The potential application of natural zeolite for greywater treatment
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Received 20 November 2006; accepted 16 February 2007

Abstract

Natural zeolites are good potential material for water and wastewater treatment. It is due to the advantages of low cost, ion-exchange and adsorption capability of the natural zeolites. It can also be modified and regenerated. This paper thus looks at efforts made in exploring the potential application of natural zeolites and modified natural zeolites in water and wastewater treatment especially for reducing or removing contaminants in greywater, a type of wastewater originated from bathrooms and laundries in household. The material to be reviewed in this paper includes (i) major greywater contaminants and (ii) the potential of natural zeolites for greywater treatment.

Keywords: Natural zeolite; Greywater; Treatment; Reuse

1. Introduction

Greywater, a type of wastewater originated from bathrooms and laundries in households, office buildings, hotels, and schools, etc, excluding wastewater from kitchen and toilet flushing [1], is an alternative water source for reuse due to the high volume of domestic wastewater produced each day. About 50–80% of domestic wastewater originated from dishes, showers, sinks and laundries [2]. Therefore, it is potentially feasible for reuse especially for non potable uses both for indoor applications, such as toilet flushing which accounts for around 20–30% of the total household water usage, usually of the drinking water quality [1,3] and outdoor applications, such as lawn watering which comprises about 34% of the...
There are a number of health and regulatory issues facing the reuse of untreated greywater. The risk of spreading of diseases because of microorganisms in the water can be dangerous by ingestion and through hand or mouth contact, especially pathogens due to faecal indicator bacteria [1]. The risk of pollution of soil and receiving waters is another problem owing to accumulation of chemical contaminants such as phosphate and xenobiotic organic compounds (XOC) from chemical product used in households [1]. Therefore, reuse of greywater has to overcome potentially adverse health effect, environmental impact as well as technical and economic feasibilities due to the complex nature of the contaminants in grey water. A method that produces satisfactory quality of treated greywater with an affordable cost is always a challenge.

Natural zeolites are good potential material for water and wastewater treatment. The potential advantages for application of natural zeolites in water and wastewater treatment are promising. Firstly, a benefit gained by using natural zeolites for water and wastewater treatment is their ability to undergo ion-exchange and adsorption. In addition, natural zeolites can be treated and modified in order to trap contaminants in water and wastewater. Also, natural zeolites exist abundantly; low cost and only low technology systems are required. An additional benefit of using natural zeolites is their regenerative properties.

This paper reviews the major greywater contaminants and looks at efforts made in exploring the potential application of natural zeolites and modified natural zeolites in water and wastewater treatment especially for reducing or removing contaminants in greywater.

2. Major greywater contaminants

This section reviews the major greywater contaminants by looking at the sources of contaminants in household.

2.1. Chemical contaminants

Significant chemical contaminants in greywater are derived from chemicals used in household laundries and bathrooms. An understanding of the sources of contaminants will help to identify the heterogenous chemical contaminants in greywater.

First of all, the major chemical contaminants found in laundry and bathroom wastewater are surfactants (surface active agent) since they are used as the main ingredient in household products such as laundry detergents and hygiene products to provide cleaning action. Surfactant can be classified based on the nature of the chain-carrying part of their molecular structure when dissolved in water, namely anionic, cationic, non-ionic or amphoteric. The most common surfactant used in detergents for laundry and general cleansing is anionic surfactants [4]. Linear alkylate sulfonate or linear alkylbenzenesulfonate (LAS) is the most common anionic surfactant found in laundry detergents due to their very interesting foaming characteristics that can be controlled easily by foam inhibitors, low prices, well formulated in all-purpose surfactant formulas, biodegraded and easily analysed [5]. Another common surfactant is cationic surfactants such as quaternary ammonium salts, dialkyldimethylammonium chlorides, distearyldimethylammonium chloride and alkylidimethylbenzylammonium chlorides that are used in fabric softeners and laundry disinfecting agents [4–6]. They constitute a source of ammonium contaminants in greywater.

