

Capture of mercury ions by natural and industrial materials

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Abstract

In this paper the technical feasibility of various adsorbents for mercury removal from contaminated waters has been studied. Adsorption isotherms of mercury ions in aqueous solution have been experimentally measured on a granular activated carbon (Aquacarb 207EATM), a char, a pozzolana and a yellow tuff. The experimental evidences show that the mercury capture capacity of yellow tuff and char is of few tenths of milligrams per gram of sorbent while for the pozzolana and the activated carbon this value is of the order of 1 mg/g of sorbent. Moreover, for a mercury concentration as high as 3000 $\mu\text{g/l}$ the pozzolana shows the highest adsorption capacity. This result seems to be quite interesting, especially in consideration of the extremely low cost of this natural sorbent.

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1. Introduction

Mercury is universally recognised as one of the most dangerous inorganic pollutants. Its high toxicity is mainly related to the capacity of its compounds to accumulate in the aquatic food web reaching humans through the food chain. Mercury ingestion typically results in neurological pathologies which may cause fatal injuries and eventually death [1]. The widespread of anthropogenic sources as well as the long atmospheric life of elemental mercury and its compounds lead to a planetary scale problem.

The direct anthropogenic sources of mercury in water bodies are related to numerous industrial applications (e.g. chloro-alkali productions, pharmaceutical and cosmetic preparations, electrical instruments, pulp and paper industries, etc.) and to many products of common use (e.g. thermometers, batteries, medical drugs, etc.) [2–4]. However, the combustion of fossil fuel and solid waste is the main anthropogenic sources of mercury since it is a trace element both in coal (0.1–0.3 mg/kg) and in municipal solid waste (0.5–3 mg/kg) [5,6]. The wet and dry deposition of gaseous mercury and particulate matter leads to the contamina-

tion of soils and superficial water bodies. Pacyna and Munch [7] and Seigneur et al. [8] reported that the global emissions of mercury from major anthropogenic sources in nineties have reached the level of 2000 tonnes/year, while natural emissions account for about 2500 tonnes/year. Data on typical untreated wastewaters from different civil and industrial activities show that the mercury concentration is usually smaller than 2000 $\mu\text{g/l}$ [2,3].

Because of its high toxicity, the European Union considers mercury as a priority and hazardous pollutant and defines a maximum permissible concentration of total mercury as low as 1 $\mu\text{g/l}$ for drinking water and 5 $\mu\text{g/l}$ for wastewater discharge. Moreover, under Directive 2000/60/CE the European Union stated the cessation or phasing out of discharges, emissions and losses within 2020. Before this deadline the complete remediation of polluted water bodies has to be realized.

Hence, it is necessary to remove mercury from wastewaters before they are discharged into the environment and to provide a requalification of polluted natural water bodies. Commonly adopted methods to remove mercury from industrial wastewaters include sulphide precipitation, membrane filtration, iron and alum coagulation, ion exchange and activated carbon adsorption.

Adsorption of mercury by solid materials seems to be a suitable choice for final purification of wastewaters thanks to the appreciable removal efficiency and the intrinsic flexibility of this unit operation [9]. The typical sorbents for mercury ions removal are activated carbons and zeolites [10–12] but in the

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Nomenclature

c	total mercury concentration in solution ($\mu\text{g/l}$)
c°	total mercury concentration in the initial solution ($\mu\text{g/l}$)
V	volume of the aqueous solution (l)
m	mass of the sorbent (g)
K	Langmuir parameter (l/g)
H	linear isotherm parameter (l/g)
<i>Greek letter</i>	
ω	amount of mercury captured per gram of sorbent (g/g)
ω_{max}	Langmuir parameter (g)
ε	experimental error

past few years several natural and refuse derived materials have been tested as alternative low-cost sorbents [13–17]. The good adsorption and ionic exchange capacities shown by several minerals and soils [18–22] suggest the possible use of these materials for the protection or the remediation of natural water bodies. Recently, the realization of permeable “vertical layers” of adsorbing material for groundwater remediation and for the control of the transport of pollutants between superficial and underground waters without affecting the water flow regimes has been suggested [23,24]. In these cases, the unit cost, the availability, the environmental hazard and the possible reuse of a given sorbent are fundamental parameters as well as its capture capacity.

