

THE USE OF A LOCAL SOIL IN ENVIRONMENTAL ENGINEERING PROCESSES

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ABSTRACT

The local soil used in this study was found to contain 68% quartz, 24% muscovite, and 8% hedenbergite and to have a surface area, a pore volume, and a pore diameter of $173 \text{ m}^2/\text{g}$, $0.16 \text{ c}^3/\text{g}$, and 40.5 \AA , respectively. This study investigates the use of soil in environmental engineering processes. In the first part of the study, hexadecyl trimethyl ammonium bromide (HDTMA) surfactant was mixed with the soil in order to investigate its effect on the soil sorptive capacity. Phenol adsorption isotherms conducted under different combinations of HDTMA and dissolved oxygen (DO) revealed that both HDTMA and DO enhance the adsorptive capacity of the soil for phenol. HDTMA-induced increase of 48.5% and 312% in the soil sorptive capacity was attained at a residual phenol concentration of 100 mg/l under anoxic and oxic conditions, respectively. In the second part of the study, the use of the aforementioned soil as a destabilizer for an emulsified colloidal wastewater was investigated. While, preliminary investigation of the pollutants in the raw wastewater revealed their poor solubility and settleability, the reduction of the supernatant COD increased with the increase in soil mass and time until equilibrium. Destabilization of the polymers by the soil was attributed to the adhesion enhanced by the large soil surface area and the expansive nature of muscovite while sedimentation was believed to occur because of discrete and zone types of settling. Considerable removal efficiency was achieved in the first hour and in the time range of (6-24 hours) resulting in removal efficiencies up to 95%.

KEYWORDS

Soil, Surfactant, Dissolved Oxygen, Phenol, Polymeric Waste, Adsorption, Coagulation.

INTRODUCTION

Purification of industrial wastewater prior to discharge is necessary to keep our environment clean. However, industrial wastewater treatment is becoming very costly, particularly, if chemicals are used. Therefore, research on less costly treatment is crucial.

Clays or soils containing clay are prevalently used as containment liners in hazardous landfill sites and in slurry walls at hazardous waste sites, with the primary objective of retarding the migration of contaminants from the site (Chion (1983)). A major mechanism of organics-soil interaction involves a partition process that depends on solute water solubility (Chion (1983)) and the soil organic matter Garbarini (1986). Thus, low organic matter soils need to be modified to enhance their efficiency in altering organics mobility. Boyd et al. (1988) and Lee et al. (1989) have used cationic surfactants to modify soils and increase their retardation of nonpolar organics. Bouchard et al. (1988) have demonstrated that ethyl hexadecyl dimethyl ammonium (EHDDMA) has increased toluene sorptive capacity of an aquifer material by two orders of magnitude. Most earlier works (Garbarini (1986); Boyd et al. (1988), Smith et al. (1990); Smith and Faffe (1991)) have focused on nonpolar organic contaminants. Recently, however, Haus and Uchrin (1995) demonstrated the viability of soils modification with cationic surfactants to retard polar organics such as 2-hexanone and o-chlorophenol. Sorption studies conducted with dodecyl dimethyl ammonium bromide and hexadecyl trimethyl ammonium bromide (HDTMA) revealed that HDTMA is a better partitioning media on organic matter basis (Lee et al. (1989); Haus and Uchrin (1995)).

Pristine groundwaters contain very low dissolved oxygen and are thus readily rendered anaerobic by contamination. Furthermore, landfill leachates are rarely aerobic and maintain low dissolved oxygen concentrations. Thus the interaction of organics with aquifer soils or containment clays occurs predominantly under oxygen limiting conditions. The influence of dissolved oxygen on sorption of organics is obfuscated by its seemingly indirect involvement except in so far as biodegradation is concerned.

Dissolved oxygen (DO) was found to enhance the adsorptive capacity of granular activated carbon (GAC) for phenolics (Vidic and Suidan (1991); Abuzaid and Nakhla (1994); Abuzaid and Nakhla (1995); Abuzaid et al. (1995); Abuzaid and Nakhla (1996a); Abuzaid and Nakhla (1996b); Emmanuel et al. (1996). While Abuzaid et al. (1995) found the enhancement phenomenon valid for phenol, o-cresol, 4-nitrophenol, and water containing aromatics, Vidic and Suidan (1991) reported that it was not observed for trichloroethene.

Polymers-containing wastewater are difficult to treat because of their complex nature. This difficulty increases when those polymers are neither soluble nor settleable. Soluble waste can be treated by adsorption (Abuzaid and Nakhla (1996)) or biodegradation ((Nakhla and Harazin (1993)) while for settleable waste, sedimentation is the most used process. For emulsified suspended polymeric waste, on the other hand, the constituents of the wastewater need to be coagulated and flocculated before it can be clarified.

Colloidal dispersion may be stabilized by electrostatic repulsion between particles, arising from ions either adsorbed onto or dissolved out of the solid surface. Destabilization of those colloids is essential in order to bring them in contact and aggregate. Coagulants destabilize colloidal particles by four mechanisms: double layer compression; charge neutralization; enmeshment in a precipitate and interparticle bridging (Benefield et al. (1978)).

Separation of colloidal industrial wastes were usually undertaken by the use of chemical coagulants. Shut'ko (1986) investigated the treatment of petroleum wastewater by aluminum-containing coagulants. The findings of the study revealed that aluminum-containing coagulants were effective for the separation of petroleum wastewater, particularly for effluents with high suspended matter. Coagulation of pulp and paper mill

effluents using alum and chlorinated copperas was carried out by Soetopo (1984). In his study, it was reported that significant reduction of pollution parameters were achieved. Wastewater from a textile plant was effectively treated by alum coagulation resulting in disperse dye reduction of 85 per cent (Colak and Arikan (1986)). The cost involved in the purification of the aforementioned wastewater by chemical coagulants should not be overlooked. As mentioned earlier, cheaper alternatives should be investigated.

