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Research article

Adsorptive removal of arsenic by novel iron/olivine composite: Insights into preparation and adsorption process by response surface methodology and artificial neural network

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ABSTRACT

Olivine, a low-cost natural material, impregnated with iron is introduced in the adsorptive removal of arsenic. A wet impregnation method and subsequent calcination were employed for the preparation of iron/olivine composite. The major preparation process parameter, viz., iron loading and calcination temperature were optimized through the response surface methodology coupled with a factorial design. A significant variation of adsorption capacity of arsenic (measured as total arsenic), i.e., 63.15 to 310.85 mg/kg for arsenite [$As(III)^T$] and 76.46 to 329.72 mg/kg for arsenate [$As(V)^T$] was observed, which exhibited the significant effect of the preparation process parameters on the adsorption potential. The iron loading delineated the optima at central points, whereas a monotonous decreasing trend of adsorption capacity for both the $As(III)^T$ and $As(V)^T$ was observed with the increasing calcination temperature. The variation of adsorption capacity with the increased iron loading is more at lower calcination temperature showing the interactive effect between the factors. The adsorbent prepared at the optimized condition of iron loading and calcination temperature, i.e., 10% and 200 °C, effectively removed the $As(III)^T$ and $As(V)^T$ by more than 96 and 99%, respectively. The material characterization of the adsorbent showed the formation of the iron compound in the olivine and increase in specific surface area to the tune of 10 multifold compared to the base material, which is conducive to the enhancement of the adsorption capacity. An artificial neural network was applied for the multivariate optimization of the adsorption process from the experimental data of the univariate optimization study and the optimized model showed low values of error functions and high R^2 values of more than 0.99 for $As(III)^T$ and $As(V)^T$. The adsorption isotherm and kinetics followed Langmuir model and pseudo second order model, respectively demonstrating the chemisorption in this study.

1. Introduction

Arsenic present in the earth crust as the 12th most abundant element as well as in human body as the 20th (Mandal and Suzuki, 2002). This ubiquitous trace element is recognized as toxicant and is classified as a Class I human carcinogen by the International Agency for Research on Cancer (IARC) (Chen et al., 2009). The chronic exposure to arsenic results in cancer in skin, lung or bladder (Ng et al., 2003). Additionally, hyper and hypopigmentation, keratosis, non-pitting edema of feet/hands, multiple organ damages, such as lung, liver, cardiovascular or pulmonary system, etc. are associated with the arsenic intake (Guha Mazumder and Dasgupta, 2011; Ng et al., 2003). A prominent correlation between epidemiological evidence of the arsenic poisoning and the prevalence of arsenic in drinking water attract global attention towards arsenic pollution (Rossman, 2003). The prevalence of arsenic is predominantly found in Bangladesh, India, Chile, China, Ghana, United States, etc. affecting a population to the tune of 200 million (Naujokas et al., 2013; WHO, 2008). The arsenic contamination in ground water mainly occurs naturally from arsenic bearing minerals and mostly present in trivalent or pentavalent forms (Duker et al., 2005). The public health hazard associated with arsenic lead to enforce a stringent standard of arsenic in drinking

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water as 10 μg/L (WHO, 2011). Nevertheless, the removal of arsenic is one of the biggest challenges in the drinking water sector and creates tremendous stress in the research area in the field of environmental management.

Many treatment technologies have been attempted for the elimination of arsenic from drinking water. The conventional methods, such as oxidation precipitation, coagulation-precipitation and filtration, co-precipitation, adsorption, etc. and some advanced techniques, i.e. electrocoagulation, membrane filtration, reverse osmosis, etc. are used for arsenic removal (Jadhav et al., 2015; Mohan and Pittman, 2007). Among them, adsorption is one of the preferred technique for technical simplicity and economic viability (Lata and Samadder, 2015; Mohan and Pittman, 2007). Many studies have been conducted for the adsorptive removal of arsenic species, which bring forth many natural and synthetic adsorbent (Chaudhuri and Mohammed, 2012; Kundu and Gupta, 2007a, 2007b). Among them, iron-based substances are found to have strong affinity and high selectivity for the inorganic arsenic species in the sorption processes, which results in predominantly use of the various form of iron, mostly as oxides, for arsenic removal (Chowdhury and Yanful, 2010; Dickson et al., 2017; Wen et al., 2014). Furthermore, the vast applicability of iron lies in its well-known low-cost, eco-friendly nature and non-occurrence of secondary pollution after treatment. However, one of the biggest challenges in using most of the metal oxides in sorption studies is their low specific surface area and difficulties in separation after treatment. The use of composite materials consisting of a base material supporting metal oxides stems from this concern.

