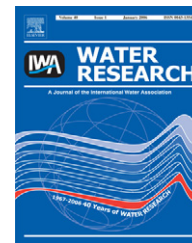


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Experimental and modelling analysis of As(V) ions adsorption on granular activated carbon

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ABSTRACT

In this work the adsorption of pentavalent arsenic on a granular activated carbon (GAC) has been experimentally studied. The effects of arsenic concentration, pH, temperature and salinity on equilibrium adsorption capacity have been investigated. Experimental results show that the adsorption capacity is the highest at neutral pH conditions, low salinity levels and high temperatures. A model for the description of the arsenic adsorption mechanism is reported. This is based on the multicomponent Langmuir adsorption theory applied to the ionic species in solution. The model points out that the adsorption capacity is proportional to the concentration of arsenic anions in solution and decreases by increasing the concentration of competitive ions such as hydroxides and chlorides, allowing a correct interpretation of the pH and salinity effects on the adsorption capacity. Finally, one of the main goals of the proposed model is to preserve the exothermicity of the adsorption phenomena despite the observed trend of experimental results: the increase of adsorption capacity with temperature appears to be related to a higher arsenic dissociation.

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1. Introduction

Arsenic and its compounds are toxic pollutants for the environment and all living organisms (Jain and Ali, 2000; Mondal et al., 2006), that derive from several natural phenomena and antropic activities (Singh et al., 1988; Viraraghavan et al., 1999; Mondal et al., 2006). Arsenic mainly reaches humans through water supplies where it generally occurs as As(V) and As(III), depending on pH and redox conditions (Lorenzen et al., 1995; Mohan and Pittman, 2007). Typical concentration of arsenic in natural waters can rise up to 3 mg/l (Viraraghavan et al., 1999; Mondal et al., 2006). The pentavalent species (arsenates) predominate in aerobic and oxidizing conditions; the trivalent species (arsenites), prevail in anaerobic and moderately reducing conditions (Lorenzen et al., 1995; Viraraghavan et al., 1999).

Due to their high toxicity and to the widespread of their emissions, arsenic and its compounds are strictly controlled by environmental regulations all over the world (Mondal et al., 2006; Mohan and Pittman, 2007). In particular, under directive 2000/60/CE, the European Community has already defined new and more restrictive limits of 1.6 and 1.4 µg/l for superficial waters to be achieved within 2008 and 2015, respectively.

For these reasons, appropriate depuration technologies are required to comply with the environmental regulations. Treatments for arsenic containing wastewaters include coagulation–precipitation, ion exchange, membrane filtration and adsorption (USEPA, 2000; Mondal et al., 2006; Mohan and Pittman, 2007). As a general decisional criterion, an appropriate treatment must reduce pollutant concentration at low costs, good commercial availability and minimum environmental impact. Adsorption processes seem to have these

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Nomenclature			
[X]	concentration of the species X, M or mg/l	K_{Cl^-}	Langmuir model constant for Cl^- species adsorption reaction, M^{-1}
A_i^-	As(V) anions ($i = 1 \dots 3$, i.e. $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-})	K_i	Langmuir model constant for the i -th arsenic species adsorption reaction, M^{-1}
As(III)	trivalent arsenic species	K_{OH}	Langmuir model constant for OH^- species adsorption reaction, M^{-1}
As(V)	pentavalent arsenic species	OH^-	hydroxide ion
C	concentration of a species in solution, M or mg/l	P	lower limit for normality test
H^+	hydrogen ion (proton)	R^2	coefficient of determination
ΔG_{Cl^-}	Standard Gibbs free energy variation for the Cl^- adsorption reaction, kJ/mol	R_{adj}^2	adjusted coefficient of determination
ΔG_i	Standard Gibbs free energy variation for the i -th adsorption reaction, kJ/mol	t	t-statistic test
ΔG°	Standard Gibbs free energy variation for arsenic ions i -th formation reaction, kJ/mol	T	temperature, $^\circ C$ or K
ΔG_{OH^-}	Standard Gibbs free energy variation for the OH^- adsorption reaction, kJ/mol	W	residual weight for regression analysis
F	F-statistic test	ω	adsorption capacity of activated carbon, mg/g
I	salinity, M	ω_i	experimental value of adsorption capacity for the i th sample, mg/g
		ω_{MAX}	maximum adsorption capacity, mg/g
		ω_{ri}	adsorption capacity estimated by the model for the i th sample, mg/g