The soil used in this study is a local soil found in the Eastern Province of Saudi Arabia, named Khoweldi by the local people. The Khoweldi soil will be named K-soil throughout the paper. The first part of the study is focused on the effect of surfactant and DO on the sorption capacity of the soil for phenol while the second part investigates the efficiency of the K-soil soil for the purification of the polymeric colloidal wastewater described earlier.

Soil Characterization

The surface area of the K-soil was determined by measuring the nitrogen adsorbed on a degassed sample at different relative pressures and liquid nitrogen temperature. The procedure involves measuring the nitrogen physically adsorbed on the surface of the soil at the liquefaction temperature of nitrogen using the theory developed by BET.

X-ray diffraction (XRD) is an indispensable technique for investigation of the crystal structure of the solid matter. In this study, the XRD analysis was performed on a Philips PW 1700 automated diffractometer with a monochromator and spinner. Diffraction patterns were generated on a vertical goniometer attached to a broad focus X-ray tube with a copper target operating at 45 kV and 30 mA. The K-soil specimens were powdered with a pestle and mortar. A homogeneous sample of this powder was packed in to a sample holder and scanned from $4 - 80^\circ 2\theta$ at a speed of $0.01 2\theta$ °/s.

Fourier transform infrared spectrometer (FTIR) was used for the qualitative identification of the organic molecules in the K-soil samples. FTIR analysis of a K-soil sample treated with HDTMA and that untreated were carried out on the Nicolet 20 SXB FTIR. Routine parameters using DTGS detector, and a resolution of 8 cm^{-1} and in the middle of the infrared range (4000 to 400 cm^{-1}) were utilized for this analysis. Potassium bromide (KBr) pellets were prepared for the K-soil samples.

Isotherm Experiments

Single-solute stock solution (1000 mg/l) of phenol (BDH Chemicals, U.K.) was prepared and subsequently buffered with KH_2PO_4 to maintain neutral pH. The soil under study was categorized into two categories; treated soil (TS) (with HDTMA) and the untreated soil (UTS) (without HDTMA). For each of the two categories of the K-soil, two sets of 160-ml bottles containing varying amounts of the K-soil were prepared and subsequently filled with 100 ml of phenol solution. One set was purged with nitrogen until oxygen was completely stripped (anoxic condition), and the bottles were quickly closed with a rubber stopper. The other set of bottles was purged with pure oxygen until saturation was achieved as evidenced by a DO concentration around 30 mg/l (oxic condition). Each set included two bottles without soil to serve as blanks in order to check for sorbate volatilization and adsorption of

sorbate on to the walls of the container. All bottles were placed on a rotary shaker at a room temperature of 21 ± 2 °C for a period of 7 days. At the end of the equilibration period, samples were withdrawn from each bottle, filtered through 0.45- μm Millipore filter paper, and analyzed for sorbate residual concentrations using Spectronic 21 spectrophotometer (Boush and Lomb Model,UV-D) at a wavelength of 271 nm.

Coagulation of the Polymeric Waste

Twenty-liter sample was collected from the wastewater effluent of the factory and stored in a fridge to prevent any biological activity. One liter of the wastewater was taken from the stored sample and divided into two portions. One portion was filtered using 0.45 μm filter paper. The filtered and unfiltered portions of the wastewater were analyzed for COD in order to know the soluble and total COD, respectively. The COD was analyzed in accordance with the procedure outlined in the Standard Methods (1980).

The settleability of the polymeric colloids in the wastewater was tested in order to check the extent of stability of the colloids. A one liter volume measuring cylinder was filled with the wastewater and monitored for a period of 24 hours for COD. The settleability test was conducted according to the procedures outlined in the Standard Methods (1980).

For the coagulation study, jar tests were conducted using a multiple stirring apparatus. Seven batches were run simultaneously, each was filled with 500 ml of the wastewater. Six different masses of the K-soil (0.5, 1, 1.5, 2, 2.5, and 3 grams) were added to the aforementioned beakers. The seventh beaker was run without soil to serve as a blank. In the beginning, the beakers were mixed at high speed (100 rpm) for 1 minute. Subsequently, the mixing speed was lowered (40 rpm) and continued for another 10 minutes. The speed was further reduced to 8 rpm and the mixing process continued for another 10 minutes. After the mixing stage, COD of the supernatant was monitored with time.

RESULTS AND DISCUSSION

Soil Characterization

Physical characterization of any potential adsorber is of extreme importance since it gives an idea about its chances to be used as an adsorber. Identification of the three pertinent variables i.e. surface area, volume of pores, and pore diameter, resulted in values of 173 m^2/g , 0.16 c^3/g , and 40.5 A, respectively. Those results, particularly, the surface area show that the K-soil has, relatively, good sorptive ability. In fact, the surface area of the K-soil is more than 20 % of that related to activated carbon (Abuzaid (1993)).

Since, previous research (Garbarini (1986); Boyd et al. (1988), Smith et al. (1990); Smith and Faffe (1991) Haus and Uchrin (1995)) established the effect of surfactant treatment on clay's sorptive capacity, X-ray diffraction (XRD) tests were conducted, mainly, to investigate the existence of clay in the K-soil. The results of the XRD analysis showed that the K-soil contains 68% quartz, 24% muscovite, and 8% hedenbergite. The XRD pattern for the K-soil is given in Fig. (1,a) while the patterns of its constituents i.e. quartz, muscovite (clay), and hedenbergite are shown in Figs. (1,b), (1,c), and (1, d) respectively.

