ORIGINAL ARTICLE

Lead separation by sorption onto powdered marble waste

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Abstract   Batch adsorption experiments were carried out, aiming to remove lead ions from aqueous solutions and water samples using powdered marble wastes (PMW) as an effective inorganic sorbent, which is cheap, widespread, and may represent an environmental problem. The main parameters (i.e., solution pH, sorbent and lead concentration, shaking time, and temperature) influencing the sorption process, were investigated. The results obtained showed that the sorption of Pb^{2+} ions onto PMW was fitted well with the linear Freundlich and Langmuir models over the concentration range studied. From the Dubinin–Radushkevick (D–R) isotherm model it was found that the adsorption was chemical in nature. Thermodynamic parameters *viz*. the change in Gibbs free energy change ($\Delta G°$), enthalpy ($\Delta H°$) and entropy ($\Delta S°$) were also calculated. These parameters indicated that the adsorption process of Pb^{2+} ions on PMW was spontaneous and endothermic in nature. Under the optimum experimental conditions employed, the removal of about 100% of Pb^{2+} ions was attained. The procedure was successfully applied to remove lead ions from aqueous and different natural water samples. Moreover, the adsorption mechanism is suggested.

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1. Introduction

The current pattern of industrial activity alters the natural flow of materials and introduces novel chemicals into the environment (Fasal and Hasnain, 2004; Igwe and Abia, 2006). The rate at which effluents are discharged into the environment especially water bodies has been increased as a result of urbanization. Toxic metal ions (among them is lead) are dangerous contaminants that are accumulated by the living organisms and, up to now, there are no widely accepted methods to have them removed. Therefore, the best solution is preventing the entrance of toxic metals in the ecosystem (Chubar et al., 2003). Furthermore, the removal of toxic metal ions and recovery of valuable metal ions from mine wastewaters, soils and waters have been important in economic and environmental viewpoints (Monteagudo and Ortiz, 2000; Capponi et al., 2006).

Lead is accumulated in the human system through a number of processes like breathing air from industrial environment (lead smelting, refining and manufacturing industries), ingesting plants or food grown on contaminated soils and breathing

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in fumes from hobbies that use lead (leaded-glass, ceramics). The health hazards resulting from the presence of lead in domestic and industrial wastewaters are of extreme concern to the public, government and industries for over the past decades up to now (Ho et al., 2001). Because of the great mobility, the heavy metal may reach the main source of drinking water such as surface and underground water (Ghazy and Ragab, 2007a). The deleterious effects of lead on neurobehavioral development (Dietrich et al., 1990) and brain cell function (Goldstein, 1990) have been investigated. The maximum allowable Pb^{2+} concentration in drinking water was restricted to be 20 ng ml^{-1} (Senkal et al., 2007) and by international regulations on water quality, by World Health Organization (WHO, 2000) and European Commission Directive (EUD, 1998). Hence, easy, effective, economic and eco-friendly techniques are required for fine tuning of effluent wastewater treatment. The search for a low cost and easily available adsorbent has led to the investigation of materials applicable to most treatment systems.

A number of technologies have been developed over the years to remove heavy metals from industrial wastewaters. The most important technology includes coagulation/flocculation (Amuda et al., 2006). Other conventional chemical methods include precipitation, ion-exchange, electrochemical processes, solvent extraction, and membrane technology. All the chemical methods have proved to be much costlier and less efficient (Preetha and Viruthagiri, 2005). Adsorption is one of the most interesting methods for the removal of heavy metals from various effluents. The main properties of adsorbents for heavy metal removal are strong affinity and high loading capacity (Sari et al., 2007). Natural organic or inorganic sorbents, which are particularly abundant and inexpensive, have generally these properties (Sari et al., 2007; Ghazy et al., 2005; Kumar et al., 2000). Therefore, the study of the adsorption process is of utmost importance for the understanding of how heavy metals are transferred from a liquid mobile phase to the surface of a solid phase (Bradl, 2004). Finally, researchers strive-themselves to find a low cost and easily available adsorbent.