The present work reports some experimental studies on capture of mercury ions by several sorbents, namely a non-impregnated activated carbon (Aquacarb 207EATM by Sutcliffe Carbon) and three natural materials: a char derived from a South African coal, a pozzolana and a tuff from a volcanic zone near Naples, in the South of Italy. These four materials have been chosen for their potential capture capacity as well as for their cost effectiveness and environmental safety which make them suitable sorbents for both natural and industrial water treatments.

2. Experimental study

2.1. Materials

The four materials have been chosen in order to make a comparison between a well tested industrial sorbent (the activated carbon Aquacarb 207EATM), a low-cost precursor of activated carbons (the char of South African coal) and two natural materials (the pozzolana and the yellow tuff).

The Aquacarb 207EATM is a commercially available non-impregnated granular activated carbon produced by Sutcliffe Carbon starting from a bituminous coal. This material has a narrow particle diameter distribution between 0.6 and 1.5 mm, with an average diameter of 1.2 mm. The char of South African coal presents a higher average particle diameter, around 2.25 mm, and a wider particle diameter distribution, with more than 90% of the material almost uniformly distributed between 1.3 and

Table 1

Characteristics of Aquacarb 207EATM and char of South African coal

	Aquacarb 207EA	Char of S.A. coal
Origin	Bituminous coals	South African Coal
Activation method	Steam activation	–
Specific area, BET (m^2/g)	950	
Density (g/cm^3)	480–520	641
Effective density (g/cm^3)	950	1850
Average diameter (mm)	1.20	2.27
Average pore diameter (\AA)	4670	
Porosity (%)	16.8	12
Humidity (% w/w)	5	5
Ultimate analysis (% w/w) on dry basis		
Carbon	87.00	77.00
Hydrogen	0.17	1.70
Oxygen	2.00	2.15
Nitrogen	1.10	1.80
Sulphur	n.d.	0.75
Ash	9.58	16.00
pH _{PZC}	8.00	8.85
Boehm's titration analysis (acid/base adsorption)		
HCl (mmol/g)	2.44	0.97
NaOH (mmol/g)	0.40	0.34

3.2 mm. The densities of these materials are between 500 and 650 kg/m^3 . Both the carbons present a slightly basic character as well stressed by the value of their pH_{PZC} (evaluated by mass titration analysis [25]) and by the concentration of surface functional groups measured following Boehm's acid/base titration method [26]. The main chemical and physical characteristics of the two materials are listed in Table 1.

Pozzolana is a natural volcanic slag, while yellow tuff is a relatively soft porous rock usually formed by compaction and cementation of volcanic ash or dust. The origin of yellow tuff may be also related to the diagenesis of the pozzolana even if this is a much debated issue. In particular, the two materials used in this work are extracted from two quarries in the Phlegrean fields, a volcanic area at the north of Naples (Italy). Detailed analyses of the chemical and mineralogical characteristics of these two materials are reported in Scherillo and Scherillo [27] and de'Gennaro et al. [28]. The chemical compositions and the physical properties of tuff and pozzolana used in this work are resumed in Table 2. In both cases particle diameters are widely distributed from 5 μm to 4 mm. The two materials have been previously washed and sieved and only the granulometric fraction above 50 μm has been used in all experimental runs.

Mercury aqueous solutions have been realized by dissolution of mercuric chloride (HgCl_2 , reagent grade) in distilled water. This salt has been chosen because chloride ions are almost ubiquitous in natural and industrial waters and, among the inorganic mercury complexes, mercuric chloride is one of the most abundant and soluble in aqueous solutions. Moreover, according with the theory of ions adsorption in aqueous solutions [29], the dominant presence of non-ionic species as HgCl_2 leads to a decrease of sorption capacity as shown, for example, by Manohar et al. [17]. Thus, the use of mercuric chloride gives a more realistic and conservative estimation of the mercury removal efficiency of the sorbents.