Fourier transform infrared (FTIR) tests showed interaction between the K-soil and HDTMA. Fig. 2 shows the FTIR spectrums for both natural K-soil (untreated with HDTMA) and that treated with HDTMA. The figure shows that the two spectrums are identical with the exception of clear peaks at wave numbers in the range of (2800-3000) in the case of the HDTMA treated soil which is a characteristic of a hydrocarbon. Since HDTMA was the only treatment to the natural K-soil, in Fig. (1, a), it is obvious that such hydrocarbon is HDTMA. The aforementioned result is important since it establishes the interaction between the K-soil and HDTMA.

Isotherm Studies

After the determination of phenol residual concentrations, the corresponding carbon load for each liquid phase concentration was calculated as $(q = \frac{V(C_0 - C)}{M})$, where q is the adsorber (K-soil) capacity (mg phenol /g K-soil), V is the liquid volume (L), M is the mass of the K-soil (g), and C_0 and C are initial and final phenol concentrations, respectively, (mg/l). The single-solute isotherms for phenol obtained at room temperature (21°C) and neutral pH for each of the cases under study were described by the Freundlich equation ($q = K C^n$), where q and C are as described previously and K and n are model constants. The Freundlich constants are listed in Table 1 for each of the phenol isotherms. From Table 1, it can be seen that for the untreated soil, the two isotherms are near to linear denoted by n values of 0.941 and 0.911 for the anoxic and oxic cases, respectively, which agrees with the findings of Magdalena et al. (1992). However, for the treated soil, the oxic isotherm is close to linear (n=1.16) while the anoxic one exhibited non linear behavior.

Table 1. Freundlich Constants for Phenol Isotherms.

Soil Category	DO Designation	k (mg/g)(L/mg) ^{1/n}	1/n	R ²
Untreated	anoxic	0.02	0.94	0.96
	oxic	0.029	0.91	0.82
Treated	anoxic	2.26X10 ⁻⁵	2.50	0.99
	oxic	0.0378	1.16	0.98

R² is the coefficient of determination

In order to have a general idea about the effects of DO and/or HDTMA, the adsorption capacity of the K-soil under the four conditions presented in Table 1 were calculated and presented in Fig. 3. The aforementioned capacities were calculated at a residual phenol concentration of 100 mg/l using the Freundlich isotherm with the pertinent constants from Table 1. Fig. 3 clearly shows that at a residual phenol concentration of 100 mg/l, the adsorption capacity of the K-soil increases as much as 5.18 folds when was oxygenated and treated with HDTMA. In general, the adsorption capacity is a function of the residual concentration, hence, a more detailed analysis regarding the effect of residual concentration will be presented in the upcoming discussions.

The sole effect of modification by surfactant on the adsorptive capacity of the K-soil can be revealed based on the data of Figs. 4 and 5. The aforementioned figures present the effect of treatment with surfactant on soils under anoxic (no oxygen) and oxic (DO = 30 mg/l)

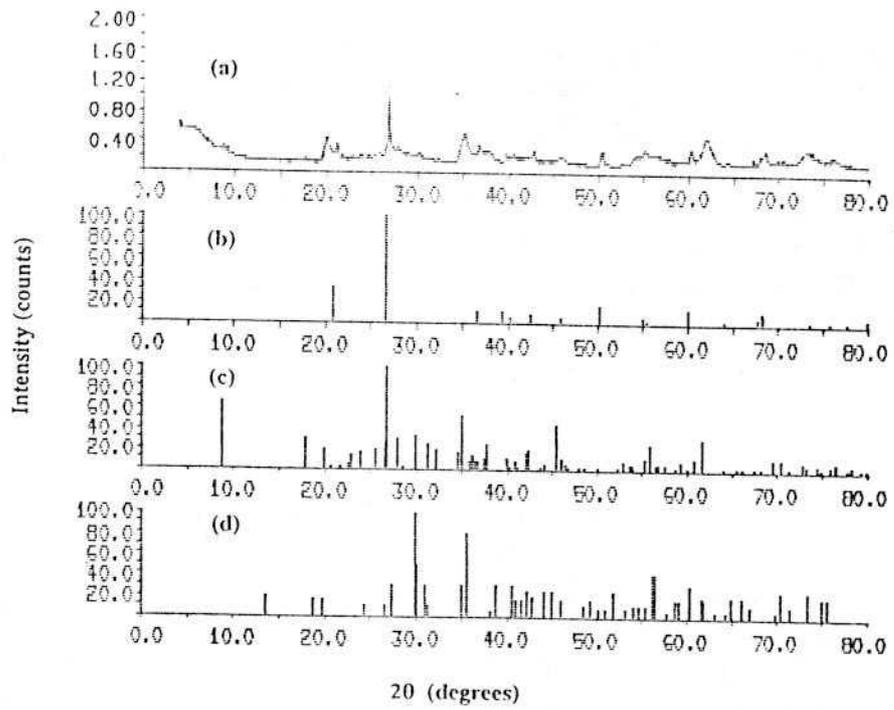


Fig. 1. XRD Patterns of the K-soil (a), Quartz (b), Muscovite (c), and Hedenbergite (d).

