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Mercury(II) Adsorption onto the Magnesium Oxide Impregnated Volcanic Ash Soil Derived Ceramic from Aqueous Phase

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ABSTRACT: The present investigation attempted to develop the ceramic adsorbent media in order to remove the mercury from aqueous phase. Two ceramics, akadama volcanic ash soil-ceramic (A-ceramic) and improved magnesium oxide impregnated akadama volcanic ash soil-ceramic (MA-ceramic) were produced from raw akadama volcanic ash soil by heating process to determine the Hg(II) adsorption capacity. Adsorption experiments of ceramics were performed as a function of contact time, solution pH, adsorbent dosage and adsorbate concentration following the batch mode operation. Results clearly revealed the adsorption capacity (265±8.5 μg/g) of MA-ceramic is significantly higher (~12 times) than that of the A-ceramic material. The adsorption was largely pH dependent and neutral pH was associated with higher mercury adsorption capacity in both ceramics. MA-ceramic showed 94% and 86% desorption and resorption capacities, respectively.

Key words: Adsorption, Akadama olcanic ash soil, Ceramic, Magnesium oxide, Mercury

INTRODUCTION

Mercury (Hg), one of the most toxic pollutants, poses serious risk on various forms of life through the network of food chains by bioconcentration, bioaccumulation and biomagnification phenomena. It is considered as a hazardous heavy metal among the 11 most toxic priority substances in the list of pollutants contained in the Water Framework Directive (Directive, 2000/60/EC) not only due to public health demeanor but also for the severe environmental impacts (Horvat *et al.,* 2003; Fang *et al.,* 2004). The toxicity of Hg depends on its chemical forms and the route of exposure such as, methylmercury (CH_3Hg) is the most toxic form over the elemental Hg (Hg^0) and is adsorbed more readily leading to an unrecoverable penalty scale problem in organisms. Mercury and its derivatives recognized as dangerous and insidious poisons and can be adsorbed through the gastrointestinal tract, skin and lungs (Bidstrup, 1964) these are responsible for the development of carcinogenic, mutagenic, teratogenic and also promotes several health problems, tyrosinemia, paralysis, serious intestinal and urinary complications, dysfunction of the central nervous system and in more severe cases of intoxication, death. In late 1950s and 1960s, we witnessed the environmental tragedy of Hg toxicity in Minamata Bay, Japan, where hundreds of individuals suffered from Hg poisoning through fish - Minamata disease (Zhang *et al.,* 2005). The U.S. Environmental Protection Agency (EPA) estimated 630,000 newborns in America are at risk of unsafe levels of Hg exposure Mahaffey, 2004).

Mercury is a naturally occurring trace element found in rocks, soils, sediments and atmosphere. Worldwide coal burning, municipal solid waste incineration, electronic, paper, pharmaceutical industries (Biester *et al.,* 2002; Sznopek and Goonan, 2000; Tack *et al.,* 2005) and tailings of gold mines were identified as the major anthropogenic origins of Hg emission. Besides, emissions of Hg from chlor-alkali, paint, oil refining, rubber processing, fertilizer industries (Kadirvelu *et al.,* 2004) and indiscriminate use in barometers, thermometers, pumps, lamps have also been determined to have a significant impact on the environment (Davis *et al.,* 2000; Chantawong *et al.,* 2003). Morency (2002) reported annual emissions of Hg from anthropogenic sources are to be over 2000 tonnes, of which about 30% are from coal-fired power plants.

Once injected into the environment, it persists for decades by means of continuous cycling between air, water, and land which inviting the above mentioned

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serious environmental impacts. Therefore, it has drawn the attention to remove Hg from polluted environment as well as from contaminated effluents before discharging into the environment to protect the whole lives on earth. In this respect, a number of methods have been evolved for the removal of Hg from water during the last few decades, but commonly adopted methods include sulphide precipitation, membrane filtration, iron and alum coagulation, ion exchange and activated carbon adsorption. Adsorption of Hg by solid materials is a suitable choice for final purification of wastewater with appreciable removal efficiency (McGuire and Suffett, 1983). Although several natural and refuse derived materials have been tested as alternative low-cost sorbents (Kawamura *et al.,* 1997; Arpa *et al.,* 2000; Ekinci *et al.,* 2002), but activated carbon and zeolites are the typical sorbents for removing Hg ions from aqueous solution (Hannah etal., 1977; Palmer *et al.,* 1988).

