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Chemical and Biological Regeneration of HDTMA-Modified Montmorillonite after Sorption with Phenol

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Hexadecyltrimethylammonium (HDTMA)-modified montmorillonite (HMM) has recently been recognized as a potential sorbent to remove organic contaminants from environmental systems. Potential applications of this material highly depend on the efficiency of regenerating contaminant-sorbing HMM. In this study, we investigated a chemical (NaOH solution) and a biological (yeast *Pityrosporum* sp.) method to regenerate phenol-sorbing HMM. Our results showed that the sorption coefficient of phenol to HMM is not a linear function of the ratio of the substitution of HDTMA in HMM. Chemical regeneration of HMMs (0−0.7 times of its cation exchange capacity (CEC)) proved the existence of a phenol residual amount of about 3 mg·g^{-1} in the HMMs tested when aqueous pH is maintained above 11. In addition, the obvious deductions in the sorption capacity of the chemically regenerated HMMs were observed after four cycles of sorption-regeneration. However, the sorption capacities of intermediate substituted HMMs (0.3−0.7 CEC) can be completely restored by bioregeneration with yeast for extended cycles of reuse. The results imply that the bioregeneration method with yeast could be a promising technique for in-situ bioremediation of phenol-contaminated groundwater in the subsurface or for treatment of phenol containing wastewater.

Introduction

The immobilization of nonionic organic contaminants by natural montmorillonite can be enhanced by modifying this mineral with quaternary cationic surfactants, such as hexadecyltrimethylammonium (HDTMA). HDTMA ions can replace the native inorganic cations at the exchange sites between clay interlayers, forming a HDTMA-modified montmorillonite (HMM). Like other organoclay that is modified with surfactant, HMM has a much higher affinity to nonionic organic contaminants than the natural montmorillonite (*1*−*5*). This property of HMM, together with its rich natural deposit, makes this organoclay a promising sorbent to remove many species of organic contaminants from surface and groundwater (*6*). However, during sorption, HMM only transfers contaminants from liquid phase to solid phase and does not decompose them into harmless forms. Like other sorbents, HMM has its own sorption capacity for an organic compound. Once the sorption capacity is reached, HMM will not be able to sorb additional organic molecules from the aqueous solution. Therefore, it is of practical significance to remove the sorbed organic molecules from HMM and potentially recycle the regenerated HMM for contaminant removal.

Till now, the most widely used sorbent for removing contaminants from surface and groundwater is still activated carbon. A lot of regeneration methods, including thermal, vapor, wet air, chemical, and catalytic methods, to remove the sorbed contaminants from this sorbent have been reported (*7*−*9*). Additionally, Yoichi et al. (*10*) even studied the bioregeneration of trichloroethylene-adsorbing granular activated carbon (GAC) and found the adsorption capacity of the regenerated GAC was much lower than the fresh GAC, inferring that bioregeneration of GAC is not an effective approach to regenerate TCE-sorbing GAC.

The regeneration methods used in the previous studies, however, are not quite applicable for HMM. For instance, the use of high temperature and steam may destroy the structures of HMM and surfactants. After the treatments, the regenerated HMM loses surfactants. Therefore, the remodification of the reclaimed clay is required. Michot et al. applied a burning method to treat the chlorinated phenol sorbed to the modified clay and found all the sorbed chlorinated phenol molecules and surfactants were removed without altering the interlayer structure of the clay (*11*). After remodification with surfactants, this recycled clay can be repeatedly used for subsequent contaminant removal. In a similar study, Li and co-worker (*12*) successfully regenerated the organobentonite sorbing phenol and *m*-chlorophenol by burning the used organobentonite at 200 °C for 2 h. Although the thermal method has been proven to be an effective process to remove certain organic contaminants, it is not cost-effective for HMM regeneration because of the decomposition at elevated temperature of surfactant molecules, which are not the target substances to be removed. On the other hand, secondary pollutants may be generated during the thermal process. In view of this negative effect in thermal regeneration of organoclay, researchers have tried looking for other regeneration processes such as bioregeneration. Crocker and colleagues (*13*) studied the bioregeneration of a naphthalene-laden organosmectite and achieved a 50% naphthalene removal.

