Use of fly ash and fly ash agglomerates for As(III) adsorption from aqueous solution

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The objective of the present study is to assess the efficiency of fly ash and fly ash agglomerates to remove arsenic(III) from aqueous solution. The maximum static uptakes were achieved to be 13.5 and 5.7 mg$_{As(III)/adsorbent}$ for non-agglomerated material and agglomerated one, respectively. Isotherm studies showed good fit with the Langmuir (fly ash) and the Freundlich (fly ash agglomerates) isotherm models. Kinetic studies indicated that the sorption of arsenic on fly ash and its agglomerates follows the pseudo-second-order (PSO) chemisorption model ($R^2 = 0.999$). Thermodynamic parameters revealed an endothermic nature of As(III) adsorption on such adsorbents. The adsorption results confirmed that fly ash and its agglomerates can be used for As(III) removal from aqueous solutions. Fly ash can adsorb more arsenic(III) than agglomerates, which are easier to use, because this material is less dusty and easier to separate from solution.

Keywords: arsenic, fly ash, fly ash agglomerates, adsorbents, adsorption.

INTRODUCTION

Arsenic contamination of land and natural waters has recently become a problem on a global scale. Arsenic is one of the most toxic trace elements. Present in drinking water, has a significant impact on human health. As a carcinogenic factor, it has an effect on the skin, liver, lungs and nervous system[1, 2, 3]. Arsenic contamination may result from a highly developed industry (mining and metallurgy) and geothermal processes. Origin of natural arsenic also increases the amount of this element in water. Over 300 minerals contain arsenic in their structures, for example: arsenopyrite and loellingite. Arsenic occurs mainly in the form of oxides of As(III) as arsenite ($H_2AsO_3^-$) and As(V) as arsenate ($H_2AsO_4^-$)[1, 2]. Arsenic(III) is the most toxic and mobile form in the environment. Arsenic compounds may migrate through the soil layer to groundwater, which constitutes a potential threat to human life and health.

Removal of As(III) ions from aqueous solution can be achieved by various techniques, e.g. oxidation/precipitation, coagulation/coprecipitation, ion exchange, membrane filtration and adsorption[1, 2, 4, 5]. Adsorption is a very popular method, because the process is efficient and economically viable[6–12]. For the adsorption process such sorbents as: macrofungus (*Inonotus hispidus*) biomass[1], iron oxide-coated cement (IOCC)[4], biogenic schwertmannite[13], synthetic siderite[14], nanosized iron oxide–coated perlite[15] or fly ash[16–20] can be used. The literature provides numerous data of maximum uptake for As(III) adsorption onto various adsorbents, such as soot (29.9 mg$_{As(III)}$/g$_{adsorbent}$)[26], biogenic schwertmannite (113.9 mg$_{As(III)}$/g$_{adsorbent}$)[13], iron oxide-coated cement (IOCC) (0.69 mg$_{As(III)}$/g$_{adsorbent}$)[4], synthetic siderite (9.98 mg$_{As(III)}$/g$_{adsorbent}$)[14], nanosized iron oxide-coated perlite (0.39 mg$_{As(III)}$/g$_{adsorbent}$)[15] and fungal biomass (51.9 mg$_{As(III)}$/g$_{adsorbent}$)[1], as well as commercial product ArsenX^{np} resin (38 mg$_{As(III)}$/g$_{adsorbent}$)[21]. Maximum adsorption of As(V) onto fly ash reported by Wang was 0.8 mg$_{As(III)}$/g$_{adsorbent}$[18].

Fly ash is a waste material generated from power plants. Approximate 500 million tons of fly ash are produced per year throughout the world[18, 20]. Fly ash has a pozzolanic property and therefore it is a valuable and desirable material. When added to water this material is strongly alkaline, with pH of 10–13. The chemical composition of fly ash depends on the type of material burnt as well as on the method of combustion. Consisting of mostly silica and silicates, fly ash has been used effectively in many areas, such as ALC manufacturing, building materials, soil amendment and fillers[22]. Fly ash can be used also as a sorbent of heavy metals (Tl, Cd, As and Cu)[22, 23] and organic materials[22] from water and waste water.

In industry, fly ash is converted into a more convenient compressed form of granules, pellets and briquettes. This facilitates the operation of the adsorption process because this sorbent is less dusty and easier to separate from the solution. Granulated fly ash has been used as a sorbent for heavy metals, such as arsenic[16, 19], cadmium[23, 24] and copper[24] from aqueous solutions.