With respect to consumer needs, surfactants are commonly combined with other ingredients such as builders. Detergent builder is another major ingredient in household chemical product. They play an important role to build the effectiveness of the detergent formulation [5]. The
builder function is in-activate calcium and magnesium ions as water hardness ions. There are several types of builders which are alkali substances, complexing agents, and ion exchange [4,5]. The alkalis (monophosphate) and phosphorus complexing agent such as sodium tripolyphosphate (STPP) are the main source of phosphate contaminant in greywater.

In relation to measurement of the traditional wastewater parameters such as biological oxygen demand (BOD) and chemical oxygen demand (COD), it is reported that, COD in greywater is much greater than BOD. The COD:BOD ratio can be as high as 4:1 [7,8]. It is believed that the high levels of XOC will result in higher level of COD in the greywater [1,8]. XOC is a heterogenous group of compounds found in common household products such as surfactants, builders, bleaches, softener and solvents [4,5]. Other measurements of the traditional wastewater parameters are nitrogen and phosphate. The nitrogen source in greywater is mainly ammonium rather than nitrate and nitrite [1]. The source of ammonium in greywater particularly in laundry wastewater may be from the use of cationic surfactants as fabric softeners and laundry disinfectant agents. The primary source of phosphates found in greywater is from detergent builders used in washing detergents [1].

2.2. Micro-organism contaminants

Micro-organisms found in greywater are faecal contaminants from mainly hand washing after toilet use and diaper changes of babies. The major indicator of faecal contaminants is Escherichia coli as recommended by the WHO Guidelines [9]. E. coli is abundant in human and animal faeces and also found in sewage, treated effluent and all natural waters. Another indicator of faecal contaminant with regard to the coliform bacteria is total or faecal coliform. It is due to their correlation with the possible presence of bacteria, virus, protozoa and other pathogens [3]. Moreover, the traditional indicator of faecal contaminant belong to the genera of Citrobacter, Enterobacter, Escherichia and Klebsiella [9]. Therefore, total or faecal coliforms have long been recommended as a suitable microbial indicator of drinking water quality as well as treated wastewater, and it is easy to detect.

3. The potential application of natural zeolite for greywater treatment

This section looks at efforts made in exploring the potential application of natural zeolites in water and wastewater treatment especially for reducing or removing contaminants in water and wastewater that are found as significant contaminants in greywater. The reviewed material to be presented includes: (a) zeolites to remove cation contaminants, (b) modified natural zeolite to remove anion contaminants and (c) natural zeolite and modified natural zeolite to remove microorganisms.

3.1. Removal of cation contaminants

Zeolites are composed of three dimensional frameworks of aluminosilicate tetrahedral where the aluminum and silicon structure atoms are bound by covalent bonds over shared oxygen atoms to form interconnected cages and channels [10]. Each aluminum (Al³⁺) atom substitution for silicon (Si⁴⁺) in the zeolite framework generates one negative charge on the framework. The greater the aluminum atom substitutions, the higher the negative charges of zeolite [11]. The negative charges within the pores are balanced by positively charged ions (cations) such as Na⁺, K⁺, Ca²⁺, Mg²⁺ on the pore surfaces of the zeolite [12]. These cations are bound to the aluminosilicate structure by weaker electrostatic bonds that cause ability to exchange with certain cations in solutions [13]. With this unique structure, zeolites possess high cation-exchange capacity.

As a cation exchange agent, zeolites have been widely investigated and applied to remove cation contaminants in waters and wastewaters. It is due
to the fact that zeolite has negative charge within the pores balanced by positively charged ions such as Na\(^+\), K\(^+\), Ca\(^{2+}\), and Mg\(^{2+}\) with weaker electrostatic bonds [12]. Those cations, therefore, can be easily exchanged with cations such as NH\(_4\)\(^+\) in waters and wastewaters [11–22], Cs\(^+\) and Sr\(^{2+}\) in nuclear wastewaters [23–25] as well as heavy metals in waters and wastewaters [26–32].

Ammonium is the most common cation in waters and wastewaters affecting human and animal health. It is also found as one of major greywater contaminant especially originated from bathroom. The performance of natural zeolites from many countries to remove ammonium have been investigated widely such as using natural zeolites from New Zealand [11], Australia [17,33–35], Turkey [21], Hungarian [36], Chilean [10], Croatian [13], and Chinese [16]. It is believed that structural characteristic of natural zeolite used (e.g. mordenite, clinoptilolite, erionite, chabazite and phillipsite) affects the performance of natural zeolite [37]. Several studies reported the effective use of natural zeolite for ammonium removal (Table 1). Reviewing Table 1, it appears that natural zeolites have their greatest potential in removing ammonium.

