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The performance of diphenyldichlorosilane coated ammonium exchange zeolite and its application in the combination of adsorption and biodegradation of hydrocarbon contaminated ground water

Junchao Ma, Geoffrey W. Stevens, Kathryn A. Mumford*

Particulate Fluids Processing Centre, Department of Chemical and Biomolecular Engineering, The University of Melbourne, VIC 3010, Australia

HIGHLIGHTS

- Modified zeolite possessed good toluene adsorption and ammonium release ability.
- Toluene concentrations do not impact ammonium release from this material.
- Potassium concentrations influence toluene adsorption by this material.
- Langmuir isotherm fitted the toluene adsorption process best.
- This material possessed good regeneration and long-term stability.

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ABSTRACT

To enhance biodegradation in Permeable Reactive Barriers, Diphenyldichlorosilane (DPDSCl) coated ammonium zeolite was selected as a reactive material for development as it undergoes reversible hydrocarbon adsorption and may release nutrients in a controlled manner. To investigate its hydrocarbon adsorption and nutrient release behaviour under varying conditions, batch tests were conducted by immersing the material into different concentrations of toluene and potassium chloride solutions.

Results showed that the ammonium release ability was not significantly influenced by a change in toluene concentration, while the toluene adsorption performance was affected by fluctuations in potassium concentration. The overall ammonium release capacity of the modified zeolite was 0.8 mmol/g, and the toluene adsorption capacity was 9.355 μmol/g without KCl in solution. The Langmuir isotherm fitted the adsorption isotherms well, indicating that the adsorption mechanism of DPDSCl coated ammonium zeolite is the same as that of DPDSCl coated natural zeolite: a monolayer and homogenous surface adsorption process. Regeneration tests showed that this material may be used, cleaned and reused at least three times without a significant reduction in performance.

1. Introduction

Accidental spills during transportation, or improper use and disposal of chemical waste from industrial facilities has led to an increasing number of contaminated sites, including both soil and groundwater [1]. Among them, petroleum hydrocarbons and heavy metals are the main recognised pollutants [2]. Biological, physical and chemical technologies are used to treat pollutants and reduce the potential hazards to safe and acceptable levels [3]. Conventional methods include pump and treat systems [4] that extract and intensively treat contaminated groundwater, and air sparging [5] that inject air to volatilize contaminants as well as promote degradation. Though these methods are beneficial, they have limitations and are not applicable to all environmental conditions [2]. As such, new technologies, including Permeable Reactive Barriers require development. These systems have been proven effective for use in extreme environmental conditions, including low temperatures [6]. To promote the efficiency and longevity of Permeable Reactive Barriers, physiochemical adsorption may be combined with in-situ bioremediation for special applications [7].

Current limitations of groundwater bioremediation in temperate regions are insufficient nutrient concentrations [8], inaccessibility of the contaminant to microbial communities, and unfavourable environmental conditions in contaminated sites [9]. Additionally, due to temporal hydraulic fluctuations, rapidly changing environmental

* Corresponding author.
E-mail addresses: junchaom@student.unimelb.edu.au (J. Ma), gstevens@unimelb.edu.au (G.W. Stevens), mumfordk@unimelb.edu.au (K.A. Mumford).

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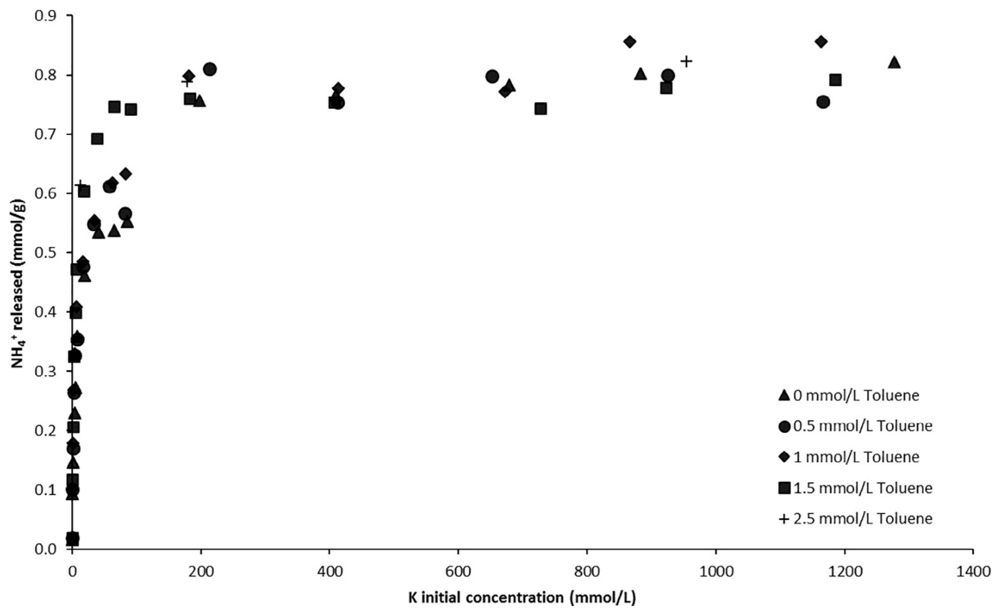

