

Magnesium oxide-impregnated tuff soil-derived ceramic: a novel cadmium(II) adsorbing media

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Abstract The contamination of cadmium (Cd) in the aquatic environment is one of the serious environmental and human health's risks. The present study attempted to develop the potential magnesium oxide (MgO)-impregnated tuff soil-derived ceramic (MITDC)-based novel adsorbent media for adsorbing higher rate of cadmium [Cd(II)] from water phase. A potential MITDC adsorbent media was developed using volcanic raw tuff soil and its Cd(II) adsorption capacity from water phase was evaluated comparing with the raw tuff soil. A series of studies were carried out in an agitated batch method at 20 ± 2 °C to characterize the adsorption capacity of MITDC under different conditions of factors, such as contact time (0–360 min), initial pH (3–11) of solution, dose of MITDC (2, 5, 7.5 and 10 g/L), and initial concentration of Cd(II) (5, 10, 20, 30, and 40 mg/L), influencing the adsorption mechanism. MITDC exhibited the equilibrium state of maximum Cd(II) adsorption at the contact time 120 min and pH 4.7 (removed 98.2 % Cd) when initial Cd(II) concentration was 10 mg/L in the present study. The dose

of 7.5 g MITDC/L showed maximum removal of Cd(II) from water. Experimental data were described by the Freundlich and the Langmuir isotherms and equilibrium data fitted well with the Langmuir model ($R^2 = 0.996$). The Cd(II) adsorption capacity of MITDC was 31.25 mg/g. The high Cd(II) adsorption capacity indicated that novel MITDC could be used as a potential ceramic adsorbent media to remove high rate of Cd(II) from aqueous phase.

Keywords Magnesium oxide · Tuff soil · Impregnation · Cadmium ion · Adsorption · Isotherm

Introduction

Heavy metals are nonbiodegradable hazardous priority pollutants (Tewari et al. 2005; Bayrak et al. 2006) having significant properties of bioaccumulation and biomagnification. Indiscriminate and uncontrolled discharge of hazardous heavy metals into the environment by means of anthropogenic as well as geogenic activities leads to the contamination of water, soil, and air. The heavy metal contamination has been identified as one of the prime environmental problems worldwide due to posing hazardous and detrimental impacts on all forms of life (Kratohvil and Volesky 1998; Gupta and Gupta 1998; Aksu 2005; Bolger and Szlag 2002; Bidstrup 1964; Lin et al. 2000).

Cadmium (Cd) is one of the biologically nonessential, nonbiodegradable, persistent type of priority hazardous and heavy metal pollutants (Campbell 2006), which easily accumulates in sediments and aquatic organisms (by bioaccumulation and biomagnifications), and thus causing a gross biological impact. It is poisonous for plants, animals, and humans (Gupta and Gupta 1998) and recognized

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as a human carcinogen (IARC 1994). Human exposure to low level of Cd can result in renal diseases, osteomalacia, and lung cancer as well as damage the cardiovascular system, liver, and reproductive system (USEPA 1992; Hrudey et al. 1995; Belimov et al. 2005). Excessive intake of Cd leads to damage of human enzyme tissue, lung, kidney and renal system, skeletal deformation (Japanese itai-itai diseases), cardiovascular diseases, and hypertension (Nogawa et al. 2004; Argun et al. 2007).