Adsorption of arsenic on fly ash was found to conform to Freundlich isotherm[19, 22] and adsorption efficiency was comparable to that of activated carbon. Some authors reported better fit with the Freundlich than with the Langmuir model[23, 25]. Determination of the kinetics parameters and explanation of the mechanism in heterogeneous systems is often a complex procedure[26]. Most often the pseudo-second-order (PSO) kinetic model is used. This model has been tested very often for the simulation of the experimental results of the adsorption of heavy metals[23, 25]. However, some authors found other well described models of adsorption of arsenic(III), such as pseudo-first order (PFO) model[1, 14–16] or Elovich equation[16].

The purpose of this study was to investigate the possibility of the utilization of coal fly ash as a low cost and effective adsorbent for arsenic removal.

MATERIALS AND METHODS

The fly ash coming from brown coal and biomass burning, has been brought from power plant "Zgierz"

(Poland). The X-ray diffraction of fly ash showed the presence of the following minerals: anhydrite (44.2%), quartz (29.5%), calcium oxide (16.9%), carbon (5.5%), calcite (2.4%) and hematite (1.5%). Particle analysis was undertaken using a Mastersizer 2000 laser diffractometer (Malvern), equipped with HydroMu dispersion unit (Malvern). The particle size analysis showed the median diameter (d_{50}) of about 105.5 μm, while d_{10} and d_{90} were 17.8 μm and 212.6 μm, respectively. The density of fly ash was determined using a pycnometer and was found to be 2.70 g/cm³. The surface area was measured by BET method for helium/nitrogen mixture using FlowSorbII apparatus (Micromeritics) and was 17.3 m²/g for fly ash powder.

Tumble agglomeration experiments have been conducted in the 6-liter and 185 mm-diameter plastic container placed horizontally and rotated by motor at 60 r.p.m. Water (110 mL) was used as the binder liquid. In each experiment, 200 g of fly ash samples were taken and the agglomeration process was carried out for 1 hour. After this time the agglomerates were formed. The wet product was cured in a curing chamber at room temperature for one week to fulfill the hydration of cementitious components. After drying, the agglomerates were divided into the following fractions: <0.125 mm, 0.125–0.5 mm, 0.5–1.0 mm, 1.0–1.6 mm, 1.6–2.5 mm, 2.5–5.0 mm, 5.0–6.3 mm and >6.3 mm. The surface area increased to 25.9 m²/g for fly ash agglomerates as a result of C--S-H phase creation. The experiments of arsenic (III) adsorption from aqueous solution were carried out on the fly ash agglomerates of the size between 2.5 mm and 5.0 mm.

The study of the mechanical strength of agglomerates was conducted on the universal testing machine (Material Test System 810). Four different fractions of agglomerates were analyzed: 5 mm, 6–8 mm, 12–13 mm and 18–19 mm. The study showed that the larger agglomerates could withstand compression to 45.9 N, while the smaller ones crumbled already at a force of 16–20 N.

Adsorption experiments of the effect of pH were studied with initial arsenic(III) concentration of 500 mg/L and adsorbent dose 0.25 g of or 2.5 g per 10 mL solution of fly ash and fly ash agglomerates (2.5–5.0 mm), respectively. The contact time was 24 hours and was executed at room temperature (25ºC). The initial solution pH was adjusted by using 0.1 M HCl and 0.1 M NaOH.

The static uptakes of arsenic(III) by fly ash and fly ash agglomerates were determined for the following adsorbent/arsenic ratios: 0.05, 0.1, 0.125, 0.25, 0.375, 0.5, 0.625 and 0.75 g/mL of 1000 mg/L arsenic solution and 0.05, 0.075, 0.1, 0.25, 0.5 and 0.65 g/mL of 1000 mg/L arsenic solution for fly ash and fly ash agglomerates, respectively.

Adsorption isotherms of arsenic(III) were determined as follows: the fly ash powder (0.25 g, 0.5 g and 1 g) or fly ash agglomerates of the size 2.5–5.0 mm (1 g, 2.5 g and 5 g) were added to 10 mL of acidic arsenic(III) solution (10–1000 mg/L). The resulting suspensions were shaken at 25ºC for 24 hours. The thermodynamic studies were done with an initial arsenic(III) concentration of 1000 mg/L at 25, 35, 45 and 55ºC. The measurements were conducted for 0.25 g or 2.5 g/10 mL of arsenic(III)

solution of fly ash and fly ash agglomerates (2.5–5.0 mm), respectively. The equilibrium time for thermodynamic studies was 24 hours.

After 24 hours, the equilibrium solute concentration of arsenic(III) (c_{eq}) was analyzed spectrophotometrically (UV-Vis Helios Gamma, ThermoFisher) by means of the molybdenum blue method, according to the standard procedure. The value of equilibrium solute concentration (c_{eq}) enables to determine the amount of arsenic(III) adsorbed at equilibrium (q_{eq}) [$mg_{As(III)}/g_{sorbent}$].