Although reports on bioregeneration of organoclay are very limited, biodegradations of many environmentally important contaminants in their aqueous solution have been extensively studied. Of the research, emphasis has been put on phenol-degrading microorganisms, such as *Pseudomonas* sp. and *Bacillus* sp. (*14*−*17*). Yeast has been also confirmed that is capable of degrading phenol. Lee et al. screened a species of yeast (*Yarrowia lipolytica* Y103) that can degrade the phenol at 47 mg·L^{-1} (*18*). In a similar study, Shivarova and Godjevargova found that *Trichosporon* sp. (*T. cutaneum*, *T. cutaneum*) can mineralize phenol from its aqueous solution as high as 1000 mg·L^{-1} (*19, 20*). The advantage of using yeast over bacteria in phenol degradation resides in its strong tolerance to a high concentration of phenol solution and large cell volume, the later of which favors greatly subsequent cell-water separation by gravity sedimentation. Furthermore, the single cell protein of yeast can be directly harvested during the process of phenol-containing wastewater treatment.

The objective of this study is to identify an effective method to regenerate HMM after adsorption with phenol. Chemical regeneration was investigated using NaOH solution, while biological regeneration was explored with a lab-screened

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yeast to look for the feasible techniques which can restore the sorption capability of the used HMM. The work described here is of practical significance to potential applications for phenol-contaminated water both in natural and engineered systems.

Materials and Methods

Materials. The natural montmorillonite with a cation exchange capacity (CEC) of 94.5 cmol·kg^{-1} was obtained from Nanjing Geology Research Institute. It contained hardly any organic matters. HDTMA ($(CH_3)_3NC_{16}H_{33}Br$) was purchased from Shanghai Reagent Company. All chemicals were of analytical grade or better quality.

The phenol-degrading yeast, *Pityrosporum* sp., used throughout this study, was originally screened in our lab from the activated sludge that had been acclimated in a lab-scale fluidized bed reactor for about 2 months. This yeast is able to metabolize phenol as sole carbon and energy source at aqueous phenol concentration as high as 1000 mg·L^{-1}.

Preparation of HMM. HMM was prepared by adding 100 g of natural montmorillonite to 1 L of 0.03−0.09 mol·L^{-1} HDTMA bromide solution based on the designed degree of surfactant modification for this clay. HMM suspension was equilibrated at room temperature with continuous stirring for approximate 5 h. HMM particles were then separated by gravity sedimentation followed by several cycles of washing with distilled water until no Br^- can be detected from the supernatant. The sample was then desiccated and ground in an agate mortar and finally allowed to filter through a 80-mesh sieve before use. The HMM generated in this way was then categorized by the extent of substitution of the total cation exchange capacity of the natural montmorillonite with HDTMA ions. For instance, 0.5 CEC HMM denotes a surfactant-modified montmorillonite that theoretically 50% of its ion exchange sites are exchanged by HDTMA ions.

Sorption of Phenol to HMM. In our preliminary experiment, the effect of solid concentration of HMM in the suspension on the removal of phenol from the aqueous solution was investigated. It was found that the removal rate of phenol increased with the HMM concentration. However, no substantial increase in phenol removal was observed if HMM concentration was maintained at 20 g·L^{-1}. So, this critical concentration was used throughout the experiments. To start a sorption experiment, we introduced 0.5 g of dry HMM or natural montmorillonite to a 150 mL flask containing 25 mL of phenol solution (pH 7.0). The flask was then shaken with 150 rpm for 120 min at 30 °C. The preliminary experiment also showed that sorption equilibrium can be reached within 90 min, and the loss of phenol through volatilization could be ignored if phenol concentration in the aqueous solution was lower than 1500 mg·L^{-1}. So, a 120-min incubation time was used to ensure sorption equilibrium, and the initial phenol concentration was confined to 100−1000 mg·L^{-1}. After the sorption experiment, the suspension was centrifuged at 4000 rpm for 10 min to separate HMM particles from the aqueous solution. All the separated HMM particles (still being wet) were directly used for the subsequent chemical sorption or bioregeneration tests without further treatment. The aqueous concentration of phenol was determined spectrometrically at 560 nm following the method reported by Banat (*21*). The detection limit for this method is about 0.05 mg·L^{-1}. The amount of phenol sorbed to HMM particles was calculated by the amount of differences of phenol in the aqueous solution between before and after the sorption. All tests were conducted in duplicate.

The Effect of pH on Phenol Sorption of HMM. Batch experiments were performed to identify the effect of pH on sorption capability of HMM. Briefly, 0.5 g of HMM was added to a 25 mL phenol solution of 800 mg·L^{-1}. The suspension was shaken with 150 rpm at 30 °C for 120 min to reach equilibration. The phenol-laden HMM was then separated following the procedures as described above. In this experiment, the separated phenol-laden HMM particles were resuspended in 25 mL of distilled water, and the final pH of the suspension was subjected to adjust to 8−13 as designed. To have a stable pH, the suspension was shaken with 150 rpm for 120 min at 30 °C, while 2 M NaOH was added. The amount of phenol sorbed to HMM at corresponding pH could be determined by the way as mentioned above.