Industrially, natural zeolites have been applied in drinking water production, municipal, agriculture, and industrial wastewaters treatment. The plant at Lake Tahoe, California, for instance, uses several hundred tons of clinoptilolite-rich material from the Hector deposit in California to treat municipal wastes with 27,000 m\(^3\)/d capacity [23]. In Virginia, USA, two plants with 45,000 and 245,000 m\(^3\)/d capacities using clinoptilolite are used for municipal wastewater treatment [23].

Regeneration of ammonium-zeolite is an important process so that the zeolite may be reused. 1 N NaCl or KCl solutions can be used to regenerate an ammonium-exchanged zeolite with the exchange site Na\(^+\) and K\(^+\). The regeneration efficiency has been achieved at pH above 11 by addition of lime [23]. Another regeneration of ammonium-exchanged zeolite is biological method [23,38]. The mechanism of this regeneration is a nitrification process. Nitrifying bacteria converted NH\(_4\)\(^+\) to NO\(_3\)\(^–\) on the surface of zeolite in oxygen-enriched air. Rahmani and Mahvi [38] reported that the efficiency of bioregeneration was 87.7–99.8% in period of 3.5–5.5 h. Heating process is also an alternative method in regeneration. Heating used zeolite between 350–450ºC in a stream of air can evolve ammonium from exchange sites [23]. After devolatilization of ammonium, H-exchange sites are formed and the zeolite can be reused. However, this method demands excessive energy, thus it is not generally used in water treatment.

Other cation contaminants removed by natural zeolites are radioactive cations such as Cs\(^+\) and Sr\(^{2+}\). 80–100% of \(^{137}\)Cs was removed in static batch processes and 40–80% in dynamic flow-through column systems with adsorption capacity for clinoptilolite-rich tuff 4×10\(^–6\) Ci/kg [23]. In application, at installations of the British Nuclear-Fuels and Central Electricity Generating Board
Table 1
Removal of some contaminants in water and wastewater using zeolite

<table>
<thead>
<tr>
<th>Contaminants</th>
<th>Zeolite performance</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_4^+$ in water and wastewater using natural zeolite from many countries</td>
<td>30–98% removal depending upon the type of zeolite used, contact time, initial concentration, temperature, and amount of zeolite loading and particle size</td>
<td>[10, 11, 13, 16, 21, 33, 34, 36]</td>
</tr>
<tr>
<td>Cs$^+$, from the discharge water of nuclear power plants in the Trans-Carpathian area of Ukraine</td>
<td>80–100% removed in static batch processes and 40–80% in dynamic flow-through column systems</td>
<td>[23]</td>
</tr>
<tr>
<td>Mn$^{2+}$ from drinking water</td>
<td>67% more reduced by K-clinoptilolite from northern Romania</td>
<td>[7]</td>
</tr>
<tr>
<td>Hg$^{2+}$</td>
<td>Nearly 50% Hg$^{2+}$ removed using Na-clinoptilolite and heulandite</td>
<td>[32]</td>
</tr>
<tr>
<td>Metals such as Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn from the composting process in Psittallia, Athens</td>
<td>10–100% removed using natural zeolite</td>
<td>[40]</td>
</tr>
<tr>
<td>PO$_4^{3-}$</td>
<td>90% removed using cetylpyridinium-modified-zeolite with 80 meq/100 g dosage of cetylpyridinium chloride and pH 10 at initial phosphate concentration of 10 mg/L.</td>
<td>[39]</td>
</tr>
<tr>
<td>Arsenate from the simulated leachate</td>
<td>95% removed at all concentrations tested.</td>
<td>[40, 41]</td>
</tr>
<tr>
<td>Total coliforms, faecal coliform and faecal streptococci</td>
<td>100% removed at the tenth day for faecal coliform and seventeen days for faecal streptococci at particle size between 0.5 mm and 2.0 mm using natural phillipsite columns in a percolation reactor at a constant solution flow rate</td>
<td>[42]</td>
</tr>
<tr>
<td>COD and BOD</td>
<td>75% COD retention at the twenty-fifth day and 100% BOD in the ninth day using natural phillipsite columns in a percolation reactor at a constant solution flow rate</td>
<td>[42]</td>
</tr>
<tr>
<td>E. coli and bacteriophage MS2</td>
<td>100% removed using HDTMA-modified zeolite, particularly clinoptilolite rich tuff from the St. Cloud, New Mexico.</td>
<td>[43]</td>
</tr>
<tr>
<td>E. coli from drinking water</td>
<td>100% removed by clinotsid, a modified clinoptilolite with amine groups-polyhexamethylene guanidine chloride (PHMG) linked with epichlorohydrine, (Coli index &lt;3) depending upon the filtration flow rate and the particle size of the organozeolite</td>
<td>[44]</td>
</tr>
</tbody>
</table>