Despite these, a good adsorption and ionic exchange capacities of several minerals and soils (Stumm *et al.,* 1996; Larsen *et al.,* 1992; Sakadevan and Bavor, 1998) have been suggested to possible use for the remediation of natural water bodies. Recently, varieties of low cost clay/soil based ceramic pot filter (Van Halem *et al.,* 2008) and membrane filters (Belouatek *et al.,* 2008; Benito *et al.,* 2007) have been developed to treat the wastewater by removing various metals, nutrients and dyes (Bhakta and Munekage, 2009a). The development of low-cost inorganic membranes is one of the challenges for future from naturally occurring materials such as clays and apatites which need lower firing temperature than metal oxide materials (Mukasyan *et al.,* 2001; Li *et al.,* 2001) and have high flux performance to treat large volumes of liquid effluent. Inorganic membranes (oxide based inorganic membranes such as Al_2O_3, ZrO_2, TiO_2 and hyteropolysiloxanes) have attracted more attention due to high mechanical, chemical and thermal resistance, which can be potentially applied in liquid separation, gas separation, pervaporation, membrane reactors, etc. (Larbot *et al.,* 1994; Medoukali *et al.,* 1999; Belouatek *et al.,* 2005). Kanazirev (2009) used copper oxide as a potential adsorbent to remove Hg from contaminated water. It has also been proposed that ceramic derived from clay is an alternative adsorbent to remove Hg (Bhakta *et al.,* 2009b), arsenic and lead (Salim *et al.,* 2007; 2008).

On account of the above points of view, though varieties of adsorbent media have been developed to remove Hg but many of them are costly, inaccessible and in some cases having negative environmental impact in practical field for the treatment of massive Hg contaminated effluents of the industrial sectors especially in the underdeveloped countries. Therefore, the development of low cost materials is still needed in this regard. Understanding of these facts, the present study has focused to develop the low-cost environmentally sound metal oxide impregnated ceramic adsorbent from akadama volcanic ash soil for the removal of Hg from water phase.

MATERIALS & METHODS

The present study used raw akadama volcanic ash soil as major precursor material collected from Tochigi (36°45' *N*, 139°45' *E*), Japan. Our previous study on Hg removal capacity of some soils reported the availability, physical and chemical characteristics of the raw akadama volcanic ash soil (Bhakta and Munekage, 2010). It has already been determined, though raw akadama (fluffy loamy brown in colour and a bit heavier than kanuma) and kanuma (fluffy, less dense, very lightweight and whitish yellow in colour) are potential Hg removing soil agents (Bhakta and Munekage, 2010), but the capacity of heat treated akadama was a bit superior to that of the kanuma (results are not shown). Therefore, the present study considered akadama volcanic ash soil for the development of Hg removing metal oxide impregnated ceramic based adsorbent media. To select the best Hg removing metal oxide incorporated soil ceramic, a screening experiment was performed impregnating seven metal oxides, MgO, MnO_2, Al_2O_3, Fe_2O_3, CaO, ZnO and CuO into akadama soil @ 10% and developing ceramic by heating process. Mercury removal capacity of the developed ceramics from water was performed using one control of each metal oxides and another control of only akadama soil received no metal oxides in the screening process. Though Hg(II) removal was higher in CuO and MgO impregnated ceramics compared to that of the rest ceramics and control groups but we selected MgO for impregnation into the akadama soil to avoid the further environmental pollution of cupper. Finally, we prepared two types of ceramic: (1) only akadama volcanic ash soil ceramic which was called as A-ceramic and (2) MgO (10%) impregnated akadama volcanic ash soil derived ceramic which was named as MA-ceramic to compare the adsorption capacity in different experimental conditions. All experimental ceramic adsorbents were produced from EKOAIRANDO Co. Ltd. Kochi, Japan. According to the ceramic production process of company, the mixtures (Akadama soil and MgO) were heated gradually increasing the temperature @ 10°C / min from 100 to 900°C in an advanced furnace system. Two ceramics were ground by mortar and pastel and sieved through 0.42 mm mesh for using in the experiments.

Prepared ceramics were used for BET surface area analysis by an equipment NOVA-2000 (Quantachrome Corporation, Version 5.01). Mineralogical characterization of these ceramics was performed by X-ray diffraction (XRD, X'Pert PRO, Philips) in the Center for Advanced Marine Core Research, Kochi University, Japan.