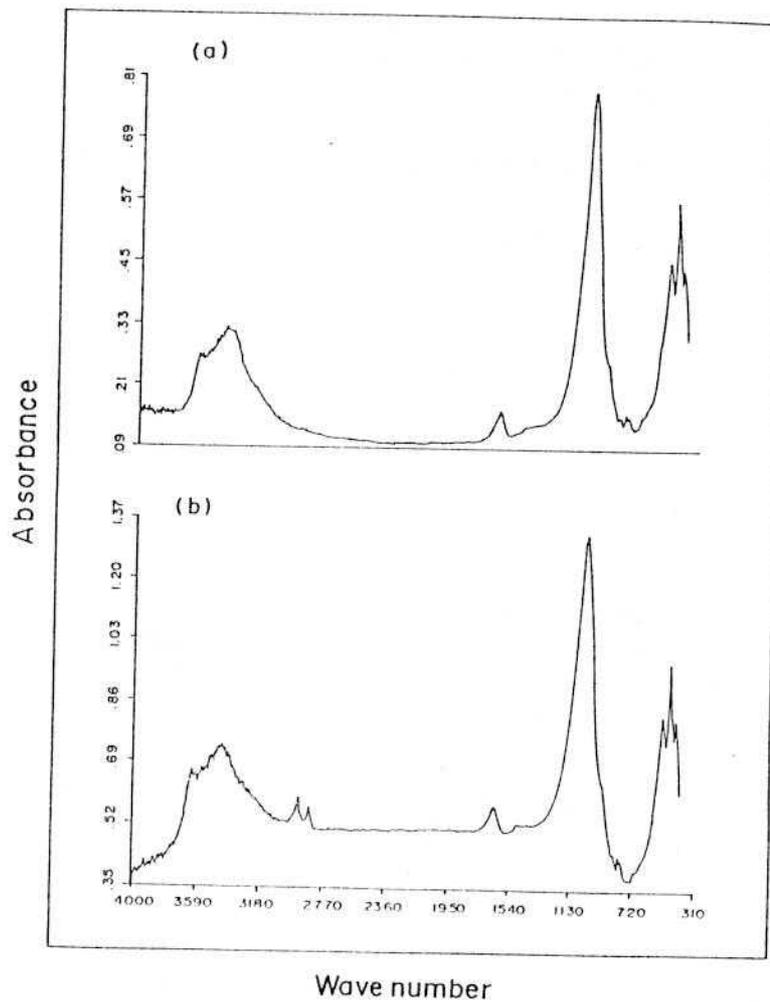


Fig. 2. FTIR Spectrums for the Natural K-soil (a) and K-soil Treated with HDTMA (b).

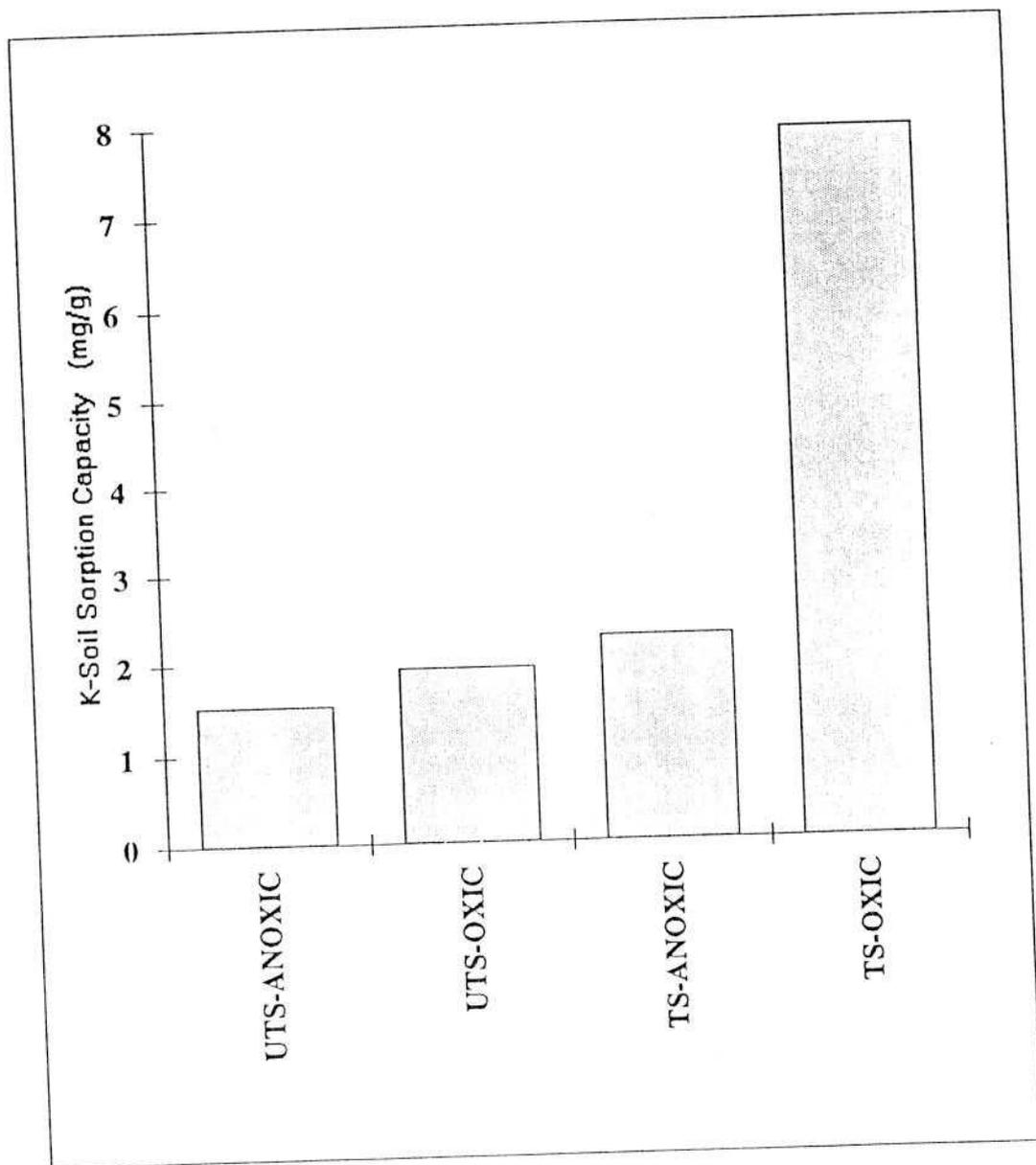


Fig. 3. The Adsorption Capacity of the K-Soil at Phenol Residual Concentration of 100 mg/l under Different DO and HDTMA Treatment Conditions