characteristics, combined with a great versatility and flexibility to guarantee a very rapid adaptation to changes in the pollutant concentrations. The operational cost for adsorbents is mainly related to the price of the sorbent. The unit cost of commercially available activated carbons, which are the most largely used in water and wastewater treatments, is usually higher than 1000 euros/tonne. For this reason, the assessment of the optimal working conditions to maximize the sorbent capture capacity (and consequently minimize their quantities) is absolutely required.

The adsorption mechanism depends on many factors, such as temperature, pH, salinity, arsenic concentration, properties of the sorbent and, finally, the distribution of other chemical species in the aqueous solution.

The analysis of adsorption phenomena in model aqueous solutions has been extensively studied (Mohan and Pittman, 2007). These experiments can be deemed too simplified for the industrial scale-up; the high variability of natural and industrial waters is a severe limitation to the extension of pilot plant results to different working conditions and do not allow any further analysis of the adsorption mechanism apart from the estimation of the overall depuration efficiency. Nevertheless, experiments in model aqueous solutions allow a more reliable analysis of adsorption phenomena and a meaningful method for the comparison of sorbents adsorption capacity.

The typical adsorbents for arsenic capture are activated carbons (Huang and Fu, 1984; Lorenzen et al., 1995; Pattanayak et al., 2000; Chuang et al., 2006), ferric, aluminium and manganese oxides (Singh et al., 1996; Halter and Pfeifer, 2001; Altundogan et al., 2002; Ladeira and Ciminelli, 2004; Deschamps et al., 2005) and iron oxides/chloride impregnated activated carbons (Vaughan and Reed, 2005; Gu et al., 2005; Chen et al., 2007). Several studies have been focused on the effects of arsenic concentration and pH, while variations in temperature and salinity levels have been rarely considered.

The solution pH influences the adsorption of arsenic species as it determines the distribution of arsenic ions

concentration in solution and, first of all, their presence as As(V) or As(III) (Lorenzen et al., 1995). Chuang et al. (2006) noted a reduction in As(V) capture capacity when the initial pH values increase from 5 to 8 by using a oat-hulls-derived activated carbon. Huang and Fu (1984), Lorenzen et al. (1995) and Pattanayak et al. (2000) found that As(V) removal onto activated carbons is favourable for pH between 3 and 5. Similar results for As(V) have been obtained by Singh et al. (1996), Halter and Pfeifer (2001), Altundogan et al. (2002) and Deschamps et al. (2005) for different types of Fe, Al and Mn oxides. Vaughan and Reed (2005) found that adsorption on an iron oxide impregnated activated carbon is highest at low pH values and it decreases when pH increases from 4 to 10, while Gu et al. (2005), working on ferrous chloride impregnated carbons, found that adsorption is maximum between 4 and 9 and it decreases for higher pH values. The adsorption of As(III) is less studied even though these species are more toxic than As(V) (Mondal et al., 2006). Experimental results show that the optimal pH for As (III) removal is around neutrality for both activated carbon (Singh et al., 1988; Pattanayak et al., 2000) and ferrous and aluminium oxides (Altundogan et al., 2002) while Deschamps et al. (2005) found that the maximum uptake of As(III) on Fe–Mn oxides is at pH 3.

The effect of temperature on arsenic adsorption is still a debated subject. Adsorption is an intrinsically exothermic phenomenon, meaning that any increase in temperature has to result in a smaller adsorption capacity. This behaviour is also confirmed for Fe and Al oxides (Singh et al., 1996; Halter and Pfeifer, 2001) while experiments on activated carbon have shown different and indefinite results (Lorenzen et al., 1995; Pattanayak et al., 2000). Although the exothermicity is a specific property of adsorption processes, there is no clear experimental evidence of this phenomenon.

Finally, Lorenzen et al. (1995) showed that the arsenic adsorption capacity onto an activated carbon decreases by increasing NaCl concentration in the aqueous solution, while Gu et al. (2005) found that adsorption on ferrous

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