Mining and smelting processes of lead and zinc, nickel-cadmium batteries, polyvinyl chloride plastics, paint pigments, insecticides, fungicides, and commercial fertilizer industries are the primary sources for causing Cd contamination as well as for causing the aforesaid tremendous environmental and human health risks. However, the effluent of Cd is a big environmental problem, since Cd is variously used as important component in producing different commodities required for daily life of human. In recent years, therefore, Cd removal from industrial wastewater is an important issue. Various technologies, chemical precipitation, ion exchange, membrane filtration, carbon adsorption, and co-precipitation, have been developed for the removal of heavy metals and many other pollutants (such as dyes) from aqueous solution over the years (Kentish and Stevens 2001, Saleh and Gupta 2012a, Gupta et al. 2012a, b). Adsorption is one of the effective techniques in removing Cd from wastewater (Aggarwal et al. 1999; Bhakta and Munekage 2009) because its sorption process largely depends on surface area, porous character, and chemical nature of adsorbent. Various relatively low-cost adsorbents have been developed and used for the removal of Cd and other various heavy metals [Such as Lead (Pb), Mercury (Hg), Copper (Cu), etc.] from aqueous solution (Sari and Tuzen 2009) such as silica ceramic (Salim et al. 2008), chitin (Benguella and Benaissa 2002), wood (Rafatullah et al. 2012), granular red mud (GRM) (Zhu et al. 2007a, b), fertilizer waste-derived activated carbon (Gupta et al. 1998), Mn₂O-loaded D301 resin MITDC and RT (Zhu et al. 2007a, b), alumina-coated multiwalled carbon nanotubes (Gupta et al. 2011), and manganese dioxide-coated multiwalled carbon nanotube (Saleh and Gupta 2012b). Soil, mud, waste materials, and its derivatives have great potential as inexpensive and efficient sorbents of various pollutants including metals and dyes due to their easy availability, chemical and mechanical stability, high surface area, and favorable structural properties (Gupta et al. 1998, Mittal et al. 2009a, b, 2010, Mittal et al. 2010, Vakili et al. 2014). Bhakta and Munekage (2013) identified some soil adsorbents including tuff soil (Meadow soil association, Japan) as potential media for removing hazardous metal(loid)s (cadmium, lead, and arsenic) from water phase. The studies also proved the ceramics derived from soil is a promising media

for removing heavy metals from water (Bhakta and Munekage 2009; Bhakta et al. 2009). There is an upsurge of interest in recent years to utilize natural minerals for the remediation of Cd-contaminated environment (Gupta and Bhattacharyya 2006; Lackovic et al. 2004). It has also been proposed that tuff soil has high Cd removal ability from water (Bhakta and Munekage 2013). However, no study has been attempted so far to develop the magnesium oxide (MgO)-impregnated tuff soil-based low-cost ceramic adsorbent media in order to remove the high rate of Cd from aqueous medium. Therefore, this study has firstly attempted to develop a potential tuff soil-based low-cost novel ceramic adsorbent media impregnating MgO for the removal of higher rate of Cd(II) from aqueous phase. The higher Cd(II) removal capacity of developed ceramic was evaluated comparing with the raw tuff (RT) under different conditions of physicochemical factors influencing the adsorption mechanism.

Materials and methods

Chemicals

The present study used the following chemicals: hydrochloric acid (HCl), sodium hydroxide (NaOH), magnesium oxide (MgO), and cadmium chloride (CdCl₂) were purchased from Nacalai Tesque, Inc. and Kanto Chemical Co., Inc., Tokyo, Japan. All synthetic and stock solutions were prepared using milli Q water of Millipore (Millipore, MA, USA).

Synthesis of magnesium oxide (MgO)-impregnated tuff soil-derived ceramic (MITDC)

Volcanic raw tuff soil (consists of ash, fragments of rock, and other miscellaneous materials) was obtained from Shimane peninsula, Japan and used for preparation of MgO-impregnated tuff soil-derived ceramic (MITDC) according to the following procedure: raw tuff (RT) soil [CaO (65.36 %), SiO₂ (21.83 %), FeO (7.7 %), Al₂O₃ (4.13 %), and MgO (1.29 %); specific surface area 8.69 m²/g] was dried by oven at 110 °C for 4 h, ground, and sieved to get uniform granules of RT. A screening process was performed in order to select the metal oxide having the high Cd(II) removal capacity (data not shown), and magnesium oxide (MgO) was selected as high Cd(II) up taking agent among the metal oxides tested. The selected magnesium oxide (MgO) was mixed with tuff soil at the rate of 2, 5, and 10 % to determine the best impregnation dose of MgO. The soft dough of tuff soil and MgO mixture was made by adding distilled water. Finally, the dough was air dried for 48 h and burned in different

temperatures 300, 700, and 900 °C for 5 h in order to activate and obtain the ceramic structure. After cooling, the hard composites of three MgO percentages (2, 5 and 10 %) were ground, sieved to get an equal particle size, and stored in capped glass bottles for screening and selection of optimum percent of MgO and activation temperature. Further a screening study determined that 5 % MgO-impregnated tuff soil-derived ceramic activated by 700 °C (herein called as MITDC) was the best for removing high rate of Cd(II) from aqueous solution (data not shown). Similar process was followed to develop the adsorbent media used for removing Cd(II) by adsorption on GRM (Zhu et al. 2007a, b) and mercury (Bhakta and Munekage 2011) from aqueous solutions.

