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The use of synthesized zeolites from power plant rice husk ash obtained from Thailand as adsorbent for cadmium contamination removal from zinc mining

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ABSTRACT

The people from the 12 villages near the zinc mining in Mae Sot District, Tak Province, and Northwest of Thailand, were suffering contamination with cadmium (Cd^{2+}). It was released into the main source of water supply, Mae Taw River, which also be used for agriculture. This represents a serious problem that should be addressed accordingly. This study aims to investigate the synthesized zeolite from agriculture waste by-product in Thailand such as; rice husk ash (RHA) with white part is containing high silica content which can be used as a potential zeolites precursor and used to solve the problem by removing cadmium in aqueous solution via batch experiments. The removal of Cd^{2+} in the concentration range of 50–500 mg/L. The maximum removal capacity of synthesized zeolite Na—A and Na—X for Cd^{2+} were found to be 736.38 and 684.46 mg/g, respectively. The result was compared to synthetic zeolite 3A commercial shown that synthesized zeolites from RHA have high removal capacity close to commercial one of 753.58 mg/g using 0.3 g/L dosage of zeolite and consistent with an endothermic reaction, an increase in the temperature (20–40 °C) resulted in increasing cadmium adsorption rate. Therefore, the synthesized zeolites from RHA have high potential to remove cadmium contamination in water.

Introduction

Incidences of cadmium contamination were reported in the Mae Sot District, Tak Province (Fig. 1) in 2003 by the Department of Agriculture (DOA), Ministry of Agriculture (MOA) and Department of Pollution Control (DPC) [1]. Cadmium usually occurs in association with zinc ore and is a by-product of zinc mining. Cadmium is a toxic heavy metal which causes environmental problems and can be accumulated in human body through the food chain, and also can causes damage to the ecosystem [2].

Long-term excessive oral exposure can result in chronic cadmium poisoning. Urinary excretion of cadmium is a good indicator of excessive cadmium exposure [4]. Previous studies have found high concentrations of cadmium in the rice field, where rice grain contain up to 284 mg/kg on a dry material base [5], and contamination in water, sediment, fish and shellfish with an average of 50.48 mg/L [6]. People that live in this area have high levels of cadmium in their urine and have experienced kidney and bone damages problems [7]. The World Health Organization (WHO) recommends the permissible cadmium concentration of 0.003 mg/L in potable water [8]. There are varieties of water treatment for cadmium contaminated water such as filtration, reverse osmosis, flocculation, activated carbon, chemical precipitation or coagulation, ultrafiltration and electrochemical method. However, the aforementioned techniques are not economically feasible for small or medium industries in rural area or developing countries. Recently, the ion-exchange and adsorption processes have been reported as the most common and effective process for removal of hazardous substances at very low concentrations. The use of agricultural waste products or bio-sorption are becoming the new alternative for waste water treatment and suitable for small or medium industries.

Thailand is an agricultural country and rice is the most preferred product. Production rated at 20 million tonnes/year could produce 5 million tons of husks. Nowadays, rice husk is used as energy source for power plant generating an 18% ashes from the total amount of husk. These ashes have high silica content of about 97% and are be used in some industries like cement, glass, electronic and steel industry, while the remainder is disposed of on landfills or lagoon. This practice has a hazardous impact on the

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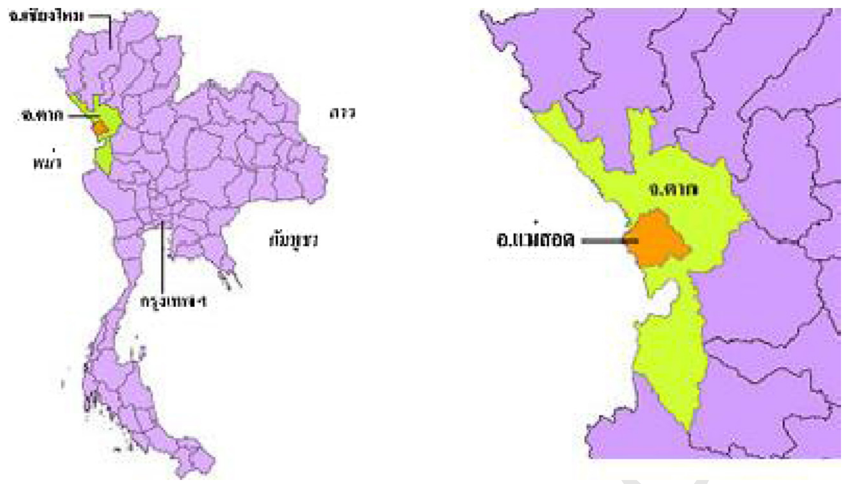

