Zeolite supported mono- and bimetallic oxides: Promising adsorbents for removal of As(V) in aqueous solutions

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HIGHLIGHTS

▶ Monometallic (Fe or Al) and bimetallic (Fe—Al) oxides were supported on zeolite.
▶ The presence of Al or Fe in zeolite enhanced As(V) removal capacity significantly.
▶ The bimetallic oxide associated zeolite had the highest adsorption capacity.
▶ The sorption patterns fit well to the Freundlich and D–R models.

ARTICLE INFO

ABSTRACT

In this study, synthesis and characterization of novel adsorbents based on natural zeolite (clinoptilolite) supported mono- (Fe or Al) and bimetallic (Fe—Al) oxides for adsorption of arsenate from aqueous solution were investigated. Batch sorption experiments were carried out at different pH (3.0, 5.0 and 7.0), temperature (298, 318 and 338 K) and initial metal concentration (0.5, 5.0 and 9.5 mg L^{-1}) values. The experimental results showed that aluminum (ZNa—Al) and iron (ZNa—Fe(II)) modified zeolites enhanced the arsenate uptake significantly. Furthermore, bimetallic oxide associated zeolite, ZNa—AlFe(III), showed highest adsorption capacity as compared to ZNa—Al and ZNa—Fe(II), indicating that aluminum impregnation can provide more adsorption sites (Al—OH) to As(V) anions. Maximum sorption capacities were achieved at pH 5 for ZNa (0.026 mg g^{-1}), ZNa—Al (1.50 mg g^{-1}) and ZNa—AlFe(III) (3.86 mg g^{-1}) and at pH 3 for ZNa—Fe(II) (3.02 mg g^{-1}). The results suggest that As(V) adsorption process is dominantly followed by surface complexation reactions for ZNa—Al and ZNa—AlFe(III) adsorbents while columbic interactions occur on the surface of ZNa—Fe(II). Dubinin–Radushkevich and Freundlich isotherm models fitted the equilibrium data better than Langmuir isotherm. The Gibbs free energy values were found as negative for all sorbents, which indicates the adsorption of arsenate on modified zeolites is feasible and spontaneous.

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1. Introduction

Arsenic is one of the growing public concerns all over the world due to its high toxicity and carcinogenicity to living organisms [1]. So far various health effects caused by arsenic have been reported [2]. It has been estimated that millions of people in Asia and America suffer from these diseases from the consumption of water contaminated with arsenic. Turkey is one of the countries where people at risk because of arsenic contaminated water according to Human Development Report [3]. The World Health Organization (WHO) and subsequently the US Environmental Protection Agency

lowered the maximum arsenic contaminant level in drinking water from 50 µg L^{-1} to 10 µg L^{-1} [4,5].

The arsenic contamination generally occurs in surface and drinking waters as well as in the industrial wastewaters. Common arsenic sources in water are arsenic-containing soils, sediments, and anthropogenic activities including metal mining, petroleum-refining, combustion of fossil fuels, semiconductors, arsenical herbicides, fungicides and insecticides [6]. In natural waters generally inorganic arsenic species exist in arsenite (H_3AsO_3) or arsenate ($H_2AsO_4^-$, $HAsO_4^{2-}$) forms and inorganic arsenic is more toxic than organic forms [2]. Technologies for the remediation of this metalloid can be listed in various categories including coagulation/flocculation [7], sorption and ion exchange [8,9], reverse osmosis, membrane separation [10] and biological methods [11]. Among

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