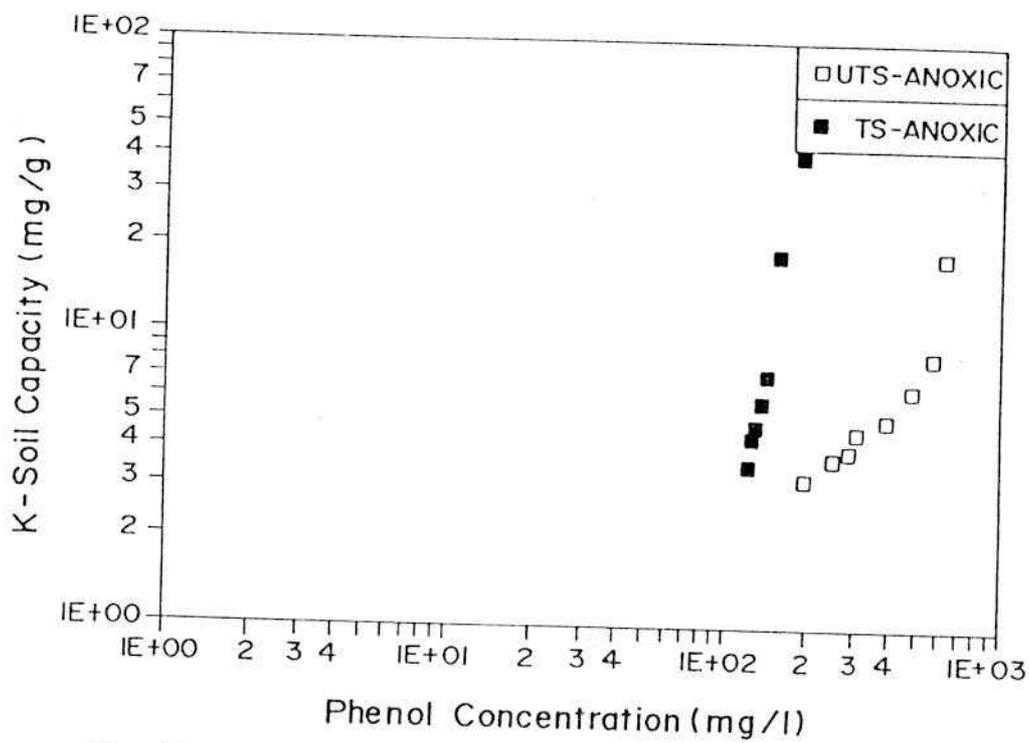


Fig. 4. Freundlich Adsorption Isotherms For the Untreated K-Soil under Oxidic and Anoxic Conditions.

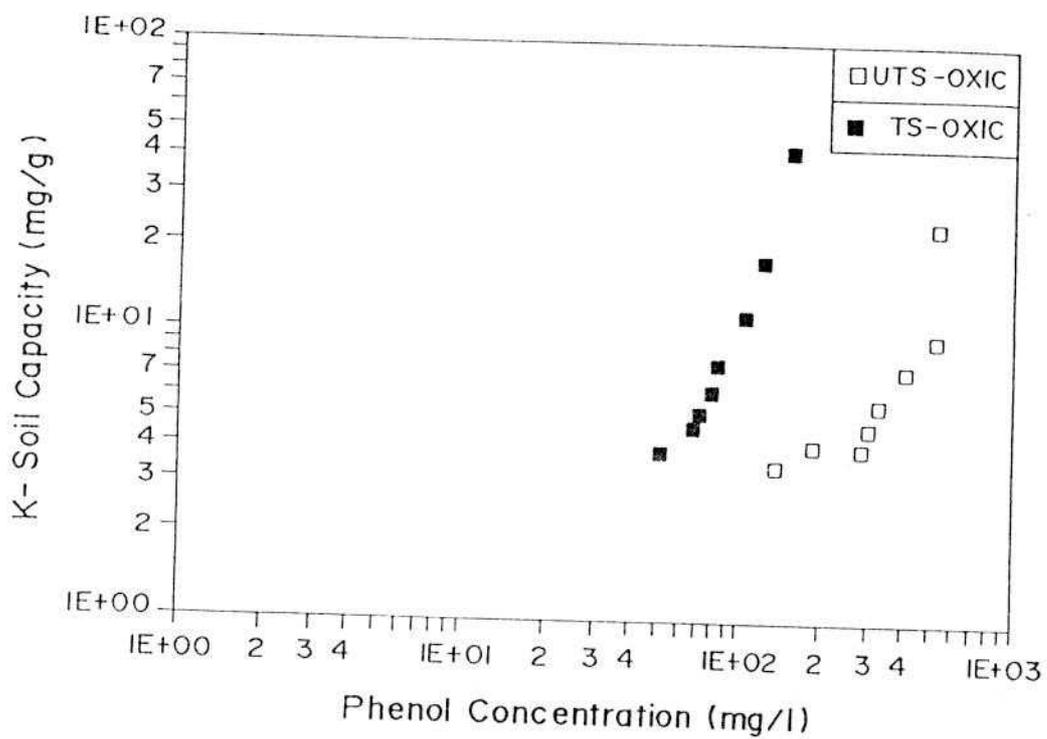


Fig. 5. Freundlich Adsorption Isotherms For the HDTMD-Treated K-Soil under Oxidic and Anoxic Conditions.