Olivine is a naturally abundant mineral rich in magnesium, silica, and iron with the chemical composition $(Fe_xMg_{1-x})_2SiO_4$ (Świerczyński et al., 2006). This sandy material is mechanically hard and attrition resistant, which made it acceptable as a potential support material in the field of catalysis (Zhao et al., 2015). Furthermore, the composite catalyst prepared by impregnation of olivine with metal is employed to increase the catalysis potential of olivine. Amongst, Fe/olivine composite is predominantly used as a catalyst for biomass gasification, steam reforming, etc. (Quan et al., 2017; Rapagnà et al., 2011; Virginie et al., 2010a, 2010b). The activation of olivine is drastically influenced by the quantum and form of iron oxide. Thus, the iron-impregnated on the natural olivine is considered to be a low-cost, eco-friendly potential catalyst (Rapagnà et al., 2011). The application of olivine on water treatment has not been explored so far. The present study originated from this gap area.

The preliminary experimentation proved that olivine can remove arsenic from water by adsorption. Hence, the olivine has been chosen as a base material for iron impregnation. The percentage of iron used for impregnation and subsequent calcination temperature are considered as the major influencing factor. Both the factors are crucial in deciding the content or phase of iron as well as the activity of olivine. The individual and interactive effect of the iron content and the calcination temperature on the catalyst activity has not been explored. In this context, the role of iron content and calcination temperature on the adsorptive potential of iron impregnated olivine targeting the arsenic removal may be essential to explore the future of the metal impregnated olivine in the adsorptive removal of pollutant.

The preparation of metal/olivine composite material mostly performed with wet impregnation method. The metal content and the calcination temperature are varied in few studies using one variable at a time (OVAT) experiment (Virginie et al., 2010a, 2010b). However, the OVAT study is incapable of predicting the combined effect of influencing parameters as well as the overall optima of the process conditions (Bezerra et al., 2008). Application of multivariate optimization may be considered as a useful tool to overcome this drawback. The response surface methodology (RSM) is found to be one of the widely adopted techniques in this direction (de Sales et al., 2015; Ghosal et al., 2015; Ghosal and Gupta, 2016; Sasidharan Pillai and Gupta, 2016; Zheng et al., 2015). Although RSM is significantly used to optimize the adsorption process, the optimization of the preparation process of the adsorbent is conducted in very few studies (Ghosal et al., 2015; Yadav et al., 2017). The pre-requisite of a multivariate optimization process is a suitable design of experiment (DoE) with the considerable efficiency and accuracy. A proper DoE coupled with RSM may demonstrate the overall optima of the system and the interactive effects of influencing parameter, which in turn encourage the production of the adsorbent with the maximum efficiency.

The adsorption process is mostly dependent on various parameters which in turn influence the performance of adsorbent. Normally the OVAT experiments are conducted to appraise the effect of various parameters. Apart from that, multivariate optimization through RSM had been conducted in many studies. However, the design of the experiment for more than four parameters was rarely conducted as the required number of the experimental run will be sufficiently high which is difficult to handle. In this context, artificial neural network (ANN) can be efficiently used for any set of experimental data as the design of experiment is not mandatory. Hence, an ANN network from OVAT experiment data of univariate optimization study can be used to model the adsorption process with a large number of input variables, which in turn reduce the requirement of number of experiments. Multivariate optimization modeling of the adsorption process for the various influencing factor from OVAT experiment data is a gap area in this field of environmental management.

In this paper, the arsenic removal potential of iron oxides and the mesoporous natural base material olivine has been chosen to prepare a composite adsorbent. A simple and low-cost technique for the preparation of Fe/olivine composite material was demonstrated through a wet impregnation method and the subsequent calcination. The major influencing factors for the preparation process were identified as the iron loading and the calcination temperature, which were optimized through a 3^2 factorial design coupled with RSM. The removal of arsenic (as total arsenic) from the aqueous solution of major prevalent species of arsenic, i.e., As(III) and As(V), were maximized as the responses in the multivariate optimization study. The physicochemical properties of the adsorbent are addressed through characterization of different sample prepared in the various process conditions in a systematic manner to explore the influence of preparation process parameters on the properties of the mesoporous composite material. The multivariate optimization of the adsorption of arsenic was modeled by ANN from the OVAT experimental data. The adsorption process was also analyzed through the isotherm and kinetic modeling.

2. Materials and methods

2.1. Reagents, chemicals and instrumentations

All the chemicals used in the study were of analytical grade and were used without further purification. De-ionized (DI) water was used for the preparation of all the solutions. The base material used was natural olivine (collected from Mahadevan Enterprises, Salem, India) of particle size that passes through the 150 μm sieve and retains on 45 μm. The iron salt solution used for the impregnation process was prepared by dissolving ferrous chloride $FeCl_2$, XH_2O (Merck), in DI water.

The inorganic forms of arsenic, i.e.; arsenite [As(III)] and arsenate [As(V)] were considered in this study. As(III) and As(V) stock solutions were prepared by dissolving sodium As(III), $NaAsO_2$