Table 2
Characteristics of yellow tuff and pozzolana

	Yellow tuff from Monte S. Severino (Quarry De Crescenzo)	Pozzolana from the basin of Pianura
Bulk density (kg/m ³)	901	896
True density (kg/m ³)	1800	2460
Diameter (μm)	50–4000	50–4000
Chemical analysis (% w/w) (Scherillo and Scherillo [27])		
SiO ₂	52.59	57.05
TiO ₂	0.40	0.40
ZrO ₂	0.05	0.06
Al ₂ O ₃	16.23	18.32
Fe ₂ O ₃	2.82	1.49
FeO	0.56	2.56
MnO	0.10	0.11
MgO	1.24	1.36
CaO	2.83	3.70
BaO	0.21	0.25
K ₂ O	6.59	7.77
Na ₂ O	3.95	4.50
Cl ₂	0.04	0.16
SO ₃	0.07	0.20
P ₂ O ₅	0.11	0.23
H ₂ O ⁻	4.66	0.20
H ₂ O ⁺	7.17	2.30
CO ₂	0.22	–

2.2. Procedures

The experimental runs have been carried out in batch mode at the constant temperature of 20 °C in a PID controlled thermostatic oven. Each sample consists of 200 ml aqueous solution containing a given mercury concentration ($c^{\circ} = 1000\text{--}5000 \mu\text{g/l}$), obtained by dissolution of a mercury chloride in distilled water. The solution is placed in contact with a mass of sorbent ranging between 0.5 and 10 g. The solution pH has been measured but not adjusted. Preliminary experimental tests on mercury capture rate show that a contact time of 48 h is sufficient to reach the equilibrium conditions for each sorbent. These experiments have been carried out both with and without mechanical stirring. No significative difference has been observed in either case, showing that the most important resistance in the mass transfer phenomena is the diffusion within the porous structure of the particles. In order to evaluate the equilibrium conditions, both the mercury concentration in the solution, c , and on the solid surface, ω , have been measured. The samples have been analysed by means of cold vapour absorption (CVAA), using NaBH₄ as reducing agent. The quantity of mercury adsorbed per gram of sorbent, ω , has been measured by leaching the adsorbing materials with *aqua regia* (HNO₃ 25%, HCl 75%) and then analysing the solution by means of CVAA. All the analyses have been realized with a Varian SpectrAA-220 atomic absorption spectrophotometer equipped with a VGA-77 hydride generator system for CVAA analysis.

Starting from the measurement of the initial and equilibrium concentrations in solution and on the solid surface, the following material balance on mercury can be written:

$$\omega M - V(c^{\circ} - c) = \varepsilon \quad (1)$$

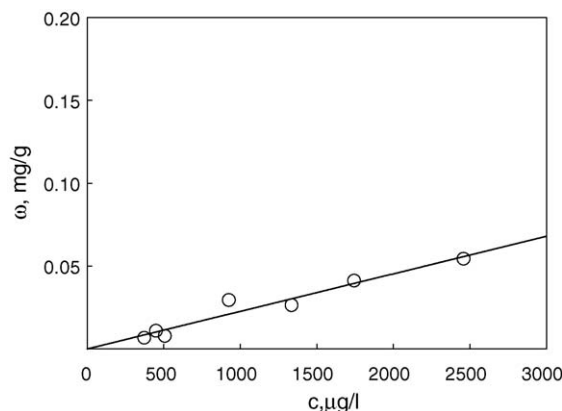


Fig. 1. Capture of mercury by char of South African coal.

where M is the mass of the sorbent, V the sample volume and ε is the experimental error. The maximum allowed error in the material balance has been fixed at 5%. Each experimental run has been repeated three times and the average values of c and ω have been considered. Moreover, the initial and the equilibrium pH of the aqueous solution have been measured by means of an Orion EA 920 digital pH-meter.

3. Results and discussion

The experimental work has been focused on the thermodynamic aspects of mercury capture by the four investigated materials. In particular, the experimental data are shown in Figs. 1–4 in terms of mercury adsorption capacity, ω , in function of the equilibrium concentration of dissolved mercury, c . The equilibrium pH for all the experiments ranges from 6.5 to 7.3 with the lowest pH value observed for higher mercury content in solution.