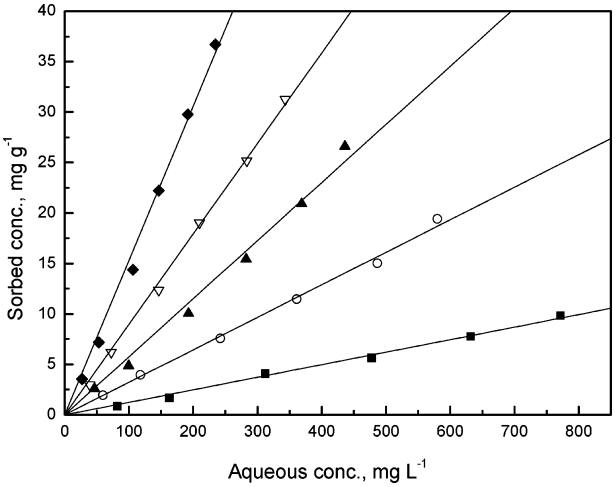

FIGURE 1. The sorption isotherms of phenol on HMMs at 0.0 CEC (■), 0.3 CEC (○), 0.5 CEC (▲), 0.7 CEC (▽), and 1.0 CEC (◆) in neutral circumstances.

Chemical Regeneration of Phenol-Laden HMM. To regenerate HMM chemically, we resuspended the wet phenol-laden HMM from the sorption experiment in 25 mL of distilled water. The pH of the suspension was then adjusted to 13 to facilitate desorption. The suspension was subsequently equilibrated for 120 min at 30 °C with shaking at 150 rpm. The HMM particles was then regenerated by separation with centrifugation as described above. The regenerated HMM particles could be used repeatedly to determine its sorption capacity of phenol.

Biological Regeneration of Phenol-Laden HMM. The phenol-sorbing HMM from the above sorption experiment was mixed with 5 mL of yeast solution with a biomass concentration of 598.6 mg·L^{-1} in 20 mL of distilled water. After appropriate nutrient salts (N, P) were added, the mixture was shaken with 150 rpm at 30 °C for a designed incubation period. In the process of the incubation, HMM particles were centrifuged, and the aqueous phenol concentrations were determined as described above in a manner of 12-h interval. The concentration of phenol sorbed on HMM was calculated using a formula derived from the sorption experiments (see the section of Sorption of Phenol to HMM). The biologically regenerated HMM could be then used directly in its wet form in the next cycles of sorption and bioregeneration experiments following the same procedures.

Results and Discussion

Sorption of Phenol to HMM. The effect of the modification degree of HMM with HDTMA on sorption of phenol from its neutral solution was investigated. As shown in Figure 1, of all the HMM tested, the sorptions of phenol from the aqueous solutions are well linearly regressed to the aqueous phenol concentration. Thus, the isotherms are linear. The higher the amount of HDTMA was exchanged on HMM, the greater the phenol-sorbing capacity of HMM is.

In some cases, the relationship between sorption of pollutants to organoclay and its aqueous concentration can

TABLE 1. Isotherms for Phenol Sorption on HMMs at Neutral pH

degree of substitution of HMMs	isotherm equations	correlation coefficients
0 CEC HMM	$C_s = 0.0124\ C_w$	0.998
0.3 CEC HMM	$C_s = 0.0322\ C_w$	0.998
0.5 CEC HMM	$C_s = 0.0576\ C_w$	0.996
0.7 CEC HMM	$C_s = 0.0897\ C_w$	0.999
1.0 CEC HMM	$C_s = 0.153\ C_w$	0.999