All adsorption studies for both A- and MA-ceramics were carried out following the batch operation technique. Experiments were conducted in 200 ml capped glass bottles using known weight of the corresponding adsorbents to equilibrate the known volume of adsorbate (100 ml) within temperature controlled mechanical shaker at 25°C. Firstly, the effect of contact periods was determined maintaining the above conditions. The effect of pH on the adsorption of Hg was studied adjusting the pH values 3, 7 and 11 with HNO_3 and NaOH. To determine the effects of initial adsorbate concentration, the range of initial Hg concentration was from 50 to 500 µg/L whereas the effect of adsorbent dosage was found using the ceramic weights varied from 0.05 to 0.25 g/L. Each experiment was performed at least twice following the identical conditions using controls of only metal solution and adsorbent without metals in solution. All experiments were conducted using the Hg solution prepared from standard stock solution of mercuric chloride ($HgCl_2$, Cica-Reagent, Kanto Chemical Co., Inc., Tokyo, Japan).

The solution was collected from each bottle and centrifuged for separating the ceramic to quantify the residual Hg present in water using the advanced RA- 3 Mercury Analyzer (Nippon Instruments Corporation, Japan).

At the equilibrium, the amount of Hg(II) adsorbed onto the corresponding ceramic, q_e (µg/g) was calculated by following mass balance relationship equation:

$$q_e = \frac{(C_0 - C_e)\ V}{M} \qquad (1)$$

The effect of contact time on the removal of Hg(II) from water phase was shown in fig. 2. The adsorption rate was rapid until 6 h, followed by slow adsorption and equilibrium state was achieved at 20h of the contact time in MA-ceramic. A slow Hg(II) uptake of A-ceramic was found until 12h and steady state thereafter. The Hg(II) adsorption was significantly higher in MA-ceramic than that of the A-ceramic at 20h equilibrium period of adsorption (Fig. 2).

Fig. 3 shows the effect of pH on the adsorption of Hg(II) onto A- and MA- ceramics. A similar pH dependent Hg(II) removal response was observed in both A- and MA- ceramics. The Hg(II) adsorption was maximum at pH 7, whereas a lower adsorption was pronounced at pH <7 and >7 in two ceramics. The MA-ceramic exhibited a greater Hg(II) adsorption in different pH (3, 7 and 11) over the A-ceramic (Fig. 3). These data clearly demonstrated that neutral pH is supposed to be optimum for adsorbing the Hg(II) from aqueous phase for both ceramics. Bhakta et al. (2009b) proposed that pH 7 is the optimum for Hg(II) adsorption for a ceramic, whereas increasing and decreasing pH levels from pH 7 govern the reduced Hg adsorption. Salim et al. (2007; 2008) also reported pH 7.5 is optimum for arsenic adsorption of ceramic. From this proposition it may be suggested that the neutral pH plays significant roles in the chemisorptions as well as surface adsorption processes at the functional groups of modified MA-ceramic by sponsoring the lower H^+ conditions for removing Hg(II). The H^+ ions are preferentially adsorbed on the surface of the adsorbents. Thus, a higher competition between the positive metal ions and H^+ ions exists in the surface of adsorbent at acidic pH. Due to this reason, the higher H^+ ions concentration prevent the positively charged metal ions to reach at the binding surface of the adsorbent. The influence of initial pH on Hg(II) removal may be explained as follows: in the acidic condition, both the adsorbent and the adsorbate are positively charged (M^{2+} and H^+) and therefore, the net interaction is that of electrostatic repulsion (Langmuir, 1918). Annadurai et al. (2002) proposed the solution pH affect both the solution chemistry and the surface binding sites of the phosphogypsum. Chen and Wang put forwarded that pH increases or the concentration of protons is reduced; this results in a production of surface metal-adsorbent complex or higher removal of metal ions (Chen and Wang, 2000).

The effect of initial concentrations of Hg(II) on the adsorption process of A- and MA- ceramics was depicted in the fig. 4. The Hg(II) uptake of A- and MA-ceramics was increased from 1.2 to 6.8 and 1.2 to 4.2 times, respectively in the highest concentration compared to lower concentrations of the present investigation (Fig. 4). An remarkably elevated amount of Hg(II) adsorption was found in MA-ceramic than the A-ceramic in different concentrations. It is also clear from the results that the amount of removal of Hg(II) was enhanced with increasing the initial concentration of Hg(II) by per gram of two employed ceramics. The results are similar to adsorption of cadmium from aqueous solution by two different rock types Pumice and Scoria (Alemayehu and Lennartz, 2009). This increase may be as a consequence of saturation of higher number of functional groups by means of

