



The effect of temperature on hydrocarbon adsorption by diphenyldichlorosilane coated zeolite and its application in permeable reactive barriers in cold regions



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ABSTRACT

Zeolite was modified and characterized for its potential application in permeable reactive barriers tasked with fuel removal. Characterization was accomplished to compare the material structure before and after the modification process. Batch sorption tests were performed to detect hydrocarbon capture performance, and regeneration experiments were conducted to determine longevity. Adsorption isotherms and thermodynamics were studied to determine the mechanism of hydrocarbon capture.

Diphenyldichlorosilane (DPDSCI), a unique reactive chlorosilane with two phenyl groups, was selected to coat the zeolite. The surface characterization results revealed that the modification was successful and the inner structure of zeolite maintained its porosity. Hydrocarbon adsorption tests presented good toluene adsorption performance even under cold temperatures. Regeneration experiments indicated this modified zeolite could be used multiple times without a significant reduction in adsorption efficiency. The Langmuir adsorption isotherm fitted toluene adsorption well and the process occurred spontaneously. Based on this performance, this modified zeolite was found suitable for future application for groundwater remediation in cold regions.

1. Introduction

Worldwide there are many reports of serious groundwater pollution resulting from accidental spills or improper disposal, including in cold regions (Bargagli, 2008). In attempts to clean-up groundwater and restore contaminated sites to near pristine conditions, scientists started to develop technologies such as the pump-and-treat method in the 1980's (Thiruvengkatachari et al., 2008). In the last decade, permeable reactive barriers (PRBs), filled with reactive materials to intercept and decontaminate groundwater plumes have become one of the most promising remediation technologies (Obiri-Nyarko et al., 2014). Compared to traditional pump-and-treat systems, PRBs generally have a longer service life and lower operational cost (Bortone et al., 2013a). In Antarctica, similar practices were also conducted to treat petroleum-contaminated and heavy-metal-laden sites (Mumford et al., 2013) and at this stage, the major areas of research to improve PRB performance include improving the longevity of the reactive materials used in these systems and their adsorption capacity towards various contaminants (Bortone et al., 2013b; Erto et al., 2014; Santonastaso et al., 2015; Snape et al., 2001). Materials that rely solely on chemical or physical processes have a limited operational life as once the materials are saturated, they will

no longer perform as required (Obiri-Nyarko et al., 2014). The combination of bioremediation and physicochemical adsorption is a more promising and viable method (Freidman et al., 2016). As long as microbes are present and stimulated, the system will operate (Thiruvengkatachari et al., 2008). Previous tests have demonstrated that indigenous microorganisms present in soil and water can migrate and thrive, on and between adsorbent materials in PRBs and degrade the hydrocarbons as they migrate through the system (De Jesus et al., 2015).

To enhance the bioremediation process, improving the reactive materials in PRBs is essential (Scherer et al., 2000). Among the available options, modified zeolites may present better characteristics than granular activated carbon (GAC) despite of its high adsorption capacity (Bortone et al., 2013b; Erto et al., 2017). Contaminants are not bonded as tightly onto modified zeolite surfaces, and so are more easily utilised by the microbes present (Mumford et al., 2015; Zhang et al., 1991). Zeolites have a rigid three dimensional structure of aluminosilicate and a highly microporous surface and a net negative charge that is balanced by electrostatically held cations (Torabian et al., 2010). Thanks to its unique structural characteristics and surface components, natural zeolite is usually used to capture target cations through ion exchange, with

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Table 1
Previous research on modified natural zeolite and synthesized zeolite.