Physical and chemical properties

The morphological properties and chemical composition of RT and MITDC were analyzed by scanning electron microscopy and energy dispersive spectroscopy (SEM–EDS, JEOL-JSM-6500F, Tokyo, Japan) in the Center for Advanced Marine Core Research, Kochi University, Japan using the process followed by Bhakta and Munekage (2013). Specific surface area of the RT particles (as shown in Synthesis of MITDC section) was measured by means of specific surface analyzer [YUASA-IONICS NOVA 2000].

Batch adsorption experiments

The Cd(II) adsorption experiments were carried out using a batch equilibrium technique to determine the optimum process parameters required for maximum Cd(II) adsorption of MITDC and RT from water. Experiments were conducted in 200-mL capped glass bottles using known weight of the adsorbents to equilibrate the known volume of adsorbate (100 mL). The effect of contact periods was determined by maintaining the initial concentration 10 mg/L, pH 5.6, and adsorbent dose 0.5 g/L. The effects of different initial pH (3–11) values on the Cd adsorption of adsorbents were determined by adjusting the pH (by 0.1 N HCl and 0.1 N NaOH) and maintaining the conditions, initial concentration of Cd(II) 10 mg/L and adsorbent dose 5 g/L. The effects of adsorbent dosages were determined using 2, 5, 7.5 and 10 g/L of adsorbent and 10 mg/L of adsorbate [Cd(II)] solution with pH 5.6. To determine the effects of initial adsorbate concentrations, the initial Cd(II) concentrations of 5, 10, 20, 30, and 40 mg/L with pH 5.6 and adsorbent dose of 0.5 g/L were used. Each experiment was performed at least twice following the identical conditions using controls of only Cd(II) solution and adsorbents (MITDC and RT) without metals in solution. All experiments were conducted using the Cd(II) solution prepared from standard stock solution of CdCl₂. In each

experiment, the capped glass bottles were shaken by mechanical shaker at the rate of 150 excursion/min within the thermostat incubator at 20 ± 2 °C.

Sampling and analysis

The samples (1 mL) were collected from glass bottles, filtered by 0.20 µm filter, centrifuged, and analyzed using an atomic absorption spectrometer (AAS 200, PerkinElmer) with 228.8 nm analytical wavelength of Cd(II) to determine the residual metal concentration in samples. Mean data obtained from least two experiments were used for analysis.

The amounts of Cd(II) adsorbed onto the MITDC and RT (mg/g) at the equilibrium state (q_e) and at specific time (q_t) were calculated by following the Eqs. 1 and 2 of mass balance relationship (Rozada et al. 2008; Bhakta and Munekage 2013; Bhakta et al. 2014a, b):

$$q_e(\text{mg/g}) = \left[\frac{C_i - C_e}{M} \right] \times V \quad (1)$$

$$q_t(\text{mg/g}) = \left[\frac{C_i - C_t}{M} \right] \times V, \quad (2)$$

where q_e (mg/g) is the amount of Cd(II) adsorbed per gram of the adsorbent, C_i (mg/L) is the initial concentration of Cd(II) in the solution, C_t (mg/L) is the concentration of Cd(II) in the solution at time t , C_e (mg/L) the equilibrium concentration of the solution of Cd(II), V (L) is the volume of the solution, and M (g) is the mass of the adsorbent.

The percentages of Cd(II) removed (R_e) by the MITDC and RT were determined using the following Eq. 3:

$$R_e(\%) = \left[\frac{C_i - C_e}{C_i} \right] \times 100, \quad (3)$$

where R_e is the percentage (%) of Cd(II) adsorbed, and C_i and C_e are the initial and equilibrium concentration of Cd(II) (mg/L) in the solution, respectively.