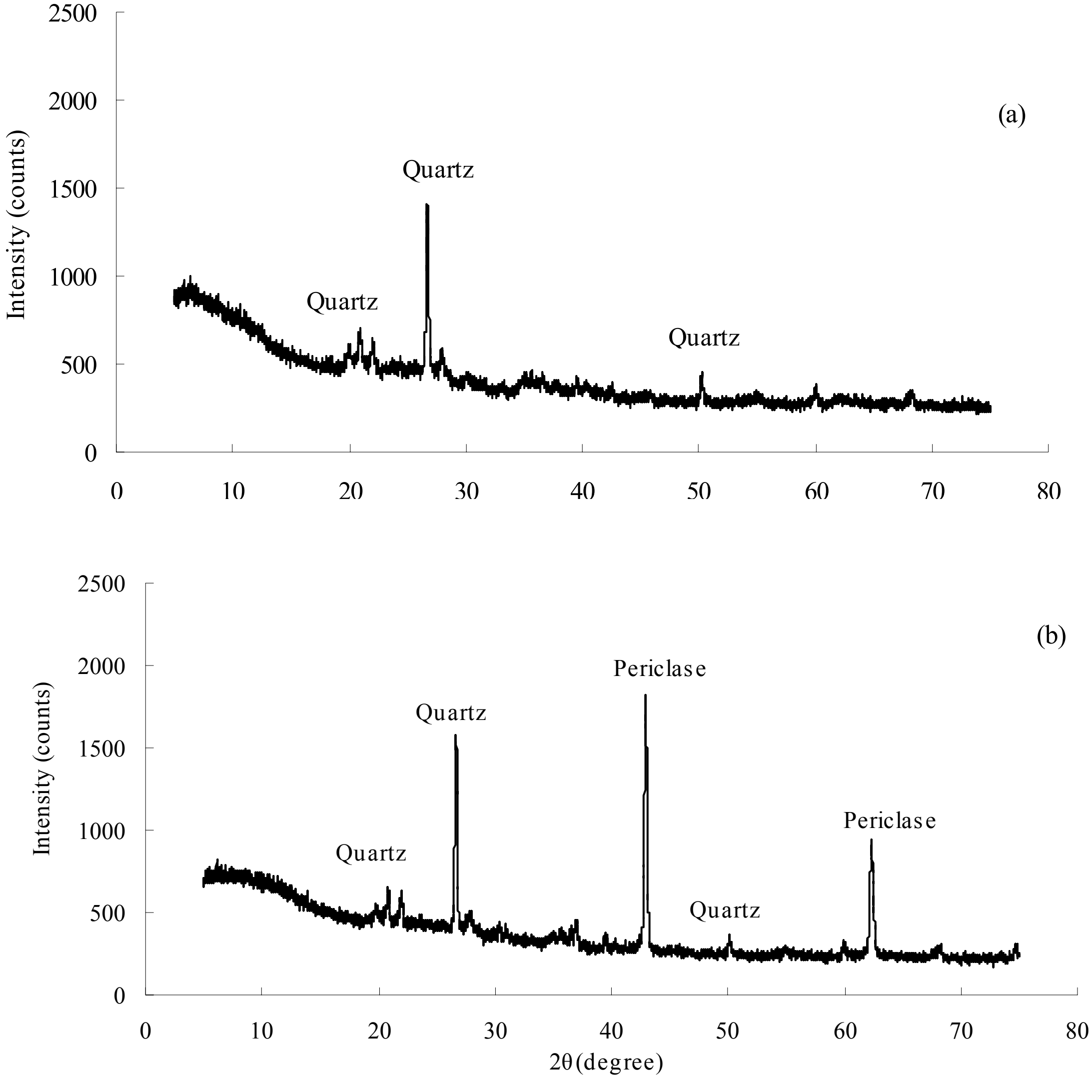

Fig. 1. XRD patterns of A-ceramic (a) and MA-ceramic (b)