The kinetic experiments of arsenic(III) adsorption were carried out in the following procedure: fly ash powder (1.75 g, 2.5 g, 5 g, 7.5 g, and 10 g) or fly ash agglomerates (10 g, 17.5 g, 25 g, 30 g and 35 g) were added to 50 mL of arsenic stock solution. Arsenic(III) concentration was monitored for 24 hours, starting at 5th minute. The measured value of the concentration of arsenic(III) at a given time (c_t) allowed to determine the amount of arsenic(III) adsorbed at that time (q_t) [$mg_{As(III)}/g_{sorbent}$].

RESULTS AND DISCUSSIONS

Adsorption study

Effect of pH

According to the literature data, the optimum pH for the arsenic(III) adsorption depends on the selection of sorbent type, e.g. for macrofungus (*Inonotus hispidus*) biomass is 6.0[1], iron oxide-coated cement (IOCC) 6.7[4], hydrous titanium dioxide 4.0[5], biogenic schwertmannite 7–10[13], ArsenX[np] resin 5.0–8.5 and carbon-based adsorbents < 7[21].

The effect of pH on the adsorption of arsenic(III) onto fly ash and fly ash agglomerates (2.5–5.0 mm) was studied at pH range 5–12 and the results are presented in Figure 1 and Figure 2. The uptake of arsenic(III) at various initial pH levels (5–12) for an arsenic concentration of 500 mg/L was studied to determine the optimum pH. The arsenic adsorption is not much affected over the investigated pH range. In all investigated ranges of pH there are no significant differences in the amount of adsorbed arsenic(III) onto fly ash powder and agglomerates. As can be seen from Figure 1. and Figure 2., the
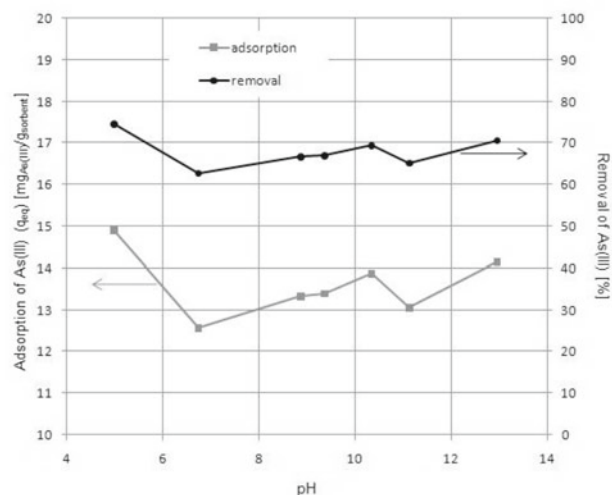

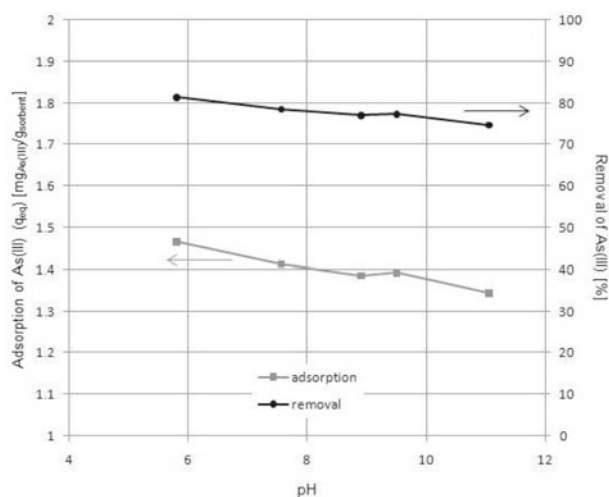**Figure 1.** Effect of pH on As(III) adsorption onto fly ash (0.25 g/10 mL of 500 mg/L As(III) solution; T = 25ºC)

Figure 2. Effect of pH on As(III) adsorption onto fly ash agglomerates (2.5–5.0 mm; 2.5 g/10 mL of 500 mg/L As(III) solution; T = 25°C)

arsenic adsorption varied from 12.6 to 14.1 and from 1.35 to 1.47 $mg_{As(III)}/g_{sorbent}$ for fly ash powder and agglomerates, respectively. Therefore the sorption experiments have been performed at natural pH, i.e. imposed by fly ash (10.40–11.50) or fly ash agglomerates (8.50–10.75).