Results and discussion

Physical and chemical properties

The SEM images of RT and MITDC shown in Fig. 1 are representing their morphological properties. The microscopic images (10,000×) clearly reveal the elaborate surface structures of RT and MITDC. The MITDC surface structure was granular, whereas RT showed more smooth surface structure. The EDS analysis indicates that RT is composed of CaO (65.36 %), SiO₂ (21.83 %), FeO (7.7 %), Al₂O₃ (4.13 %), and MgO (1.29 %) and MITDC is constituted by CaO (63.21 %), SiO₂ (19.6 %), FeO (6.84 %), Al₂O₃ (4.03 %), and MgO (6.04 %).

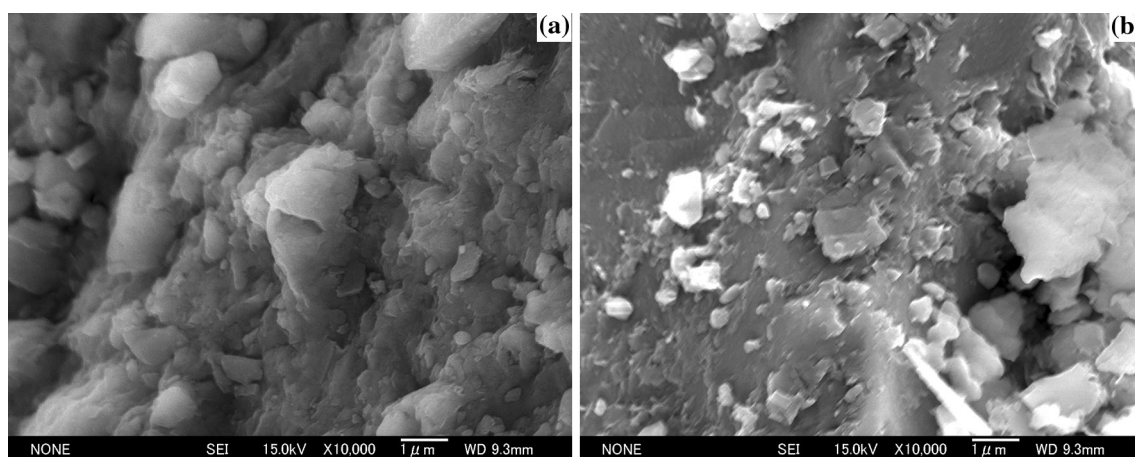


Fig. 1 SEM microimages (10000×) of RT (a) and MITDC (b)

It may be suggested that impregnated MgO (5 %) formed granular crystal structure by thermal (700 °C) activation in MITDC.

Effect of contact time

The effect of contact time on the adsorption of Cd(II) onto the MITDC and RT is shown in Fig. 2. The metal adsorption is rapid in the first 90 and 30 min of contact period for MITDC and RT, respectively. The necessary optimum times were about 120 min for MITDC and 90 min for RT to reach the equilibrium state. Beyond these contact times, the amount of Cd(II) adsorbed on the MITDC and RT remained constant as shown in Fig. 2. The amounts of Cd(II) adsorbed at equilibrium are 16.6 mg/g for MITDC and 1.98 mg/g for RT in this experiment. The amount of Cd(II) adsorbed onto MITDC was much higher than the values obtained by RT at the same interaction time period (360 min). These observations are similar with the works of Alemayehu and Lennartz (2009), Benaissa (2006), and Benguella and Benaissa (2002).

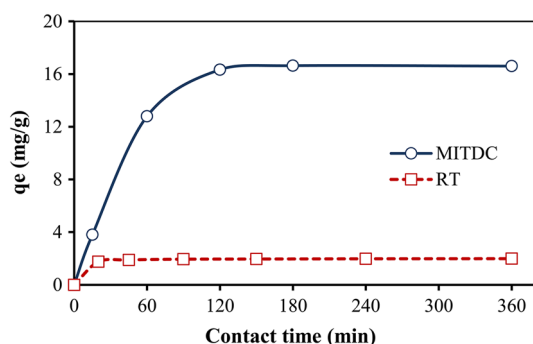


Fig. 2 Effect of contact time on adsorption of Cd(II) by MITDC and RT (initial concentration of Cd(II) 10 mg/L, pH 5.6, adsorbent dose 0.5 g/L)