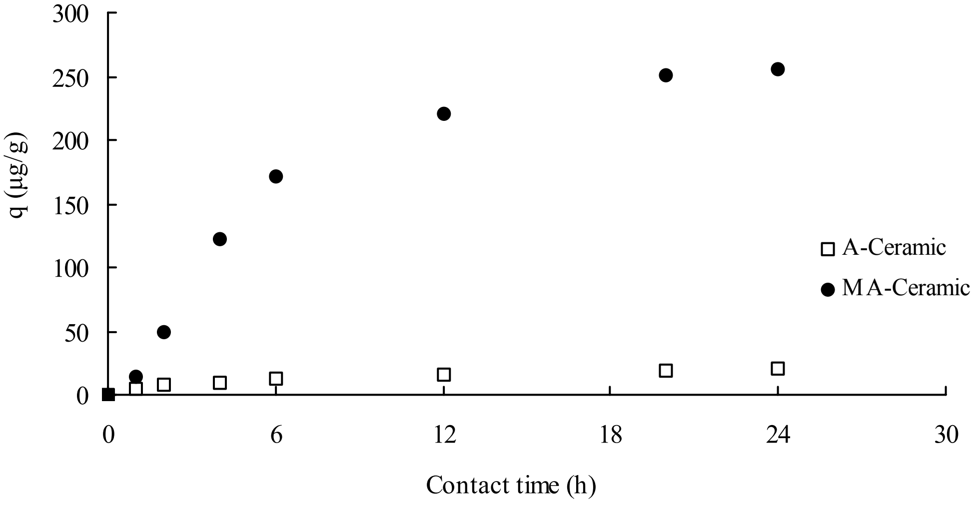

Fig. 2. Effects of contact time in the Hg(II) adsorption of A-ceramic and MA-ceramic (initial concentration 400 μg/L, dosage 0.05 g/L and pH 7)

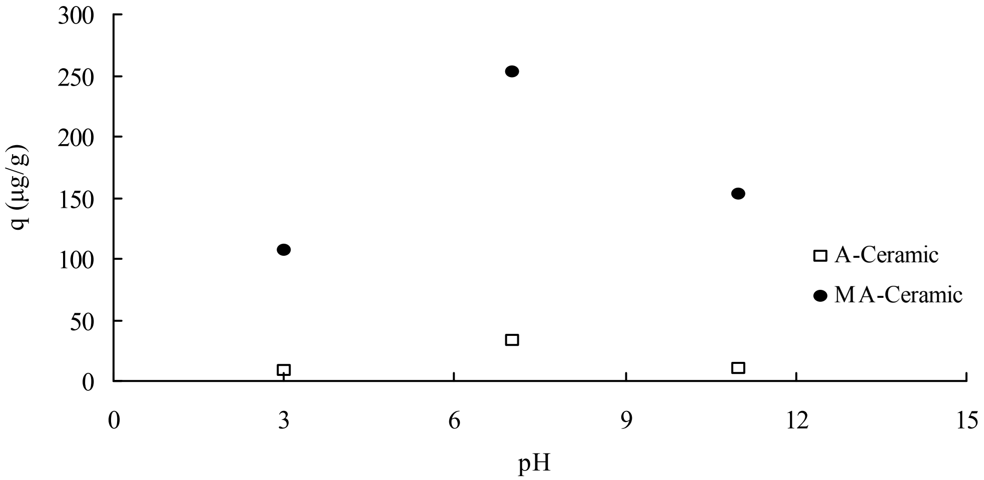

Fig. 3. Effects of initial pH on the Hg(II) adsorption onto A-ceramic and MA-ceramic (initial concentration 400 µg/L, dosage 0.05 g/L and contact time 24 h)

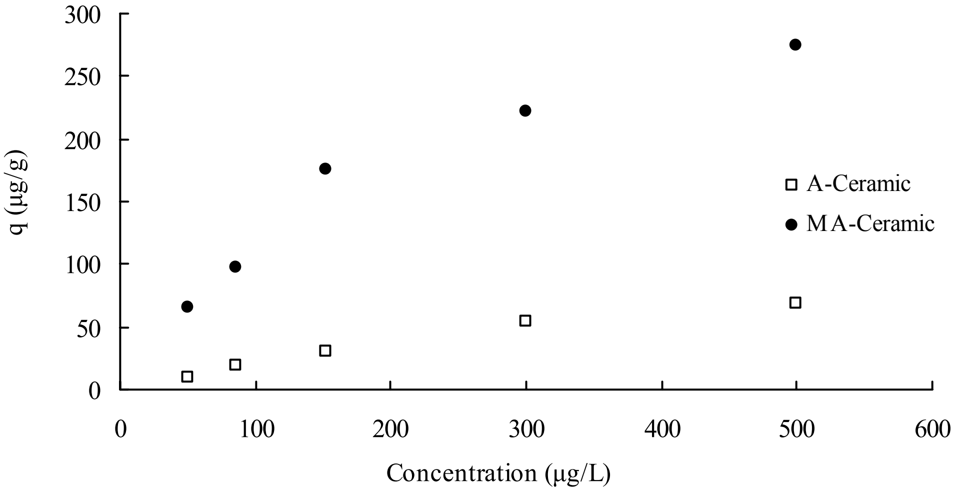

Fig. 4. Effects of initial concentration on the Hg(II) adsorption onto A-ceramic and MA-ceramic (dosage 0.05 g/L, contact time 24 h and pH 7)