In Fig. 1 the capture of mercury by char of South African coal is reported. This figure shows an almost linear dependence of ω on mercury concentration. A capture capacity of about 0.1 mg/g at 3000 $\mu\text{g/l}$ can be observed.

Fig. 2 resumes the experimental data concerning the yellow tuff. In this case an almost linear dependence of ω on mercury concentration is shown. The capture capacity results almost doubled as compared to that of char (0.18 mg/g at 3000 $\mu\text{g/l}$).

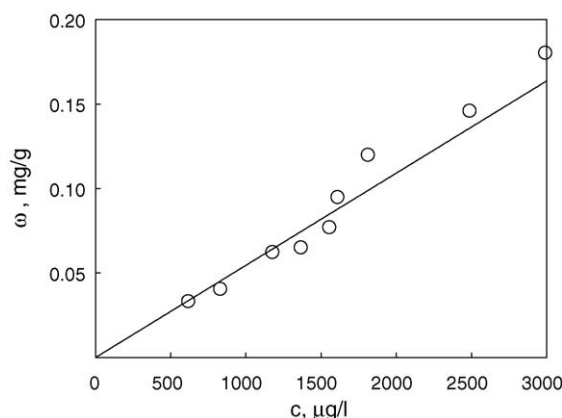


Fig. 2. Capture of mercury by yellow tuff of Phlegrean fields.

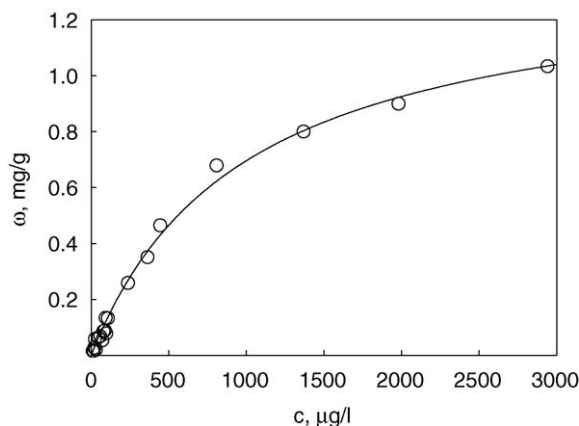


Fig. 3. Capture of mercury by pozzolana of Phlegrean fields.

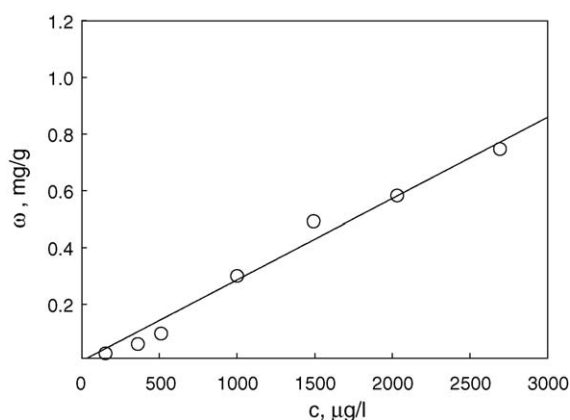


Fig. 4. Capture of mercury by Aquacarb 207EA™ granular activated carbon.

The experimental results obtained for the pozzolana are reported in Fig. 3. The figure shows that the adsorption capacity reaches an asymptotic value around 0.8 mg/g for mercury concentration as high as 1000 $\mu\text{g/l}$. For a mercury concentration up to 3000 $\mu\text{g/l}$ the adsorption capacity results almost one order of magnitude higher than the char.

Finally, Fig. 4 shows the experimental results for adsorption of mercury on the activated carbon. In this case the capture capacity follows a linear trend with mercury concentration with a value of 0.8 mg/g for a mercury concentration of 3000 $\mu\text{g/l}$, quite close to that shown by pozzolana.