Large amounts of marble are produced in some countries, such as the United States, Italy, Greece, Egypt and others. The by-product generated from marble processing is a powdered dust and may represent an environmental problem. Hence, the objective of this work was to study the possibility of utilizing PMW (naturally occurring and cheap) as a sorbent for removing lead ions from aqueous solutions and natural waters. The different parameters influencing the adsorption of lead ions onto powdered marble wastes were optimized and the results are presented in this paper.

2. Experimental

2.1. Materials and solutions

The powdered marble waste (PMW) samples used in this study were obtained, free of charge, from the dust of some private marble processing workshops located in Bilqas city, Egypt. The samples were sieved and those with size (25–63 µm) were used in the experiments. The samples contained 60% calcite, CaCO$_3$, and 6% dolomite, MgCO$_3$, (as found by chemical analysis), with the remainder being composed of common constituents such as quartz, feldspar, clays, pyrite and siderite

(Bates and Jackson, 1980). The samples were dried for 2 h in large trays in an oven maintained at 125 °C, allowed to cool to room temperature and then packed into stoppered bottles and stored in a desiccator for future use.

Functional groups of PMW were characterized through infrared analysis. The observable peaks at about 710, 875, 1420, 1807 and 2520 cm^{-1} coincided with pure CaCO$_3$. The surface area and porosity of PMW was measured using Brunauer, Emmett, Teller (BET) method. PMW samples have 18.9 Å pore diameter, 31.2 Å pore size, 0.98 cc/g pore volume and its measured surface area was 21.8 m^2/g. The pH values of points of zero charge (pHPZC) were 9.1 (not aged), 6.2 (aged 60 min) and 8.3 (aged several days) and this agreed with the previously reported data (Somasundran and Agar, 1967). Stirring or soaking of 1 g PMW sorbent in 100 ml distilled water (pH 6.9) for 1 h or more increased the suspension pH to 9.3, confirming the negative charge of the PMW surface.

Lead(II) stock solution (4 g/l) was prepared from Pb(NO$_3$)$_2$ (BDH Ltd., Poole, England) by dissolving 1 g in 250 ml doubly distilled water. The working solutions were made by diluting with doubly distilled water. Xylenol orange (XO) indicator (1%) was freshly prepared daily before use. Aqueous solutions of HNO$_3$ and NaOH were used for pH adjustment. Any other chemicals were prepared from certified reagent grade chemicals.

2.2. Apparatus

A Buck Scientific Accusys 211 Atomic Absorption Spectrophotometer (AAS), provided with micro-burettes 5 and 10 ml, was used for the determination of lead concentration at 217 nm. The infrared analyses were undertaken via a Mattson 5000 FT-IR spectrophotometer using KBr disc method. The pH was measured using a Jeanway 3311 pH meter, provided with a glass electrode. The shaking of solutions was carried out with a shaker Model (Heidolph promax 2020, Germany).

2.3. Procedure

Unless stated otherwise, all batch sorption experiments were done at room temperature ($\sim$25 °C). Known volumes of lead solutions were pipetted into quick-fit glass bottles containing 0.05 g of PMW sorbent in 50 ml aqueous solution to give concentrations ranging from 500 to 1500 mg/l. Since the pH of any resulting solution was $\sim$7.0, further controlling of pH was not necessary since pH 7 was suitable for most adsorption experiments. The resulting solution was then shaken at 270 rpm and the samples were taken at fixed time periods (1, 5, 10, 15, 20 and 25–135 min) in order to study the kinetics of the adsorption process. Preliminary experiments showed that this time length was sufficient for adsorption of Pb^{2+} ions onto PMW. The samples were subsequently filtered off and the residual Pb^{2+} ions concentration in the filtrate were analyzed using AAS at a wavelength of 217 nm, or with EDTA titration for high concentrations of lead using xylenol orange (XO) indicator.

The percentage adsorption of Pb^{2+} ions from the solution was calculated from the relationship:

$$\% \text{ Adsorption} = (C_i - C_r)/C_i \times 100 \tag{1}$$