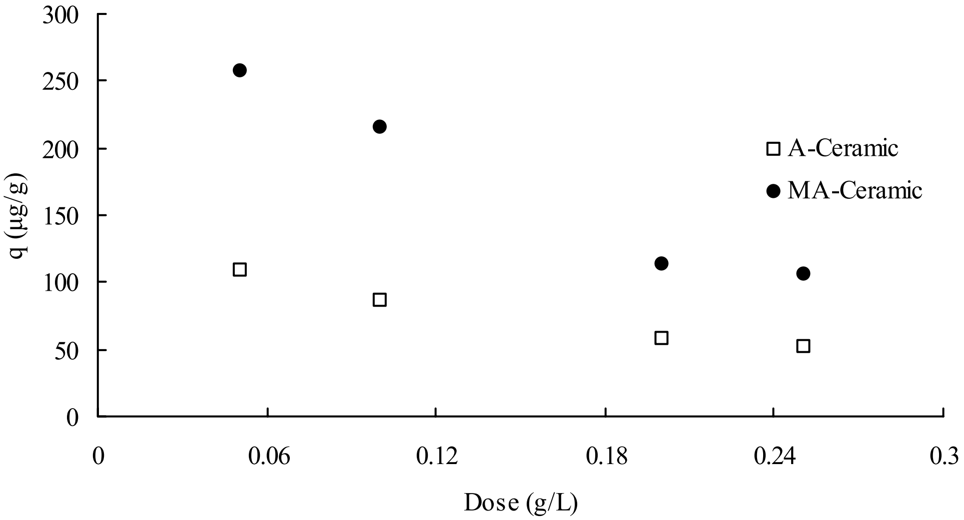

Fig. 5. Effects of adsorbent doses on the Hg(II) adsorption of A-ceramic and MA-ceramic (initial concentration 400 µg/L, contact time 24 h and pH 7)

increasing electrostatic interactions between the Hg ions and functional groups in the higher concentrations.

Fig. 5 elucidates the effect of adsorbent dosages on the Hg(II) adsorption of A- and MA- ceramics. Unlike effect of initial concentrations, though total Hg(II) adsorption increased with increasing the dosages but the amount of adsorption per gram of ceramic was higher (A-ceramic 1.2 – 2.1 times and MA-ceramic 1.2 – 2.4 times) in lower dosages than that of the higher dosages in this experiment. MA-ceramic also showed higher Hg(II) removal capacity compared to that of the A-ceramic in different dosages. The decrease in adsorption amount with the increasing adsorbent dosages may result from the electrostatic interactions, interference between binding sites, and reduced mixing for higher densities at higher dosages of adsorbent (Fourest and Roux, 1992; Montanher *et al.,* 2005). This is supported with the metal removal result of sequential adsorbent treatment using solid adsorbents (such as sand, silica, coal and alumina) proposed by Yabe and de Oliveira (2003).

In order to determine the effectiveness of various adsorbent materials, it is important to know about the relationship between the adsorbate and adsorbent under different operation conditions of the batch experiments (Benguella *et al.,* 2002; Yalcinkaya *et al.,* 2002). These relationships of equilibrium data of adsorption process are commonly known as adsorption isotherm. Though a number of isotherm equations have been proposed to describe the modelling of the adsorption process, but Langmuir and Freundlich models are commonly used models for the single and binary cases among them (Kandah, 2004).

In the present investigation, the comparison between the A- and MA- ceramics in respect to Hg(II) adsorption capacity was performed following the Langmuir and the Freundlich models. The following equations were used for describing the Langmuir (2) and Freundlich (3) isotherms:

$$q_e = \frac{Q^0 K_L C_e}{1 + K_L C_e} \qquad (2)$$

Where, Q^0 (μg/g) and KL (L/mg) are the Langmuir parameters, related with the maximum capacity of adsorption and with the binding energy of adsorption, respectively.

$$q_e = K_F C_e^{1/n} \qquad (3)$$

Where, K_F [(μg $L^{1/n}$)/(g $\mu g^{1/n}$)] and n (a dimensional) are the Freundlich parameter characteristic of the corresponding system. Generally, it is stated that the values of n in the range of 1 to 10 represents good adsorption (Oliveira *et al.,* 2005).