Effect of solution pH

The effects of different pH values on adsorption efficiency of MITDC and RT are represented in Fig. 3. The removal efficiency was high between the pH 4 and 8 and slightly decreased at pH 9 and increased with increasing pH thereafter in MITDC and RT. The MITDC showed maximum removal (98.2 %) at a pH value of 4.7, whereas RT exhibited highest adsorption of Cd(II) at 7.1 and 10.5. It indicated that though favorable adsorption takes place at pH 4.3–8, optimum pH would be 4.7 for MITDC to remove maximum Cd(II). In relation to the Cd speciation diagram, Cd(II) is the predominant ionic species at pH less than 7 (Baes and Mesmer 1986). Similar results were reported for the adsorption of Cd(II) from aqueous phase onto eucalyptus bark (Ghodbane et al. 2008) and manganese oxide-loaded resin (Zhu et al. 2007a, b). Considering the above results, it is fact that the effect of pH in the sorption process of sorbent can be explained by the surface charge of adsorbent materials and the specification of Cd(II). Srivastava (2013) reported that the effect of solution pH can

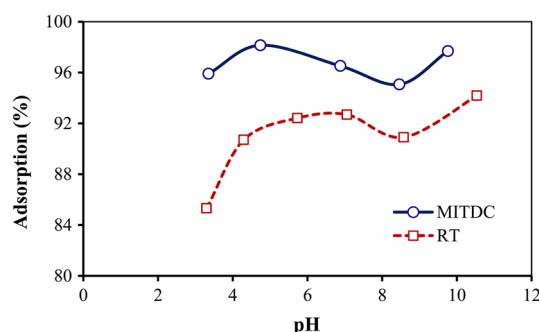


Fig. 3 Effect of pH on the adsorption of Cd(II) by MITDC and RT. (Initial concentration of Cd(II) 10 mg/L, contact time 120 min, adsorbent dose 5 g/L)

be explained by considering the surface charge of the carbon nanotubes and the degree of ionization and speciation of the sorbates.

Effect of adsorbent dose

Figure 4a and b elucidates the effects of different adsorbent dosages on the Cd(II) adsorption of MITDC and RT. The removal efficiency was higher in MITDC (85–95 %) than that of the RT (30–40 %) within the 15 min contact time. It can be seen in the Fig. 4b that the percentage of Cd(II) removal increased sharply up to adsorbent dose 7.5 mg/L and steady thereafter. It demonstrated that the adsorbent dose 7.5 mg/L is optimum for maximum Cd(II) adsorption in the experiment. It is due to the increased available surface area of adsorbent in the higher adsorbent doses, and thus, high amount of Cd(II) is attached to their surfaces (Gupta and Bhattacharyya 2006). The results also demonstrated that though total Cd(II) adsorption increased with increasing the dosages, the amount of adsorption per gram of ceramic (i.e., q_e) was higher in lower dosages than that of the higher dosages in this experiment. The decreasing trend of q_e value 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). It is also obvious that the competition between surface site of adsorbent and Cd(II) is higher in higher doses resulting in the decrease of q_e value.

Effect of initial concentration

The amount of Cd(II) adsorbed per unit mass of MITDC and RT increased gradually with an increasing Cd(II) concentration of the solution (Fig. 5). By changing the initial concentration of the solution from 5 to 40 mg/L, the absolute amount of adsorbed Cd(II) increased from 9.38 to 30.83 mg Cd(II) per gram of MITDC and from 1.99 to 7.37 mg of Cd(II) per gram of RT, respectively. The results suggested that the maximum q_e values of MITDC and RT were 30.83 and 7.37 mg/g, respectively, maintaining the environmental conditions employed in the present experiment. This trend is similar to adsorption of Cd(II) from aqueous solution by two different rock types Pumice and Scoria (Alemayehu and Lennartz 2009).