conditions, respectively. The data of the figure and Table 1 show that an HDTMA-induced increase of 48.5% and 312% in the sorptive capacity was attained at a residual phenol concentration of 100 mg/l for anoxic and oxic conditions, respectively. Apart from the DO conditions, the aforementioned enhancement is well established in the literature (Garbarini (1986); Boyd et al. (1988), Smith et al. (1990); Smith and Faffe (1991) Haus and Uchirin (1995)). The findings revealed from the previous figure and Table 1 suggest that treatment of the K-soil with HDTMA significantly, enhances its adsorptive capacity.

Figs. 6 and 7 present the effect of DO conditions on the adsorptive capacity of both untreated and treated soils, respectively. The data of the figures show that a DO-induced increase of 26% and 249% in the capacity was attained at a residual phenol concentration of 100 mg/l for untreated and treated soil samples, respectively. The aforementioned findings suggest that keeping oxic conditions while mixing the K-soil with the surfactant, would, significantly, increase the enhancement of its adsorption capacity.

Coagulation of the Polymeric Waste

The COD of the polymeric wastewater discharged by a polymer factory was found to be 13500 mg/l, while the COD of the filtered sample (soluble COD) was found to be 130 mg/l. This clearly shows that the soluble COD is less than 1% of the total COD which is negligible and thus will not be considered in further work. Actually, this finding has motivated the main objective of this work which is destabilization of the emulsified colloids in the polymeric wastewater and not treatment of the soluble constituents. The settleability test of the wastewater revealed that even after a detention time of 24 hours, the colloidal waste was totally in suspension. This stressed the colloids stability and emphasized the need for a destabilizer to flocculate the colloids and have them settled. Hence, conventional sedimentation will not be able to remove the constituents of the wastewater under study.

Fig. 8 depicts the relationship between the supernatant COD and time of sedimentation under the effect of six soil masses. The figure shows that at any time, the supernatant COD is inversely proportional to the amount of soil used. It should be noted from Fig. 8 that equilibrium with time was reached within 24 hours under all of the soil masses. It is worth to mention that negligible COD reduction was found in the beaker with no soil. The interaction between the K-soil and the colloidal polymers in the wastewater may best be interpreted in terms of surface adhesion because of the large soil surface area in general, and the expansive nature of the muscovite constituent in particular (Mitchel (1993)). Regarding sedimentation, there are two possible mechanisms; discrete settling, in which each particle settles by itself which is a characteristic of the coarse portion of the soil, and zone settling in which fine particles settle as a blanket. The second mechanism is supposed to enhance sedimentation of the colloidal polymers by sweeping them by the settling zone. Removal of colloids in this manner is frequently referred to as a sweep-floc coagulation (Benefield et al. (1978)). This is of course, in addition to the polymers-particle adhesion process.

Fig. 9 shows the temporal effect of different soil masses on the COD removal efficiency. The highest removal efficiencies (88-95%) were achieved when the soil mass was three grams, while the lowest values (50-72%) were achieved under a soil mass of 0.5 gram. Fig. 9 has a useful application regarding the design and operation of this process. If the process is to be designed for a certain removal efficiency, two parameters can be manipulated,

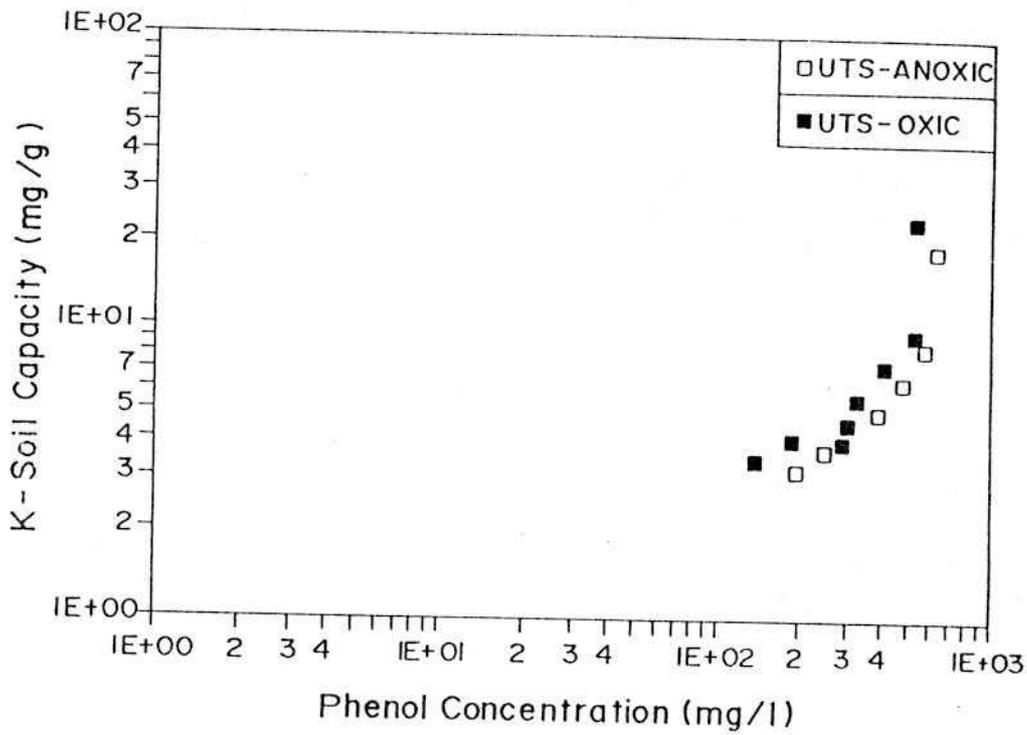


Fig. 6. Freundlich Adsorption Isotherms for the Anoxic K-Soil (Treated and Untreated with HDTMA)

