



Arsenate removal from synthetic wastewater by adsorption onto fly ash

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ABSTRACT

In this work, the removal of arsenate from synthetic wastewater by adsorption onto a coal combustion fly ash (CCA) has been experimentally studied. To this aim, a multiple analysis that included the evaluation of metal releases in aqueous solution, the assessment of arsenate adsorption capacity and a possible beneficiation treatment has been performed. The release of different metallic ions (mainly Ca, Al, K, and Si) was observed, including arsenic itself whose total content on CCA resulted to be around 0.05 mg/g. Arsenate adsorption isotherms on raw CCA at constant temperature (20 °C) have been determined in two synthetic arsenic aqueous solutions, a distilled water and a mineral water simulating a groundwater. In both the experimental conditions, CCA shows almost the same arsenate adsorption capacities. In order to increase CCA adsorption capacity and to simultaneously remove the arsenic originally present on CCA, a HCl treatment has been performed; the treated sample showed a higher adsorption capacity, likely related to a surface oxidation. Finally, the effect of equilibrium pH on arsenate adsorption on both raw and treated CCA samples has been investigated. A speciation analysis of arsenate ions revealed that for each investigated pH, the treated CCA sample showed the highest adsorption capacity.

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1. Introduction

The presence of heavy metals in natural water and subsurface aquifers represents a serious threat for human health. Based on a combined evaluation of frequency, toxicity, and potential for human exposure, arsenic is considered at the top of a priority list of hazardous substances [1]. Adverse health effects, mainly deriving from drinking water consumption, can be attributed to arsenic contaminated waters, more often in the oxidized form of arsenate (V) [14,16,17].

The occurrence of arsenic compounds in natural waters derives from anthropogenic sources as well as from natural ones, mainly due to the weathering of arsenic containing rocks. Many countries of the world such as India, Bangladesh, China, USA, Japan, etc. have experienced arsenic groundwater contaminations above the World Health Organization guidelines level, set at 10 µg/l. [14,15].

The typical sorbents for arsenic capture are activated carbons [7,9,16,21], ferric, aluminium and manganese oxides [8,12,16] and iron oxides/chloride impregnated activated carbons [6,13,16]. As a general rule, these sorbents have a high cost and, in most of the cases, they are used only once (the regeneration studies are scarcely available or economically unfeasible). In addition, their final disposal costs are comparable with their price. For these reasons, the use of low-cost materials represents a valid alternative for the challenge of a wider

diffusion of accessible treatment systems. Waste recycling and re-utilization are two energy-efficient processes that have gained popularity due to their environmental friendliness and cost-reductive advantages. An example of such processes is the utilization of waste by-products generated from different human activities for arsenic polluted water depuration, such as agricultural materials [16,26], industrial by-products, mainly in the form of oxides/hydroxides [2,3,16], fly ashes [5,16,21,27] and other alternative materials such as natural zeolites, chitosan, clays, peat, etc. [3,16].

In this work, a fly ash deriving from coal combustion (CCA) has been tested as arsenate sorbent. As a matter of fact, among the fossil sources employed to satisfy the ever-increasing world energy demand, coal is becoming more and more important due to social, political and economical reasons. Consequently, many hundreds of Mton per year of CCA are produced worldwide, and particular attention must be paid to landfill disposal of this residue, a management option that must be limited in accordance with the issuing of recent specific regulations. Therefore, reuse of CCA as sorbent appears a very interesting option. The removal of arsenic by fly ash was found to be greatly dependent on pH [16,27] and several treatments have been proposed by different authors in order to increase CCA adsorption capacity towards heavy metals [18,22,28,29]. Moreover, the CCA chemical composition can exert a great influence on arsenic adsorption, since potential leaching of metallic ions normally present on raw CCA may significantly affect the overall CCA adsorption efficiency [27]. Paradoxically, these issues have been rarely considered in literature surveys on CCA use for water remediation even if they potentially regard almost all the CCA based sorbents.

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In the present work, the suitability of CCA as low-cost sorbent for arsenate adsorption has been experimentally investigated. To this end, a multiple analysis starting from a thorough CCA chemical and physical characterization has been performed. The analysis also included the evaluation of metals release from CCA in different aqueous solutions and the assessment of arsenate adsorption capacity both in distilled water and synthetic groundwater, with the evaluation of concentration and pH effects. Moreover, a possible beneficiation treatment for the extraction of the arsenic originally present on raw CCA and the simultaneous increase of CCA arsenate adsorption capacity has been proposed.

2. Materials and methods

2.1. CCA physical/chemical characterization

The coal combustion fly ash utilized in this work came from an Italian power plant operated by Enel, in which a bituminous coal is used as fuel. Laser granulometric analysis was performed by a Malvern Instrument Master Sizer 2000 granulometer, operating down to a minimum particle size of 0.02 μm . From this characterization, a mean Sauter diameter of 10 μm can be inferred, as reported in Table 1, where the main physical and chemical CCA properties are listed. Porosimetric analysis was carried out by a CE Instruments Sorptomatic 1990 analyzer (N_2 , 77 K) and pore size distribution was obtained by operating a Thermo Finnigan Pascal 240/140 porosimeter, in a pore radius range of 3.7–1000 nm. While the surface area and cumulative pore volume resulted equal to 19 m^2/g and 69.5 mm^3/g , respectively, pore size distribution results are reported in Fig. 1, where it can be observed a broad distribution with a large peak in the radius range of 1.5–4.5 μm . CCA chemical characterization was obtained by performing X-Ray Diffraction (XRD, Philips PW1830) and proximate analysis. In Fig. 2, the XRD analysis has been reported, showing the presence of a significant silico-aluminous fraction, mainly represented by quartz (SiO_2) and mullite ($3\text{Al}_2\text{O}_3 \times 2\text{SiO}_2$) crystalline phases. Proximate analysis showed a moderate fixed carbon content, due to short mean residence times (and, therefore, limited carbon conversion degree) into the combustor. The evaluation of the pH of point zero charge (pH_{PZC}) is based on the Noh and Schwarz [19] method, which shows that the CCA has a marked basic nature.