Adsorption isotherms

An adsorption isotherm is the relationship between the adsorbate in the liquid phase and the adsorbate adsorbed on the surface of the adsorbent at the equilibrium state. The equilibrium adsorption isotherms are useful in understanding the sorption interaction of importance in the design of adsorption systems. Several isotherm models such as Langmuir isotherm and the Freundlich isotherm models have been published in several literatures to describe and evaluate the experimental data of adsorption isotherms, which show the different adsorption mechanisms. In this study, the Langmuir and Freundlich isotherms were described.

The Langmuir adsorption isotherms assume that adsorption takes place at specific homogeneous sites within

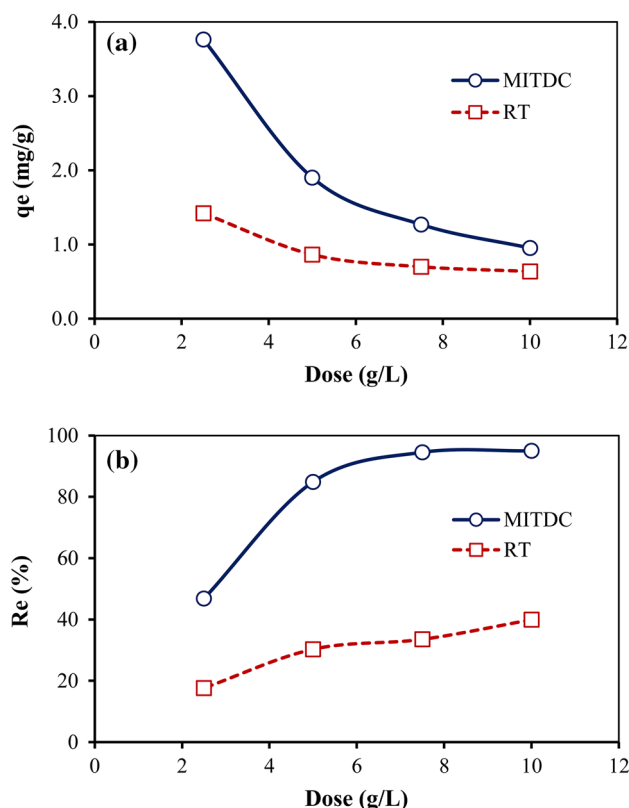


Fig. 4 Effect of adsorbent dose on adsorption of Cd(II) by MITDC and RT (pH 5.6, Cd(II) concentration 10 mg/L); **a** q_e vs dose and **b** R_e vs dose

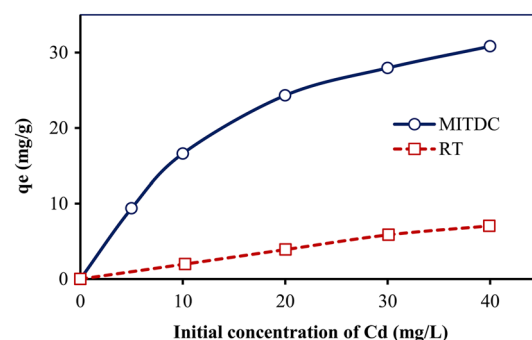


Fig. 5 Effect of initial concentration on adsorption of Cd(II) by MITDC and RT (pH 5.6, adsorbent dose 0.5 g/L)

Table 1 Langmuir and Freundlich isotherm parameters for adsorption of Cd(II) by MITDC and RT

Parameters	MITDC	RT
Langmuir isotherm		
q_m (mg/g)	31.25	9.497
b (L/mg)	3.56	1.444
R^2	0.998	0.996
Freundlich isotherm		
k_f (mg/g)	20.34	4.785
n	6.94	2.55
R^2	0.973	0.8059

the adsorbent and have found successful application to many sorption process of monolayer adsorption. The Langmuir adsorption isotherm can be written as

$$q_e = \frac{q_m b C_e}{1 + b C_e} \quad (4)$$

The Langmuir parameters were obtained by fitting the experimental data to the linearized equation derived from Eq. (4) which is Eq. 5 as follows:

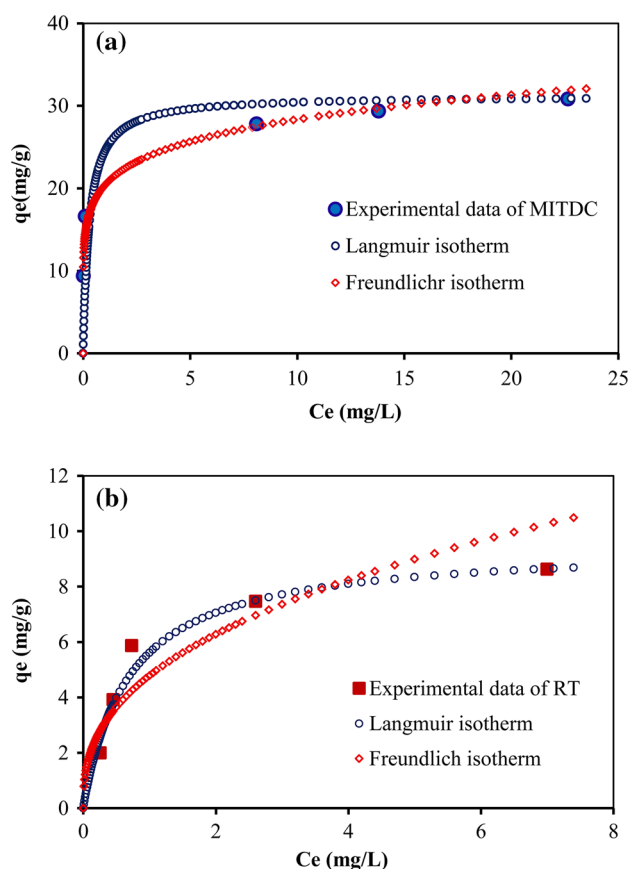
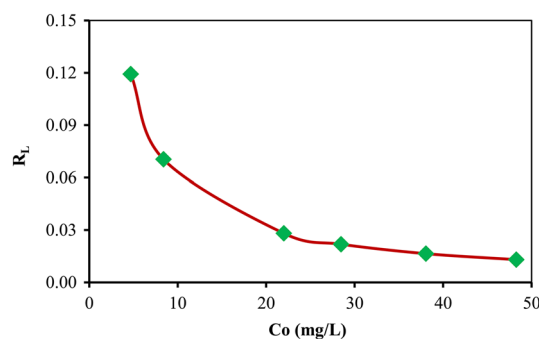
$$\frac{C_e}{q_e} = \frac{1}{b q_m} + \frac{C_e}{q_m}, \quad (5)$$

where q_e is the adsorbent amount (mg/g) of the Cd(II), C_e is the equilibrium concentration of the Cd(II) in solution (mg/L), q_m is the monolayer adsorption capacity (mg/g), and b is the constant related to the free energy of adsorption (L/mg).

Based on Eq. 5, the isotherms were fitted to the adsorption data obtained. The Langmuir adsorption exponents for Eq. 5, the q_m and b , are determined from the linear plots of C_e/q_e versus C_e (Figure not shown), and the calculated correlation coefficients for these isotherms are shown in Table 1. The values of the Langmuir constant were calculated from the slopes and intercepts of the plots. The magnitude of Langmuir constant b is 3.56 L/mg for MITDC and 1.44 L/mg for RT. The adsorption capacity q_m was determined as 31.25 mg/g for MITDC (Fig. 6a) that is larger than RT (9.49 mg/g) (Fig. 6b). In order to predict the adsorption efficiency of the adsorption process, the dimensionless equilibrium parameter R_L was determined by using the following Eq. 6 (Magdy and Daifullah 1998):

$$R_L = \frac{1}{(1 + b C_i)}, \quad (6)$$

where C_i is the initial concentration and b is the Langmuir isotherm constant. The parameter R_L indicates the shape of isotherm accordingly, as explained in Table 1. The Fig. 7 shows that the R_L values at different initial Cd(II) concentrations for MITDC indicating a highly favorable adsorption.