The isotherms of A- and MA- ceramics for Hg(II) adsorption from aqueous solution were illustrated in the Fig. 6. Fittings parameters of the Langmuir and Freundlich isotherm models derived from the experimental results of corresponding ceramic were shown in Table 1. The R^2 values of A-ceramic (Freundlich 0.96, Langmuir 0.92) and MA-ceramic (Freundlich 0.99, Langmuir 0.81) clearly apprehended that Freundlich isotherm is well fitted with the experimental data rather than the Langmuir isotherm. Therefore, monolayer coverage of Hg(II) ions on ceramic cannot be explained. On the other hand, it may be seen, the Langmuir maximum adsorption capacities (Q^o) is greater in MA-ceramic than that of the A-ceramic which indicated that the adsorption capacity is higher in MA-ceramic over the A-ceramic (Table-1). In this study, the Freundlich constant n was $1<n<10$ which pronounces the adsorption is favorable. Generally, it is also considered that the Freundlich constant $1/n$ is an indicator to evaluate the adsorption strength and the deviation from linearity of the adsorption.

However, it can be supposed the additional mineral forms a greater percentage of complex with higher number of Hg(II) which is the important driving factor for removing greater amount of Hg(II) from water phase in MA-ceramic. As previously mentioned, the surface area was smaller in the MA-ceramic than that of the A-ceramic, therefore the higher Hg(II) adsorption onto the MA-ceramic might be due to the effect of chemisorptions process rather than physical surface adsorption. It can be explained like that adsorption mechanism of Hg(II) onto the MA-ceramic is influenced by the chemical interaction between additional functional group of MA-ceramic, periclase and Hg(II) which forms a chemical complex resulting in the chemisorptions of Hg(II).

Desorption and resorption capacities of any developed adsorbent are important aspects in order to apply in practical field of water treatment. In the present study, the MA-ceramic showed about 94% Hg(II) desorption capacity at the period of 60 min by 10 mM EDTA (Fig. 7). About 86% of total adsorbed Hg(II) was released within 10 min period. The resorption study also pronounced that about 89% resorption capacity was achieved by desorbed MA-ceramic at 20 h equilibrium period (Fig. 7). High magnitude of desorption and resorption capacities clearly demonstrated that MA-ceramic can be recycled by

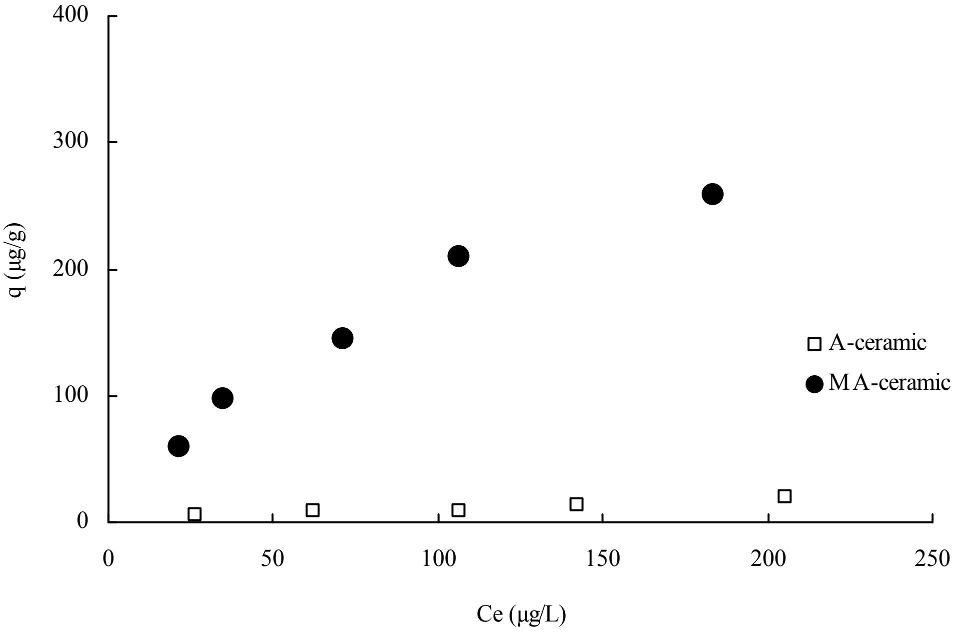

Fig. 6. Equilibrium curve for the adsorption of Hg(II) by A-ceramic and MA-ceramic (initial concentration 400 µg/L, contact time 24 h and pH 7)

Table 1. Langmuir and Freundlich parameters for A-ceramic and MA-ceramic in the adsorption isotherm of Hg(II)

Adsorbents	Langmuir			Freundlich		
	K_L (L/µg)	Q^0 (µg/g)	R^2	1/n	K_F	R^2
A-ceramic	0.007	28.7	0.81	0.55	0.94	0.96
MA-ceramic	0.002	74.6	0.92	0.75	6.13	0.99