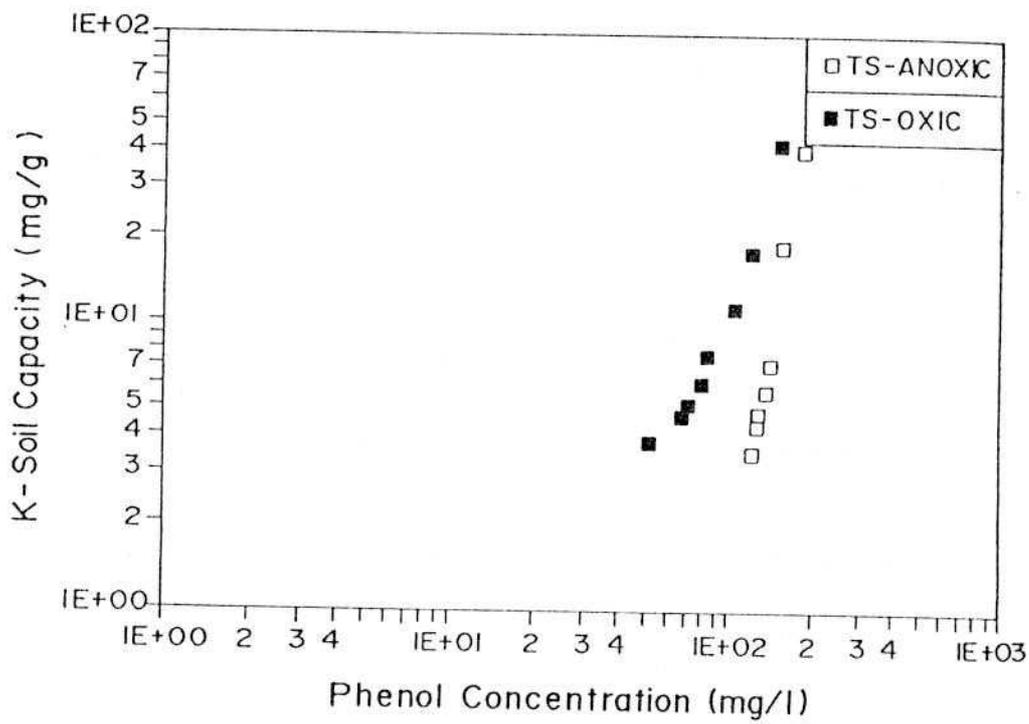


Fig. 7. Freundlich Adsorption Isotherms for the Oxidic K-Soil (Treated and Untreated with HDTMA)

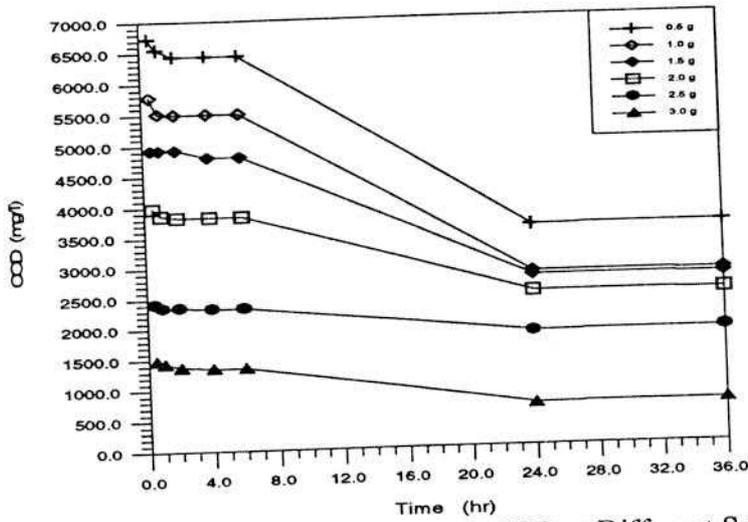


Fig. 8. Temporal Variation of the Supernatant COD at Different Soil Masses

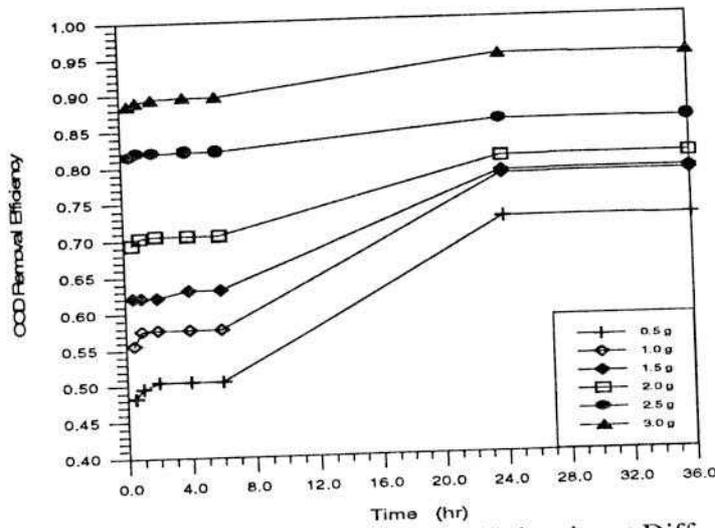


Fig. 9. Temporal Variation of the COD Removal Efficiencies at Different Soil Masses