Fig. 1. Ammonium release of DAZ at different initial concentration of potassium and toluene.

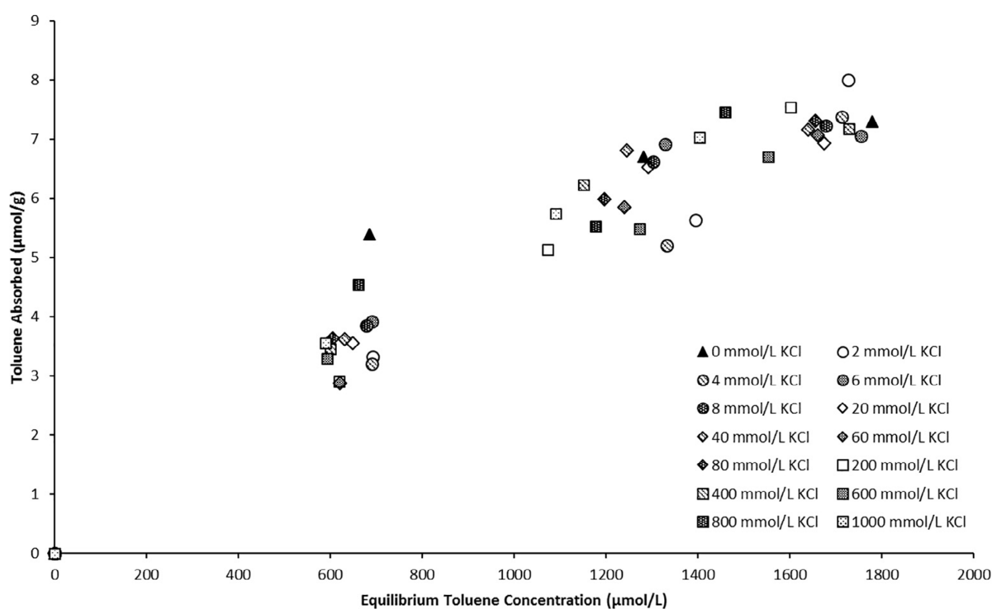

Fig. 2. Toluene adsorption of DAZ at different initial concentration of potassium and toluene.

conditions, and a transient supply of pollutants, the growth of contaminant degrading populations is further hindered [10]. Therefore, in most instances, major microbial communities are constrained or are dormant [11].

In temperate regions scientists are also developing and applying new technologies such as bioelectrochemical approaches [12,13], biosurfactants [14] and bioreactors [15] to enhance biodegradation efficiency. However, these methods require further testing at a laboratory and pilot scale before large scale field application would be appropriate [16]. Materials that are currently used in Permeable Reactive Barriers (PRBs) to enhance the performance of in-situ bioremediation include Granular Activated Carbon (GAC) and nutrient amended zeolite. GAC is used to capture hydrocarbons which may be subsequently used as a food resource for microbial communities and ammonium exchanged zeolite is used to provide nutrients [7]. However, due to irreversible hydrocarbon adsorption onto GAC, the captured hydrocarbons are not easily accessible to the microbial communities for degradation. Thus, when saturated, the GAC requires replacement [17,18].

Therefore, it is necessary to develop new materials that can capture hydrocarbons when the concentration is high and desorb at low concentrations. An additional benefit would be the release of nutrients when required. Previous work has shown that chlorosilane coated zeolite is able to adsorb hydrocarbons and may be easily regenerated across a temperature range of 4 °C–20 °C [19]. The performance was modelled using well established modelling practises [19], and the material in column tests remained stable after three regeneration cycles [20]. Furthermore, zeolite amended with ammonium may be used as a nutrient source across conditions applicable to remediation processes [21].

In this study, the combined performance of hydrocarbon adsorption and nutrient release of cholorosilane coated ammonium zeolite is tested. To prove the materials may be reused, regeneration experiments are conducted.