 mL of an aqueous solution containing 9.99 g DTMAB, 10.9 g TTAB, 11.8 g CTMAB or 12.7 g OTMAB, respectively, equal to 100% of bentonite's CEC. The mixture was stirred at 60°C for 4 h. The product was separated from water by vacuum filtration and washed with distilled water six times, then dried at 70°C for 12 h. Finally, the organobentonite was grounded to pass a 100 mesh sieve.

The retention capacity measurements were undertaken as follows [21]: 200 mL of pure pollutant liquid was poured into a 250 mL pyrex glass beaker. 5.0 g of sorbent was weighed and the value recorded. The sorbent material was spread evenly on a filter paper, placed into a circular brass wire mesh (2 mm^2) basket ($\Phi 5.0 \times 8.0 \text{ cm}$), and then lowered into the beaker so that the sorbent was completely covered by the liquid. After 15 min of immersion,

the basket with the sorbent was hung up for about 5 min to allow the excess liquid to be drained off. Then, the saturated sorbent was transferred to a suitable pre-weighed dish and weighed. Additionally, to evaluate the effect of retention time between the chosen pollutants and sorbents, the experiments were conducted with the same procedure other than the volume of pure organic liquid was reduced to 25 mL and the contact time varied from 1 to 30 min. All tests were carried out in triplicate.

The retention capacity of the sorbents and the retention rate of organic liquids on a weight basis were calculated as follows: retention capacity (g g^{-1}) = $(S_1 - S_0)/S_0$, retention rate (%) = $(S_1 - S_0)/S_2 \times 100$, where S_0 is the initial dry weight of sorbent, S_1 is the weight of the saturated sorbent, S_2 is the weight of the pure organic liquid.

2.3. Degradation by ozonation and ferrate oxidation

The experimental set-up used for wastewater treatment by ozonation consisted of a pressurized oxygen gas cylinder, an ozone generator, a gas flow meter, and a reaction vessel ($\Phi 5 \times 50 \text{ cm}$) of about 1 L in volume. The reactor was equipped with a gas diffuser at the bottom to sparge the ozone/oxygen stream into the water. The exhaust gas vented from the top of the reactor was captured in a pair of adsorption bottles containing 2% (w/v) KI solution. 500 mL of a solution that contained different contaminants at various concentrations was adjusted to the desired pH value by using either sulfuric acid or sodium hydroxide, and added to the reactor before the ozone bubbled. In this work, the ozone dosage added into the system was controlled between 5.59 mg min^{-1} and 35.1 mg min^{-1} by a gas flow meter. Samples were collected at fixed intervals and $1.0 \text{ mol L}^{-1} \text{ Na}_2\text{SO}_3$ was added to remove the residual ozone and organic radicals prior to analysis.

Batch Fe(VI) oxidation experiments were conducted in 100 mL conical flasks in which 50 mL of a solution was stirred vigorously by magnetic stirrer. $1.0 \text{ mol L}^{-1} \text{ Na}_2\text{SO}_3$ was added into the samples immediately to stop any further reaction. After the quenching, samples were centrifuged at 8000 rpm for 10 min, and then filtered with $0.45 \mu\text{m}$ filters before analysis.

All the experiments were conducted in triplicate at room temperature ($23 \pm 2^\circ\text{C}$). The elimination of the target pollutant through volatilization was evaluated and deducted. Removal is defined as (%) = $(C_0 - C)/C_0 \times 100$, where C_0 is the initial contaminant concentration or TOC.

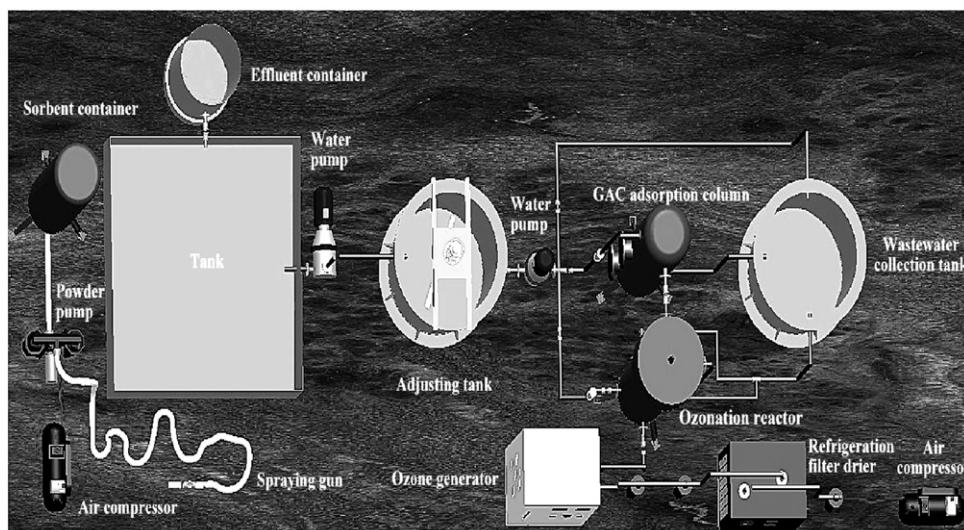


Fig. 1. The schematic diagram of the retention–oxidation–adsorption system.

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