In United Kingdom, clinoptilolite is used to treat pond waters [23]. Another application, zeolite from the Trans-Carpathia area, 500,000 tons was used after the Chernobyl accident in attempting to decontaminate rivers polluted with Cs$^+$ and Sr$^{2+}$ [23]. In addition, clinoptilolite rich tuff from the Hector deposit in California was used to remove $^{137}$Cs and $^{90}$Sr from the low level radioactive wastewater of nuclear power plants. The advantages of using natural zeolites for nuclear wastewater are their resistance to degradation in the presence of ionizing radiation, their low solubility and it can be used for long term storage of long-lived radioisotopes by drying a zeolite exchange with radionuclide cations at 200ºC and sealing in a stainless steel container [23].

Moreover, heavy metal cations such as Pb$^{2+}$, Cu$^{2+}$, Cd$^{2+}$, Zn$^{2+}$, Co$^{2+}$, Ni$^{2+}$, Hg$^{2+}$, Fe$^{3+}$ and Ag$^+$ from various types of wastewaters have been investigated for their removal using natural zeolites [26,27,31,32,45]. Natural mordenite, for instance, has been investigated to remove heavy metals from spiked motorway storm water. The mordenite reduced heavy metal concentration from...
42 to 89% in synthetic solution and 6–44% in motorway storm water [31]. Natural zeolite was also used to uptake metals such as Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn from the composting process in Psittallia, Athens [46]. The uptake effectiveness was from 10 to 100% of the total amount. Moreover, Misaelides and Godelitsas [32] reported that the uptake of Hg2+ using Na-exchanged clinoptilolite rich tuff and Na-exchanged heulandite crystal from Greece were nearly 50% from aqueous solution at low concentration (10–500 mg Hg2+/L). At higher concentration (500 mg Hg2+/L), Na-exchanged clinoptilolite removed 43 mg Hg2+/g, while Na-exchanged heulandite removed 16 mg Hg2+/g. In order to reuse the exchanged-zeolite after heavy metal removal, NaCl solution with concentration 20–25 g/L at pH 4–4.5 adjusted with HCl can be used [47]. Heavy metal ions in the regenerating solution can be precipitated as hydroxides by increasing the pH to 6 or 7, depending on the type and concentration of heavy metal ions in the regenerating solution.

3.2. Modified natural zeolite to remove anion contaminants

Due to the net negative charge on the framework, zeolites have little or no affinity for anions. It has been reported by Komarowski et al. [35], who investigated the removal of phosphate from secondary treated municipal wastewater using natural zeolite in batch experiment, that natural zeolite was less effective for reducing phosphate, which is only up to 15%. Likewise, less than 15% of phosphate removal using Australian natural zeolite was reported by Widiastuti et al. [22]. In addition, Garcia et al. [42] reported about 30% removal of phosphorus using phillipsite-rich tuff columns with 0.5–2.0 mm particle size and less than 30% phosphorus removal using natural zeolite containing about 70% clinoptilolite as reported by Sakadevan and Bavor [48]. This disadvantage stimulates further research for new application of natural zeolites for anions removal by modifying the zeolite. In an attempt to improve the natural zeolites sorptive capacities, natural zeolites have been modified in different ways.