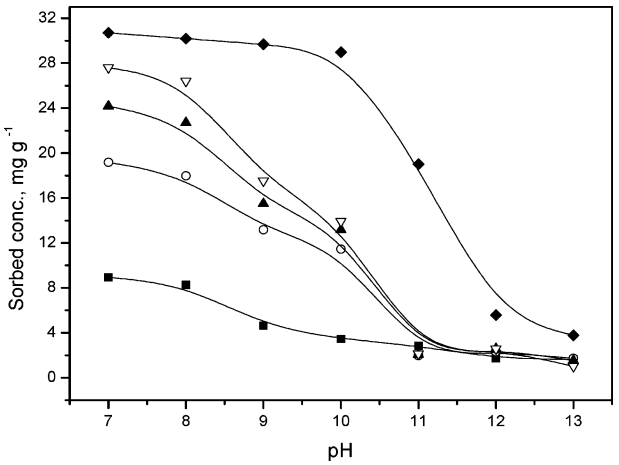

FIGURE 2. The amount of sorbed phenol on HMMs as a function of aqueous pH and degree of surfactant substitution at 0.0 CEC (■), 0.3 CEC (○), 0.5 CEC (▲), 0.7 CEC (▽), and 1.0 CEC (◆).

be described with eq 1 (*3*)

$$C_s = K \times C_w \tag{1}$$

where C_s is the amount of pollutant sorbed per gram of HMM ($mg \cdot g^{-1}$); C_w is the equilibrium pollutant concentration in the aqueous solution ($mg \cdot L^{-1}$); and K is the sorption coefficient.

In this study, our experimental data fitted the above equation. The sorption coefficients were listed in Table 1. The sorption coefficients of HMMs were found closely related to the extent of substitution of the CEC of the natural montmorillonite with HDTMA ions. The correlation between K and the degree of substitution (x) of the CEC can be described as $K = 0.102x^2 + 0.0389x + 0.0121$, where x is between 0 and 1.0.

Based on the above results, we hypothesize that a microhydrophobic phase between montmorillonite interlayers whose exchangeable ion sites were substituted with HDTMA was formed. This hydrophobic phase serves as an effective sorption location for protonated phenol. The hydrophobic interaction between the sorption location and phenol molecule is expected to become stronger as more ion exchange sites are substituted by HDTMA.

The differences between HMM and natural montmorillonite are their mechanisms of phenol removal. Phenol sorption of HMM from aqueous solution is mainly based on partitioning (*3*) with a higher degree of modification favoring to more sorption of the organic contaminant (*2*). Phenol removal by natural montmorillonite, however, is due to adsorption. As shown in Figure 1, phenol removal by adsorption with natural montmorillonite is much lower than that by sorption with HMM.

Effect of pH on Phenol Sorption of HMM. The impacts of pH on sorption capacities of HMM with different degrees of modification were investigated. As shown in Figure 2, of all the HMMs tested, the sorption capacities of HMMs decreased gradually between pH 7−9, while it declined dramatically between pH 10−11. For HMMs with modification within the range of 0−0.7 CEC, when the pHs were elevated to 11, the sorption capacities of HMMs all approached a minimum amount of about 3 $mg \cdot g^{-1}$; while for 1.0 CEC HMM, the sorption capacity reached 6 $mg \cdot g^{-1}$ at about pH 12. The observations can be explained by the deprotonation of phenol molecules (eq 2; pK_a 9.89). The hydrophobicity between phenol and its conjugated base is different. In alkaline solution, phenol undergoes deprotonation, generating its conjugate base and a proton. So, the aqueous solution with a higher pH contains a higher percentage phenol with conjugate base. The negatively charged conjugated base has a higher affinity to water, which has a lower hydrophobicity than the protonated phenol molecule. Hence, the amount of phenol partitioned to HMM is expected to decrease with increasing pH. Around the pK_a of phenol, the percentage of the deprotonated phenol in the solution changes markedly, resulting in substantial decrease in the sorption capacity of HMM. At much higher pH, the dominant form of phenol is deprononated phenol (conjugated base). Therefore, the phenol sorption capacity of the HMMs with 0−0.7 CEC all reached a common low amount. This residual adsorption of phenol may be caused by electrostatic interactions between the conjugated base and some positive charged ion sites on HMM surfaces.

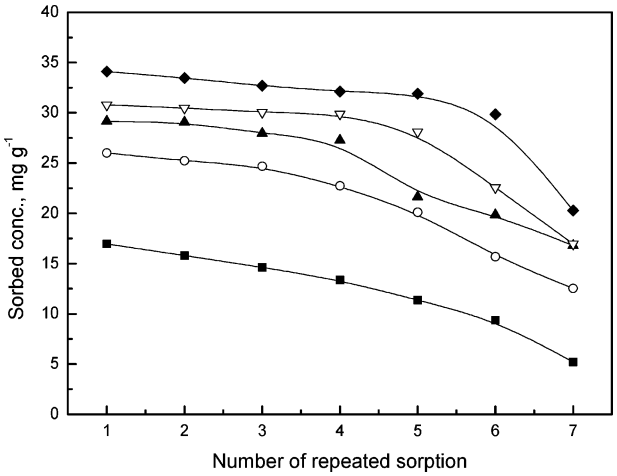

FIGURE 3. The phenol sorption capacities of HMMs as a function of chemical desorption cycles and the degree of surfactant substitution at 0.0 CEC (■), 0.3 CEC (○), 0.5 CEC (▲), 0.7 CEC (▽), and 1.0 CEC (◆) for up to seven sorption/regeneration cycles.