Effect of adsorbent dose

The maximum static uptakes of arsenic by fly ash powder and fly ash agglomerates were achieved for adsorbent-to-arsenic ratios of 0.025 and 0.030 g/mL and were 13.5 (Fig. 3) and 5.7 $mg_{As(III)}/g_{adsorbent}$ (Fig. 4), respectively. The previous research of the authors[11] pointed out that the maximum uptake by brown coal fly ash powder was 74.4 $mg_{As(III)}/g_{adsorbent}$ (for 0.025 g/mL adsorbent/arsenic ratio), while by the agglomerates was 5.4 $mg_{As(III)}/g_{sorbent}$ (for 0.1 g/mL). Comparing fly ash adsorbents, those values differ much, depending on fly ash origin as well as the adsorbent form. Even though the agglomerated fly ash exhibits bigger specific surface area, the non-agglomerated adsorbent takes up much more arsenic per gram.
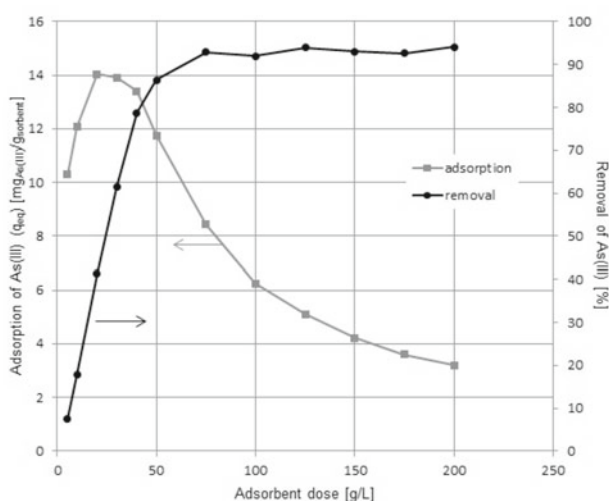

Figure 3. The sorption capacity of fly ash and As(III) removal from standard solution
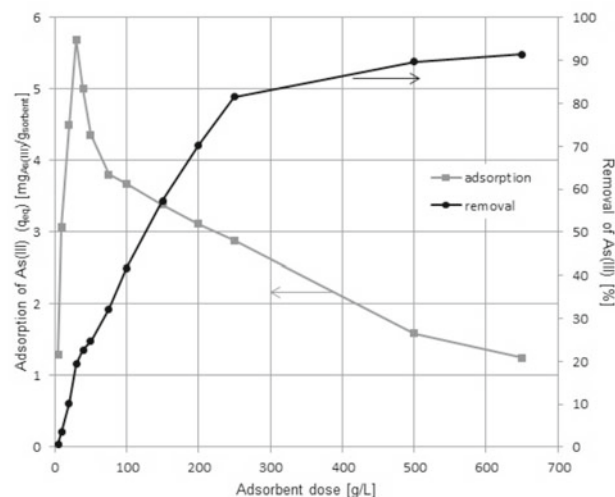

Figure 4. The sorption capacity of fly ash agglomerates and As(III) removal from standard solution

Adsorption isotherms

The equilibrium adsorption isotherms determination is of fundamental importance. The adsorption experimental results were worked up using two isotherm models: Langmuir and Freundlich. The Langmuir model assumes monolayer sorption onto a homogeneous surface with a finite number of identical sites. The Freundlich is empirical in nature[4].

The Freundlich model[4, 6, 10–12, 19] is described by the following equation.

$$q_{eq} = k_f \cdot c_{eq}^{1/n} \tag{1}$$

and a linear form:

$$\log q_{eq} = \log k_f + \frac{1}{n} \log c_{eq} \tag{2}$$

where: q_{eq} is the amount of arsenic(III) adsorbed at equilibrium [$mg_{As(III)}/g_{adsorbent}$], c_{eq} the equilibrium solute concentration [mg/L], k_f [mg/g] and n [-] the Freundlich constants representing the adsorption capacity and the adsorption intensity, respectively.

The linear form of Langmuir model[4, 7, 9] is as follows:

$$\frac{1}{q_{eq}} = \frac{1}{b \cdot Q_0} + \frac{c_{eq}}{Q_0} \tag{3}$$

where: Q_0 and b are the Langmuir constants related to saturated monolayer adsorption capacity [mg/g] and the binding energy of the sorption system [L/mg], respectively, c_{eq} is the equilibrium solute concentration [mg/L] and q_{eq} is the amount of arsenic(III) adsorbed at equilibrium [mg/g].