One zeolite modification method is to use organic compounds such as cationic surfactants. Due to a large specific surface area and a net negative structural charge, zeolites could be modified by cationic surfactants such as tetramethylammonium [44], hexadecyltrimethylammonium (HDTMA) [49], octadecyltrimethylammonium [50,51], cetylpyridinium [39], ethylhexadecyl-dimethylammonium [39] and 4-methylpyridinium [52]. Sorption of cationic surfactants onto negatively charged zeolite is controlled by cation exchange and hydrophobic interactions [53]. Ion exchange of cationic surfactants and a cation on the external surface of zeolite such Na+, K+, Ca2+, Mg2+ may balance the negative charge of the zeolite [49]. Therefore, cationic surfactants have been used to reverse the surface properties of natural zeolite from negative to positive.

Sullivan et al. [54] stated that the sorption of surfactant on the zeolite surface depending on the degree of sorption. For surfactant concentration less than external exchange capacity (ECEC), a cation exchange may play an important role, while above the ECEC, sorption may occur via intermolecular or surfactant–surface interactions and may be similar to organic partitioning or Henry’s law mechanisms [54].

Research on the use of cationic surfactant modified zeolite to remove phosphate, PO43− has been carried out [39,55]. It was reported that surfactant-modified-zeolite (SMZ) using cetylpyridinium chloride could remove phosphate ranging from 50% to 90% depending upon variables such as concentration of cetylpyridinium chloride on the preparation of SMZ; the initial phosphate concentration, contact time and initial pH on the adsorption of phosphate using SMZ [39].

Another investigation of the use of cationic surfactant zeolite to remove chromate, CrO42− and arsenate, AsO43− was reported using hexadecyl-
trimethylammonium (HDTMA) [40,49]. The adsorbed chromate on the HDTMA-modified zeolite reached 15.7 mmol/kg, whereas the arsenate was 95% removed from the simulated leachate at all concentrations tested. The simulated leachate was an alkaline leachate from a soil washing process that contained organic carbon of 45 mg/L at pH of 12.5 [40,41]. Interestingly, although the tailored zeolites have anion exchange properties, the zeolite is still accessible to sorb inorganic cations. It is because the tailoring agents are relatively large molecules, as a result they remain on the external surface of the zeolite crystal [43] and do not enter the zeolite channels [56]. The internal cation-exchange site of the zeolite remains accessible to sorb inorganic cations. In order to investigate the ability of SMZ to sorb cations, removal of Sr2+ on unmodified-zeolite and HDTMA-modified zeolite was studied. Results showed that the amount of Sr2+ sorbed by unmodified zeolite was 75 meq/kg, while the amount of Sr2+ sorbed by HDTMA-modified zeolite was 15 meq/kg [57]. It is because the Sr2+ was removed by unmodified zeolite by cation exchange and the HDTMA was also bound to the external surface of zeolite by cation exchange. As a result, there was relatively little Sr2+ that penetrated into the internal zeolite channels of SMZ and access the exchange sites. In contrast to Sr2+, removal of Pb2+ by unmodified and HDTMA-modified zeolites was almost the same [57]. Unlike Sr2+, Pb2+ as a transition metal cation can bind to mineral surfaces by specific complexation or precipitation reactions as well as by cation exchange. To prove this mechanism, further investigation was proposed [57].

Regeneration of chromate-saturated modified zeolite and PCE-saturated modified zeolite has also been studied [58]. In the study, chromate-saturated modified zeolite was regenerated using carbonate extraction method and compared to sodium dithionate method, whereas PCE-saturated modified zeolite was regenerated using air sparging. The results indicated that both saturated modified zeolites could be regenerated. The chromate-saturated modified zeolite can be regenerated partially using sodium dithionite, while PCE-saturated modified zeolite can be fully regeneration by air sparging [43].

An alternative way to enhance the zeolite ability for phosphate removal was investigated by dry grinding of low price materials such as clinoptilolite rock, FeSO4.7H2O and limestone [23]. Iron (III)-oxide-hydroxide was formed mechanochemically during the grinding at ambient temperature. The iron (III)-oxide-hydroxide may be able to react with the phosphate in the sewage.