$$C_6H_5OH \leftrightarrow C_6H_5O^- + H^+ \tag{2}$$

The impact of pH on the phenol-sorbing capacity of HMM is the basis for chemical regeneration of HMMs. In general, in order for HMM to achieve the best performance, montmorillonite needs to be modified to have a high sorption capacity and low residual phenol when regenerated. As shown in Figure 2, although 1.0 CEC HMM had the highest sorption capacity, it also had a higher residual concentration of phenol (6 $mg \cdot g^{-1}$) at elevated pH, while 0.7 CEC HMM had the higher sorption capacity with only a limiting residual sorption of 3 $mg \cdot g^{-1}$. Therefore, 0.7 CEC HMM might be more significant than 1.0 CEC HMM in further practical applications.

Chemical Regeneration of HMM. The sorption capacity of chemically regenerated HMM was also examined in this study. Figure 3 showed the effects of desorption cycles on the performance of the regenerated HMM. As depicted in Figure 3, the obvious deduction in the sorption capacity of the chemically regenerated HMM was observed after four cycles of sorption-regeneration. The reason for this deduction

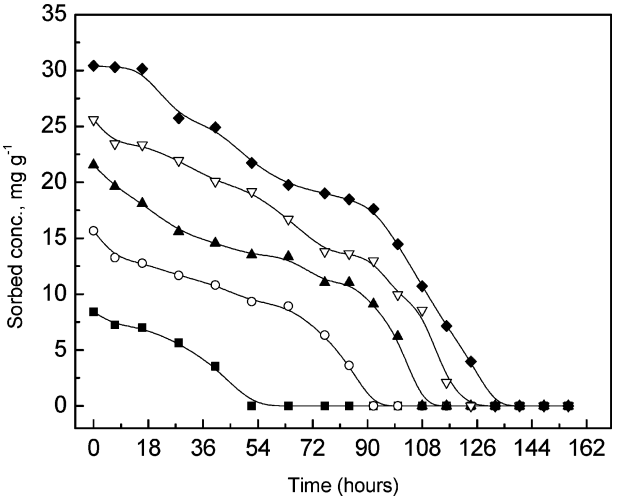

FIGURE 4. The amount of sorbed phenol on HMMs as a function of contact time with yeast and degree of surfactant substitution at 0.0 CEC (■), 0.3 CEC (○), 0.5 CEC (▲), 0.7 CEC (▽), and 1.0 CEC (◆) in neutral circumstances.

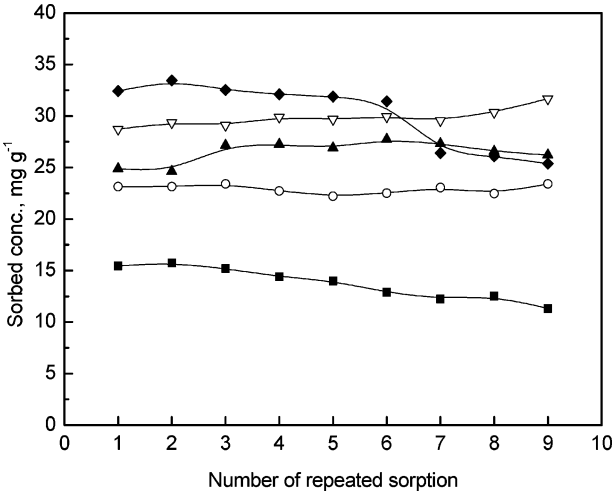

FIGURE 5. The phenol sorption capacities of HMMs as a function of biological regeneration cycles and degree of surfactant substitution at 0.0 CEC (■), 0.3 CEC (○), 0.5 CEC (▲), 0.7 CEC (▽), and 1.0 CEC (◆) for up to nine sorption/regeneration cycles.

in the sorption capacities of HMM after chemical regeneration may be due to the alteration of the surface property of original HMM. After multiple cycles of chemical treatments, more and more deprotonated phenol might be incorporated into the interlayers of montmorillonite, resulting in the decrease of the sorption capacity.