Fig. 1. Mae Sot district, Tak Province, Thailand [3].

environment by polluting the soil and groundwater. In other cases, if the remaining ashes from burned husks are not collected and treated properly, they can cause air pollution that affect the environment as well due to their small particle size and light weight. Due to the above reasons, much research work has been undertaken to develop new techniques of ash utilization. The rice husk ash (RHA) is mainly composed of high silica content and some alumina content typically used in zeolite synthesis [9], such as coal fly ash [10,11], oil shale ash [12] and bagasse fly ash [13]. Zeolites are crystalline microporous aluminosilicates with very well-defined structures that consist of framework formed by tetrahedra of SiO_4 and AlO_4. The isomorphous substitution of Al^{3+} for Si^{4+} in the tetrahedra results in a negative charge on the zeolite framework that can be balanced by exchangeable cations. Hence, zeolites can exchange cations but not anions. Zeolite may be represented by the empirical formula $M_{n/2}O\cdot Al_2O_3\cdot xSiO_2\cdot yH_2O$ [14], where generally x is equal to or greater than 2 because AlO^{-}_{4} tetrahedra are joined only to SiO^{-}_{4} tetrahedra, y is the water contained in the voids of the zeolite and n is the cation valence. The ion-exchange capacity of the zeolite therefore depends on the framework Si/Al ratio and decreases with increase in the Si/Al ratio.

Zeolites show special importance in water and gas purification, catalysts for hydrogenation, alkylation and isomerization, and sorbents for the removal of contaminants such as heavy metals, toxic gases, dyes and organic pollutants [15]. There is much research work on production of zeolites using coal fly ash as a resource and this constitutes one important issue of waste management. However, there are few researchers on production of zeolite using silicon content from rice husk ash as a resource. Moreover, Na–A and Na–X zeolites are within the group of lindes (LTA) and faujasites (FAU), respectively, and are considered to be low-silica zeolites (Si/Al < 5). They are obtained from starting mixtures differing essentially in the Si/Al ratio which is higher for Na–X than for Na–A for the temperature range of 20–120 °C, preferring temperatures around 90 °C [16].

The use of zeolites as adsorbent for cadmium removal from aqueous solutions was investigated by many researchers over the last decades due to their ion exchange properties, hydrophilic affinities, and high surface area. Until now there are still ongoing research on the removal of cadmium by zeolitic materials. Shawabkeh et al. [17], used oil shale ash formed of zeolite synthesized to uptake cadmium in wastewater, while Purna et al. [18], found that the commercial zeolite 4A and 13X can adsorb cadmium from aqueous solution. The results show that the removal of cadmium at pH 6 was 9.891 mg/g with zeolite 4A and 9.441 mg/g with zeolite 13X, respectively. However, rice husk ash used to synthesize several kinds of zeolite such as ZSM-11 [19], beta [20], NaY [21–23], ZSM-5 [24,25], NaA [26,27], NaX [28] but with a low adsorption capacity. In this study, the adsorption capacities of synthesized zeolite from rice husk ash are investigated with simple method to find a suitable way to remove cadmium contamination in rural region such as Northwestern of Thailand.

Experimental

Materials

Rice husk ash (RHA) was collected directly from electrostatic precipitators of Roi-Et Green power plant, Thailand after combustion at 650 °C. This ash was used without any pretreatment and its properties were characterized by various techniques such as Inductively Couple Plasma-Atomic Emission Spectroscopy (ICP-AES) method using SPS 7800 (SII) and X-ray diffraction (XRD) analysis using Multiflex (Rigaku). Other chemicals used in the present study were obtained from Wako Pure Chemical Industries, Ltd., Japan.

Procedure

Silica extraction from rice husk ash

RHA was sieved to remove dirt, and was dried in oven at 100 °C overnight. The particle fraction used for the synthesis was separated from the bulk RHA by mechanical sieving using standard sieve of 25 meshes (particle size less than 0.71 mm). The chemical characterization of representative of RHA was undertaken by Inductive Couple Plasma-Atomic Emission Spectroscopy (ICP-AES: SPS 7800 (II)), and the results showed high silica content (Table 1). To extract silicon from RHA particles, RHA was mixed with sodium

Table 1
Chemical composition of rice husk ash after combustion at 650 °C. Q3

Composition (wt.%)	Rice husk ash
SiO_2	91.50
Al_2O_3	2.27
K_2O	0.48
MgO	1.00
Na_2O	2.68
Fe_2O_3	0.10
Others	1.97

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