Furthermore, the CCA sample was subjected to a batch leaching test to determine the ionic metallic releases, for a preliminary evaluation of its suitability for the proposed water treatment. To this end, 1 g of raw ash was put into contact, for 72 h and under stirring, with 0.1 l of distilled water, and thereafter the liquid solution was analyzed by ICP spectrophotometry (employing an Agilent 7500ce apparatus). The results are reported in Table 2, where C represents the mg of element released per gram of CCA.

As it can be observed, significant amounts of different metallic ions are released by CCA, including arsenic itself. By considering the existing limitations of specific elements content for solids disposal in the soils, As, Cr and Mo are the most critical substances to be accounted for. To this end, batch release tests have been carried out using different leaching solutions, represented by HCl, HNO_3 and HF acid solutions, and with the same solid/liquid ratio. The results of leaching tests in various acidic solutions showed good efficiency of extraction from the solid for the

Table 1
Main physical and chemical CCA properties.

Sauter particle diameter (μm)	10
B.E.T. surface area ($\text{m}^2 \text{g}^{-1}$)	19
Cumulative pore volume ($\text{mm}^3 \text{g}^{-1}$)	69.5
Volatile content (% db)	3.68
Fixed carbon (% db)	22.3
Ash content (% db)	74.0
pH_{PZC}	9

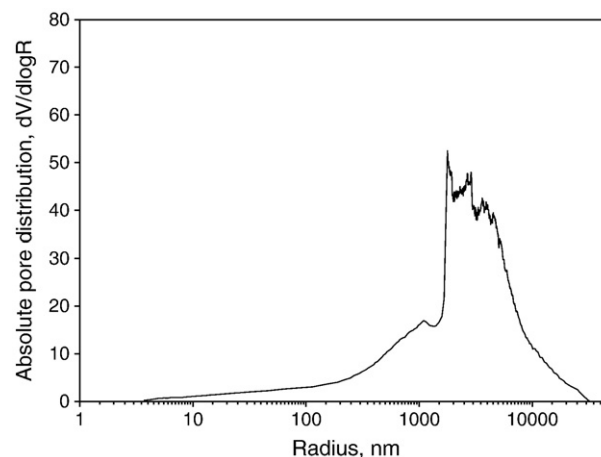


Fig. 1. Absolute distribution of particle pore radius for raw CCA sample.

three target elements; as an example, only results for arsenic releases are reported in Fig. 3.

Arsenic release per unit mass of CCA increases with HCl concentration reaching an asymptotic value in correspondence of a concentration of HCl 1 M. Moreover, at the same acid concentration (0.01 M), HCl allows higher arsenic extraction than HNO_3 . The maximum arsenic release value ($\sim 0.05 \text{ mg/g}$) is independent of the acid used for leaching (HCl, HNO_3 , HF 10 M), leading to the conclusion that a total arsenic content (ω_0) of 0.05 mg/g on CCA can be estimated. This result is perfectly coherent with the observation of Wang et al. [27] who analyzed fly ash deriving from bituminous coal combustion and found an arsenic content of 0.047 mg/g .

2.2. Beneficiation treatments

Starting from raw CCA, several acidic treatments have been tested in order to improve the adsorption properties of the sorbent. Literature indications [18,27–29] and preliminary tests indicated HCl solutions as the most promising acid treatment. Moreover, the presence of arsenic in raw ash and the possibility of its complete removal through an acidic leaching treatment suggested to carry out several treatments at different HCl concentrations, from 0.01 to 10 M. Hence, the treatments were performed by immersing 1 g of raw CCA in 100 ml of HCl solution, for 72 h under continuous stirring; the sample was then filtered and oven dried for 12 h at 105 $^\circ\text{C}$, washed with distilled water and dried again.

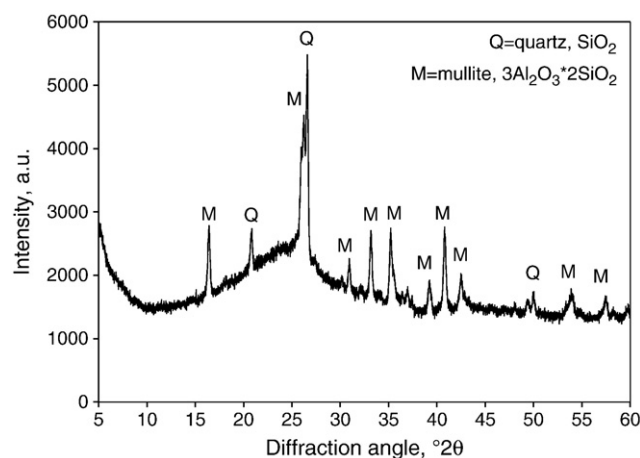


Fig. 2. XRD analysis for raw CCA (Q = quartz, SiO_2 , ASTM#33-1161; M = mullite, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, ASTM#15-776).

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