3.3. Natural zeolites and modified natural zeolites to remove microorganisms

Natural zeolites are highly porous materials with bidispersive porosity [59]. The zeolites have a system of voids, channels and cavities in their structure. Due to the unique structure, some molecules under ideal conditions can pass through the channel or cavity to fill the available adsorptive space [59]. Likewise, the large crystal surface area of the zeolites is accessible for adhering microorganisms such as bacteria [23]. Therefore, natural zeolites are a potential material for biofilter.

Research on removal of bacteria indicators of pollution and organic matter using natural phillip-site columns has also been conducted in a percolation reactor at a constant solution flow rate [42]. In this study, wastewater was obtained from the treatment plant at Santa Cruz de Tenerife city (Canary Islands). The indicator bacteria such as total coliforms, Faecal coliforms and faecal streptococci and residual organic matter evaluated as COD and BOD values were determined in the wastewater before and after the percolation treatment every two days. The results showed that the retention values of substratum for total coliforms and faecal coliforms are greater than fecal streptococci (see in Table 1). Surface morphology shows that an amorphous biofilm covers the whole material after treatment.

The potential of natural zeolites to remove
enteric bacteria and viruses from drinking water has been reported [23]. Escherichia coli, poliovirus, coxsackie virus and bacteriophages from drinking water have been effectively removed using an Al2(SO4)3 coagulant and clinoptilolite. The efficient microorganism removal is caused by the formation of microorganism-clinoptilolite-coagulant complexes.

Research to evaluate the potential for natural zeolite particularly natural clinoptilolite in drinking water preparation to remove coliform bacteria, total bacteria, phyto-and zooplankton was carried out. It was reported that the physicochemical and mechanical properties of the clinoptilolite used for 3 years were unchanged and gave a stable decrease in the number of coliform bacteria, total bacteria, phyto- and zooplankton in the finished drinking water [23].

Nikashina and Myasoedov [44] investigated the potential of natural zeolite particularly clinoptilolite to trap E. coli from drinking water by modifying the clinoptilolite with amine group-polymethylene guanidine chloride (PHMG), then linked with epichlorohydrine. This organo-zeolite was named clinotsid. The investigation results of the clinotsid for bacteria removal are presented in Table 1. Clearly, the clinotsid was an effective disinfectant for drinking water.

Another natural zeolite application to adhere bacteria from water was conducted by Bowman [43]. The natural zeolite particularly clinoptilolite-rich tuff from St. Cloud, New Mexico was modified with cationic surfactant e.g hexadecyltrimethylammonium (HDTMA). Ability of the surfactant-modified zeolite (SMZ) to remove common pathogens from sewage-contaminated water was then determined. Results indicated the complete removal of E. coli and bacteriophage MS2 by the SMZ. It is because HDTMA is bactericidal in aqueous solution even at low concentrations. As a result, the organisms are greatly removed by the surfactant bound to zeolite. The SMZ was also applied to protect drinking water wells from viruses and bacteria [51] and to remove viruses and bacteria from ground water [60].

In comparison with other adsorbents such as clay, zeolite does not need to put in a liquid colloidal state [12] and has better hydraulic properties [43]. In addition, zeolite in contrast to activated carbon has a better performance in term of cation removal.

4. Conclusions and recommendations for future work

Natural zeolites offer the possibility for removing contaminants in water and wastewater such as inorganic cations and anions as well as microorganisms. Natural zeolites are also a promising material for the use in greywater treatment. However, no thorough investigation has been carried out with regard to using natural zeolites for greywater treatment. It is because little has been known about the fundamental characteristics of greywater. Greywater has a unique matrix of contaminants, which are different from other wastewater including municipal wastewater. The composition of greywater varies significantly from source to source. In the absence of faeces, toilet paper and kitchen waste, greywater is less polluted than municipal wastewater. However, greywater contains much higher chemical and microbiological contaminants than municipal wastewater. Also, the micro-organisms tend to grow significantly in the collecting/treatment/distributing pipeline. Therefore, further research into the feasibility of using natural zeolites to remove contaminants in greywater must be investigated. An effort to improve the natural zeolite performance is also a challenge especially in application to remove anion contaminants in greywater.

Acknowledgements

N. Widiastuti wishes to thank Australian Development Scholarship (ADS) provided by the Australian Federal Government.
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