Base material	Modification material	Target	Capacity			Calculated capacity (using Langmuir isotherm q_m) ($\mu\text{mol/g}$)	Reference		
			Linear K_d (L/kg)	Langmuir q_m (mg/g)	Freundlich K_f ($\text{mg}^{1-n} \text{g}^{-1} \text{L}^n$)				
Clinoptilolite	Diphenyldichlorosilane (DPDSCI)	Toluene			0.02	5.16	(Huttenloch et al., 2001)		
		Xylene			0.05	8.90			
		Naphthalene			0.15	22.32			
	Octadecyltrichlorosilane (C18)	Naphthalene	17.30				(Northcott et al., 2010a)		
		Xylene	9.70						
	Hexadecyltrimethylammonium (HDTMA)	Benzene	79.21	16.61	0.39	212.95	(Ghiaci et al., 2004)		
			43.25	11.35	0.17	120.74			
		Aniline			0.13		(Ersoy and Celik, 2004)		
		Nitrobenzene			1.25				
		Chlorophenol		12.71	0.06	98.86			
n-Cetylpyridinium bromide (CPB)	Benzene	103.4	23.07	0.36	295.77	(Ghiaci et al., 2004)			
	Phenol	47.41	11.88	0.22	126.38				
Benzyltetradecyl ammonium chloride (BDTDA)	Phenol		1.30	0.05	13.81	(Kuleyin, 2007)			
	Chlorophenol		6.41	0.77	49.86				
Stearyldimethyl benzyl ammonium chloride (SDBAC)	Atrazine		0.43		2.00	(Lemic et al., 2006)			
	Lindane		0.99		3.40				
	Diazinone		1.35		4.40				
Octadecyldimethyl benzyl ammonium (ODMBA)	Ochratoxin A		3.41		8.45	(Daković et al., 2003)			
	Fumonisin B ₁		7.35		10.18				
Synthetic zeolite	Octylmethylchlorosilane	Toluene		215.60		2340	(Song et al., 2005)		
		Hexadecyltrimethylammonium (HDTMA)	Benzene	48.53	7.74	0.40		99.26	(Ghiaci et al., 2004)
			Toluene	48.66	9.33	0.39		101.43	
	Phenol		29.02	7.42	0.15	78.96			
	n-Cetylpyridinium bromide (CPB)	Benzene	82.46	14.95	0.39	191.67	(Ghiaci et al., 2004)		
		Toluene	80.51	16.44	0.48	178.70			
		Phenol	34.23	9.425	0.14	100.27			
	Hexadecyltrimethylammonium bromide (CTAB)	TPH				92.30%	(Saremnia et al., 2016)		

no applications in anions and organics adsorption (Mahabadi et al., 2007; Widiastuti et al., 2008). Research has focused on the development of modified zeolites with higher hydrocarbon adsorption capacities compared to natural zeolite (Wang and Peng, 2010). One of the most frequent methods is the use of cationic surfactants that are bound to the external zeolite surface via electrostatic forces. Examples include hexadecyltrimethylammonium bromide (HDTMA) or cetylpyridinium bromide (CPB) (Wang and Peng, 2010). This treatment reduces or reverses the charge of zeolite and thus leads to anions or non-polar organics being captured on the surfactant coating (Wang and Peng, 2010). This process has been extensively investigated and presented good outcomes compared with natural zeolite in removing soluble hydrocarbons from groundwater with a summary of the results presented in Table 1 (Bowman, 2003; Ghiaci et al., 2004; Kuleyin, 2007; Li et al., 2000; Michael Ranck et al., 2005). Despite this promising performance, cationic surfactants like HDTMA or CPB still face many challenges, such as the loss of the surface coating and the disintegration of the zeolite particle itself (Michael Ranck et al., 2005). Therefore, alternative techniques are needed. Here chlorosilane is selected as it can be covalently bound to the surface of zeolite thereby improving the material stability while still enhancing its adsorption capacity.

Chlorosilane is a reactive chemical compound with up to three nonpolar aliphatic or aromatic functional groups which can be covalently grafted to the silanol groups on a mineral adsorbent surface

(Huttenloch et al., 2001). This treatment provides a stable chemical linkage between organosilane and the substrate instead of the weaker electrostatic interactions of cationic surfactants, which greatly reduces the loss of hydrophobic coating during its use (Northcott et al., 2010a). Previous research has shown that octadecyltrichlorosilane modified natural zeolite possesses high capacity in removing dissolved o-xylene and naphthalene from water (Northcott et al., 2010a) and the octylmethylchlorosilane coated synthetic zeolite possesses a similar toluene adsorption capacity to GAC (Arora et al., 2011; Song et al., 2005). In addition to the use of zeolites as base materials, there are studies regarding the grafting of chlorosilane onto GAC to enhance 2,4-dichlorophenol removal, by improving its hydrophobicity in permeable reactive barriers (Yang et al., 2010). Though chlorosilane is less likely to be leached off from the surface of the substrate, one of the most significant challenges is finding the most efficient organosilane to enhance sorption (Huttenloch et al., 2001). In previous work, chlorosilane with phenyl headgroups have been shown to have better affinity for aromatic compounds compared with others (Huttenloch et al., 2001), and therefore can be viewed as potential coating material to enhance hydrocarbon capture.

The basic silanization process (Huttenloch et al., 2001) for surface modification on zeolite is shown in Fig. 1. In the presence of pyridine and elevated temperatures, the chlorine on silicon compounds has a high affinity to the hydrogen of hydroxyl groups on zeolite surface. This

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