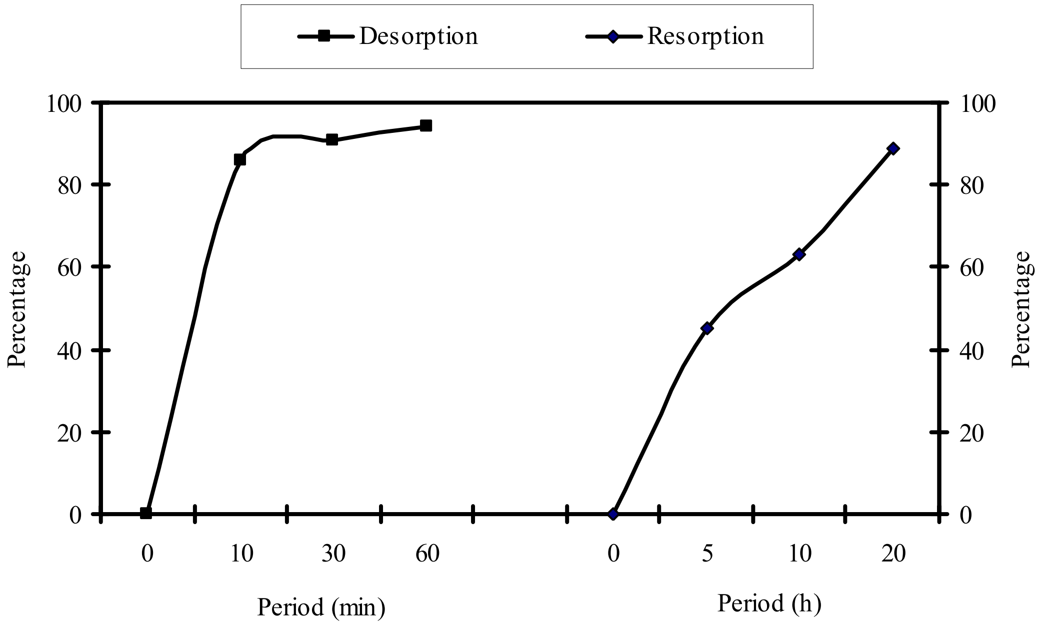

Fig. 7. Mercury desorption and resorption characteristics of MA-ceramic

desorbing Hg using EDTA. Finally, the EDTA and metal complexes can easily be separated as a solid EDTA and metal salts with the help of sulfuric or hydrochloric acid.

CONCLUSION

In the present investigation, it was clearly uncovered that adsorption capacity (265±8.5 µg/g) of MgO impregnated akadama volcanic ash soil-ceramic (MA-ceramic) is significantly higher (~12 times) than that of the only akadama volcanic ash soil-ceramic (A-ceramic) probably due to the presence of additional mineral of MgO, periclase developed by the impregnation of MgO and chemical modification by heating process. The BET and mineralogical characteristics supported the adsorption mechanism of Hg(II) onto MA-ceramic is highly influenced by the chemical interaction between additional functional group of MA-ceramic, periclase and Hg(II) which resulting in the chemisorptions process rather than only surface adsorption. Results of the contact time also revealed the adsorption equilibrium time of MA-ceramic is 20 h period. Hg(II) adsorption is largely pH dependent and neutral pH (7.0) greatly increases the adsorption capacity of the ceramics. Moreover, above results also demonstrated that ceramic dosage and initial concentration of the Hg(II) significantly influence the adsorption capacity of the ceramics. The Freundlich isotherm is well fitted with the data of present study.

However, varieties of adsorbent media including activated carbon developed in this respect, but many of them are costly, inaccessible and in some cases having negative environmental hazardous impacts of spent adsorbent in practical field. Adsorption, desorption and resorption characteristics signified that MA-ceramic would be an environmentally sound adsorbent which can safely be used in treating the Hg contaminated effluents. Furthermore, this finding would be helpful to great extend to develop the improved Hg removing filter as well as membrane filter of ceramic for producing the safe Hg free drinking water and treating the Hg contaminated effluents of the industrial sectors especially in the underdeveloped countries with low-cost. It should be mentioned clearly herein addition of some percentage of MgO as an essential ingredients for developing the ceramic filters would be an effective methods for treating the drinking water in order to safe the life from probable drinking water Hg contamination. Though Hg removal capacity is lesser than that of the activated carbon and other few developed adsorbents as published data available, but from the viewpoints of economical feasibility, availability of soil and simplicity in preparation method, the MgO impregnated akadama ceramic media might be a low-cost effective adsorbent and easy solution for the treatment of Hg contaminated effluents.

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