Experimental results show that, for all the investigated solids, capture of mercury and mercury concentration in aqueous solution result in a linear function up to a value of 1000 $\mu\text{g/l}$ with the highest slope shown by the pozzolana. At higher concentrations, only this last material tends to reach an asymptotic value. More-

over, it is interesting to observe that, despite the zeolitic nature of yellow tuff, whose cationic exchange capacity is well known [30–32], the adsorption capacity of pozzolana, an incoherent granular material which is a mineralogical precursor of the yellow tuff without any zeolitic structure, is clearly higher. This result may be explained by considering the chemical structure of the two materials (Table 2). Pozzolana has a higher concentration of sulphur and phosphate which form mercury complexes and are eventually used to increase the mercury adsorption capacity of sorbents [33–36]. Similarly the content of iron oxides, which present a good mercury adsorption capacity [37], is higher for the pozzolana rather than for the yellow tuff.

The experimental data may be interpreted by the classical adsorption models [18]. In particular, experimental data for char, activated carbon and yellow tuff can be described by a linear isotherm, while adsorption on pozzolana shows the typical trend of a Langmuir isotherm. The regression analyses have been realized by using a simple linear regression method to describe the experimental data on char, yellow tuff and activated carbon. For the case of pozzolana, where the Langmuir model has been considered, the linear regression of data is made between $1/c$ and $1/\omega$ according to the following expression of the Langmuir equation:

$$\frac{1}{\omega} = \frac{1}{K\omega_{\max}} \frac{1}{c} + \frac{1}{\omega_{\max}} \quad (2)$$

In this case, the slope of the regression line corresponds to $1/K\omega_{\max}$ and the intercept is equal to $1/\omega_{\max}$ where ω_{\max} and K are the Langmuir parameters.

The parameters of adsorption isotherms calculated by regression analysis of experimental data for each material are collected in Table 3 which also lists the value of the correlation factor, R^2 , and of the standard estimation error for each parameter. In this table, H is the proportionality constant for the linear adsorption model. As it is well known, the linear model is an approximation of the Langmuir model in conditions far from saturation and the value of the constant H may be considered as the product $\omega_{\max}K$.

From an operative point of view it is necessary to highlight that the applicability of a given process for wastewater treatment and for natural water bodies remediation cannot ignore the economical aspects of this process. In the case of adsorption, these are mainly related to the unit cost of the sorbent. Recent estimations show that the unit cost per ton of a granular non-impregnated activated carbon is around €2000, while that of coal in the United States in 2002 was around US\$ 17 [38,39]. The cost of yellow tuff and pozzolana is mainly related to their extraction from superficial quarries and it is around €4 tonne^{-1} of tuff and €1.5 tonne^{-1} of pozzolana, one order of magnitude lower than that of coal.

Table 3
Adsorption isotherms models and parameters

	Adsorption isotherm	Expression of the adsorption isotherm	Value of the parameters	R^2
Char of S.A. coal	Linear	$\omega = Hc$	$H = 0.0185 \pm 0.008 \text{ l/g}$	0.95
Yellow tuff	Linear	$\omega = Hc$	$H = 0.058 \pm 0.02 \text{ l/g}$	0.96
Pozzolana	Langmuir	$\omega = \omega_{\max} \frac{Kc}{1+Kc}$	$\omega_{\max} = 0.00138 \pm 6 \times 10^{-5}$, $K = 1016 \pm 78.9 \text{ l/g}$	0.99
Aquacarb 207EA™	Linear	$\omega = Hc$	$H = 0.314 \pm 0.10 \text{ l/g}$	0.98

In this sense, the performance of pozzolana in mercury capture seems to be of particular interest since it is very cost-effective especially considering that the typical mercury concentration in polluted natural water bodies is below 500 $\mu\text{g/l}$ [2,3].

4. Conclusions

In this paper experimental data on mercury capture by a non-impregnated granular activated carbon, a char of South African coal, a pozzolana and a yellow tuff from Phlegrean Fields are presented. Due to their chemical composition all these materials present a negligible environmental hazard. Moreover, they are relatively abundant in the environment. Experimental results point out that for a mercury concentration lower than 3000 $\mu\text{g/l}$ the capture capacity is maximum for the pozzolana and minimum for the char. The experimental results may be interpreted by a linear isotherm for char, yellow tuff and activated carbon, while for pozzolana the Langmuir model has been considered. The appreciable mercury capture capacity, the cost effectiveness and the negligible environmental hazard of pozzolana makes it a suitable choice for both natural water bodies' remediation and wastewater treatments.

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