Based on the above results, the chemical regeneration is expected to be a useful method for treatment of phenol-containing wastewater in engineered systems (in which the pH is controllable) as long as recycles of HMM are within 4 times. However, since it is difficult to control pH in natural systems, the method of chemical regeneration for phenol-sorbing HMM may not have any practical applications in natural systems such as in-situ remediation of groundwater contaminated with phenol.

To seek regeneration methods with wide applications it is necessary to explore new techniques, such as the biological method to effectively reclaim phenol-sorbing HMM.

Bioregeneration of Phenol-Sorbing HMM. Since there are no secondary pollutants generated and the degradation of the sorbed phenol molecules is complete during the bioregeneration, studies on biological regeneration of phenol-sorbing HMM is important. In this study, *Pityrosporum* sp., a strain of yeast, was chosen to regenerate phenol-sorbing HMM. When mixed with suspension containing phenol-sorbing HMM particles, *Pityrosporum* sp. can metabolize phenol to produce carbon dioxide and water. As shown in Figure 4, the concentration of phenol residual in all the HMMs tested decreased gradually with increasing incubation time with the yeast. *Pityrosporum* sp. could effectively mineralize almost all the sorbed phenol molecules as long as enough incubation time was allowed for yeast to contact with the HMMs even at a higher degree of modification. Therefore, bioregeneration is advantageous over chemical regeneration for sorbed phenol removal.

As shown in Figure 1, an equilibrium exists between the aqueous and the sorbed phenol. Mineralization of phenol in the aqueous solution by yeast broken up of the equilibrium and the lower concentration of phenol in the aqueous phase serves as a driving force to continuously desorb phenol from HMM until almost all the sorbed phenol molecules are desorbed and mineralized. Because of the complete mineralization of phenol after incubated with yeast as manifested in Figure 4, we believed that *Pityrosporum* sp. was able to degrade both protonated and depronated phenol forms. Therefore, the overall removal efficiency of phenol by yeast might be independent of a degree of modification by surfactant molecules. Compared with the chemical regeneration method, bioregeneration employed the HMM with the higher degree of modification that has the higher sorption capacity. That the yeast has the higher sorbed phenol removal efficiency makes the bioregeneration method a promising technology to regenerate phenol-sorbing HMM.

Based on the above results, the bioregeneration method with yeast *Pityrosporum* sp. could make the two-stage-bioremediation including HMM sorption and bioregeneration of phenol-laden HMM a promising technology to remediate phenol-contaminated environmental systems. When cultured without natural montmorillonite, microorganisms are extremely sensitive to the toxicity of dissolved HDTMA (*3*). However, when cultured with HMM, HDTMA's toxicity to organisms would be substantially suppressed as long as the exchanged HDTMA is lower than 1.0 CEC since HDTMA is exchanged in the interlayer of HMM (*3*). Hence, when cultured with HMM that has a low amount of surfactants, microorganisms would maintain a high level of activity (*22*). For in-situ remediation of groundwater contaminated with phenol, a barrier packed with HMM and yeast could be constructed to block the dissolved phenol from further migration. As long as sufficient hydraulic retention time is administrated, phenol in the intercepted contaminant plume could be degraded continuously by yeast, while HMM serves as an effective sorbent and reservoir for phenol sorption and releasing, which can be tailored to optimal bioremediation.

Biological Regeneration of Recycled HMM. The impact of bioregeneration cycles of HMM on its sorption capacity was investigated. As shown in Figure 5, the performance of natural montmorillonite and 1.0 CEC HMM could be reused at nearly a constant level within 6 times. For intermediate substituted HMMs (0.3−0.7 CEC), however, their sorption capacities could be completely recovered by bioregeneration for extended cycles of reuse. So, bioregeneration is much more effective than chemical regeneration in phenol removal from phenol-sorbing HMMs.

In summary, the results of this study showed the feasibility of chemical and biological regeneration of phenol-sorbing HMM. HMM can sorb phenol effectively, and the biological regeneration with *Pityrosporum* sp. has advantages over chemical regeneration for reclamation of phenol-laden HMM. The sorption capacities of the intermediate exchanged HMMs (0.3−0.7 CEC) can be completely restored by repeated

bioregeneration. Since the sorption capability of 0.7 CEC HMM is higher than that of 0.3 or 0.5 CEC HMM, a two-stage technique combining 0.7 CEC HMM sorption and biological regeneration appears promising for in-situ remediation of ground and surface water or for treatment of phenol from engineered systems.

Acknowledgments

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