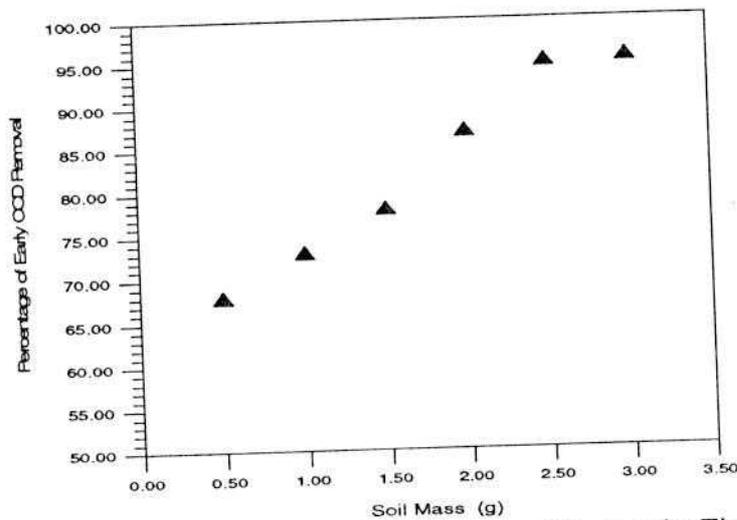


Fig. 10. Relationship between the Ratio of COD Removed During the First One Hour to the Total COD Removed and Soil Mass

namely, soil mass and detention time. For example, a removal efficiency of 70% can be achieved at detention times of 30 minutes, 8 hours, 13 hours and 22 hours, and soil masses of 2, 1.5, 1, and 0.5 grams, respectively. It should be understood that the decision on which pair of these parameters to adapt in order to reach a given removal efficiency is mostly related to economy which is outside the scope of this work. In Fig. 9, it is clear that in the first hour, considerable portion of the removal efficiency was achieved under each of the soil masses and the ratio of the removal efficiency reached to the total removal efficiency increased with the increase in soil mass. Furthermore, this removal efficiency did not increase considerably, in the next five hours for all the masses used. During the time period (6-24 hours) considerable increase in the removal efficiency was achieved. The increase in that period was inversely proportional to the soil mass while, at equilibrium, the total removal efficiency attained was proportional to the soil mass.

The kinetics of this process is very complicated because of the heterogeneity in both the wastewater and the soil. However, it will be tried to explain the aforementioned findings by plotting the ratio of COD removed during the first hour to the total COD removed versus soil mass in Fig. 10. The figure generally, shows that the percentage of the COD removed in the first hour to the total COD removed increases with the increase in soil mass up to a soil mass of 2.5 grams after which the aforementioned percentage did not change. In order to explain this behavior, one should recall that the soil used was not pure clay but a mixture of mainly, muscovite and quartz. The non clay portion was higher in quantity than clay and naturally coarser and heavier. So, it is expected that most of the early COD removal (within one hour) occurs mostly, because of the non clay portion, and clay starts settling with the attached polymers at a later time (after six hours as was shown in Fig. 9. At a low soil mass, the amount of coarse constituents is low, resulting in a comparatively low percentage of early COD removal which will give chance to the fine constituents (clay) to remove more COD with time. However, at a high soil mass, the comparatively high coarse constituents is responsible for higher percentage of early COD removals, reducing the amount of COD to be removed by the fine portion. Above a soil mass of 2.5 grams, the percentage of early COD removals reaches a fixed value of 95%. This means that 5% of the removable COD can not be removed by the coarse portion of the soil regardless of quantity. From Fig. 10, it can be concluded that if efforts were made to increase the clay quantity in the K-soil by separation of quartz, both kinetics and equilibrium capacity would be affected. As a result, kinetics may be lowered while, higher equilibrium capacities are expected to be attained due to the comparatively large surface area of clay. In other words, less soil masses would be needed but at the cost of time.

CONCLUSIONS

Based upon the findings of this work, the following conclusions can be drawn:

- The K-soil was characterized and found to contain 24% muscovite, 8% hedenbergite, and 68% quartz, and to have a surface area, a pore volume, and a pore diameter of 173 m²/g, 0.16 c³/g, and 40.5 A, respectively.
- Phenol adsorption isotherms conducted on the K-soil under different combinations of HDTMA and DO revealed that both HDTMA and DO enhance its adsorption capacity for phenol. HDTMA-induced increase of 48.5% and 312% in the sorptive capacity was

attained at a residual concentration of 100 mg/l for soils under anoxic and oxic conditions, respectively. However, DO increased the soil sorptive capacity by 26% and 249 % at 100 mg/l residual concentration for untreated and treated soils, respectively.

- The soil was found to destabilize the colloidal waste, and the reduction of the supernatant COD increased with soil mass and time until equilibrium.
- The destabilization of the colloidal polymers by the K-soil was attributed to the adhesion enhanced by the large soil surface and the expansive nature of muscovite, while sedimentation was believed to occur because of discrete and zone settling.
- Considerable portion of the removal efficiency was achieved in the first hour and in the time range of (6-24 hours), and the highest removal efficiency achieved was 95%.

ACKNOWLEDGMENTS

The authors would like to thank the Research Institute and the Civil Engineering Department, KFUPM, for supporting this research. The authors thank Mr. Bassam Al-Tawabini, Mr. Mohamed Essa and Mr. Aarif El-Mubarak for their help in the experimental work.

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