**Fig. 6** Langmuir and Freundlich isotherms for Cd(II) adsorption onto MITDC (a) and RT (b) at different initial concentrations**Fig. 7** Separation factor for Cd(II) adsorption by MITDC

The Freundlich isotherm is an experimental equation in use to describe heterogeneous systems:

$$q_e = K_f C_e^{\frac{1}{n}} \quad (7)$$

The linearized form of Freundlich equation is

$$\log q_e = \log K_f + \frac{1}{n} \log C_e, \quad (8)$$

where q_e is the adsorbent amount (mg/g) of the Cd(II), C_e is the equilibrium concentration of the Cd(II) in solution

Table 2 A comparative Cd sorption capacities of different adsorbents related to MITDC (as literature available with us)

Adsorbent	Modifying agent	q_{\max} (mg/g)	Reference
Tuff soil	Magnesium oxide	31.25	Present study (MITDC)
Tuff soil	–	9.49	Present study (RT)
Soil, Fox	–	17.0	Bolton and Evans (1996)
Chemically treated clay	–	12.6	Samir (2008)
Montmorillonite	–	40.82	Sun et al. (2014)
Red mud	–	13.03	Gupta and Sharma (2002)
Fly ash, treated	–	14.33	Chaiyasith et al. (2006)
Manganese nodule residue	–	21.2	Rout et al. (2009)
Low grade manganese ore	–	59.17	Mohapatra et al. (2008)
<i>Azolla filiculoides</i> (aquatic fern)	Magnesium chloride	86	Ganji et al. (2005)
Juniper fiber	Sodium hydroxide	29.54	Min et al. (2004)
Rice husk	Sodium hydroxide	20.24	Kumar and Bandyopadhyay (2006)
<i>Syzygium cumini</i> leaf powder	–	34.54	Rao et al. (2010)
Chemically modified peanut shell	–	95.24	Xu and Zhuang (2014)

(mg/L), K_f is the adsorption capacity of Freundlich constant (mg/g), and n is the adsorption intensity of Freundlich adsorption isotherm constant. The Freundlich adsorption (Fig. 6a and b) exponents for Eq. 8, the K_f , and n , are determined from the linear plots of $\log q_e$ versus $\log C_e$, and the respective data are shown in Table 1. Generally, it is stated that the values of n in the range of 1–10 represent good adsorption (Magdy and Daifullah 1998). In this study, the Freundlich constant n was $1 < n < 10$, representing favorable adsorption.

As shown in Table 1 and Fig. 6a and b, the Langmuir equation representing adsorption process fitted very well; the correlation coefficient, R^2 , value is higher for the Langmuir isotherm than that for the Freundlich isotherm, indicating a good mathematical fit (Langmuir isotherm $R^2 = 0.996$, Freundlich isotherm $R^2 = 0.8059$). The value of adsorption capacity of MITDC (q_m 31.25 mg/g) is comparatively greater than RT (q_m 9.49 mg/g) and some other adsorbent media developed by several studies (Table 2). Therefore, the MITDC can be used as an effective Cd(II) removal adsorbent from aqueous media. Saleh and Gupta (2012a, b) employed manganese dioxide-coated multiwalled carbon nanotube for removing lead(II) removal from water.

Conclusions

This study developed a novel magnesium oxide (MgO) MITDC, which showed a high rate of Cd(II) removal capacity compared to that of the raw tuff soil (RT) from aqueous solutions. The adsorption capacity of tuff soil was significantly improved by impregnation of magnesium

oxide (5 %) and heat activation. Although adsorption equilibrium data follow both Langmuir and Freundlich isotherm models, the Langmuir isotherm model fitted well with the experimental data. The capacity of Cd(II) adsorption was 31.25 mg/g, calculated from Langmuir isotherm using other optimum study conditions obtained. The present adsorption kinetics clearly revealed that Cd(II) adsorption of MITDC is largely influenced by initial pH, adsorbent amount, and initial concentration of ambient solution. The contact time 120 min, pH 4.7, and adsorbent dose 7.5 were supposed to be the optimum process parameters for maximum Cd(II) adsorption of developed novel MITDC adsorbent media. The dimensionless separation factor (R_L) showed that MITDC was favorable for removal of cadmium from aqueous solutions. On account of the above characteristics, it might be concluded that MgO MITDC can potentially be used as an easily accessible, inexpensive, and effective adsorbent media for removing high rate of cadmium from aqueous phase.

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Conflict of interest The author declares that there is no conflict of interests regarding the publication of this article.

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