These two adsorption isotherm models were tested to fit the experimental data of arsenic adsorption onto fly ash adsorbent. The best fit of the experimental data has been achieved using the Langmuir isotherm model for fly ash powder and the Freundlich isotherm model for fly ash agglomerates (2.5–5.0 mm). The Langmuir isotherm assumes monolayer coverage of adsorbate over a homogeneous adsorbent surface. Values of Q_0 and b were calculated from the slope and intercept of the linear plot of c_{eq}/q_{eq} versus c_{eq}. The Freundlich isotherm encompassing the surface heterogeneity, the exponential distribution of active sites and their energies[1, 3]. The linear dependence of log q_{eq} and log c_{eq} gives a slope

that is the value of $1/n$ and the y-intercept is $\log k_f$. Experimental data and model curves are shown in Figure 5 and Figure 6, In these studies and the previous studies[16, 19], the equilibrium adsorption data arsenic(III) for agglomerated fly ash did not obey the Langmuir equation, perhaps due to the heterogeneity of the sorbent surface. The designated parameters of adsorption isotherms are higher for fly ash powder than for fly ash agglomerates. These values are different depending on the amount of the sorbent used. The Langmuir and Freundlich isotherm parameters determined for various amounts of adsorbent are shown in Table 1. Other authors describe the equilibrium adsorption of arsenic(III) mostly using the Freundlich and Langmuir equations[1, 4, 5, 13–15, 17].
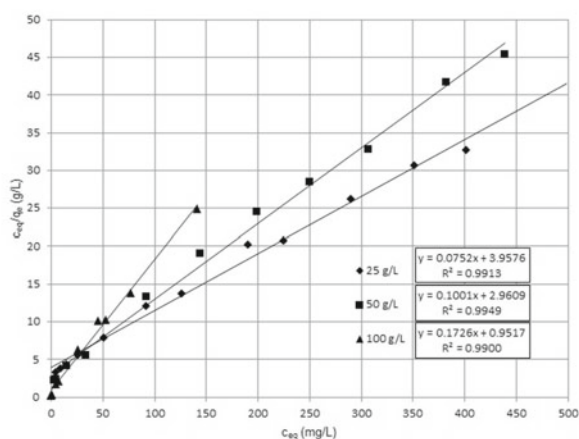

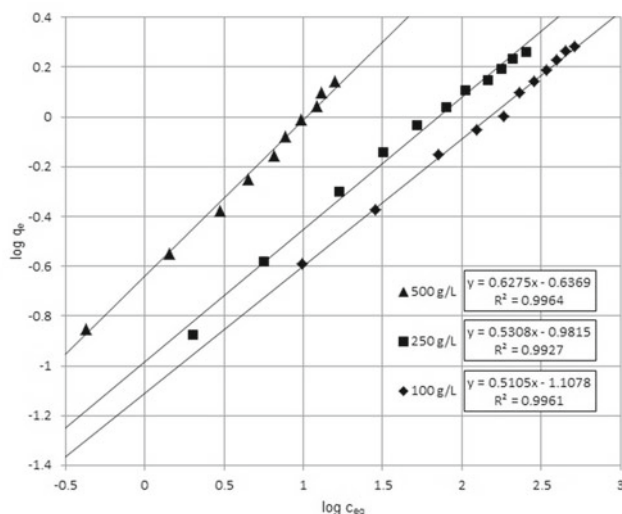**Figure 5.** Adsorption isotherms (Langmuir model) of As(III) onto fly ash (pH~11; T = 25°C)

Figure 6. Adsorption isotherms (Freundlich model) of As(III) onto fly ash agglomerates (pH~10; T = 25°C)

Thermodynamic parameters

The amount of arsenic(III) adsorbed by fly ash and fly ash agglomerates (2.5–5.0 mm) were measured at 25, 35, 45 and 55°C. The thermodynamic equilibrium constant K_0 for the adsorption process were obtained by plotting $\ln(c_{ads}/c_{eq})$ versus c_{ads} (is the equilibrium adsorbate concentration on the adsorbent [mg/L] and is calculated as $(c_0 - c_{eq})$) and extrapolating to zero using a graphical method[1, 4, 14, 22, 25, 27]. Regression straight lines were fitted through the data points by the least-squares method and from their intersections with the vertical axis, the value of $\ln(K_0)$ was estimated at four different temperatures. The standard differential Gibbs energy of adsorption (ΔG^0) can be written in terms of the equilibrium constant (K_0) as:

$$\ln K_0 = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \qquad (4)$$

where R is the universal gas constant [J/mol · K] and T the temperature [K]. The standard differential heat of adsorption (ΔH^0) and entropy (DS^0) can be determined from the equation[4, 21, 27], as follows:

$$\ln K_0 = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \qquad (5)$$

The values of enthalpy change (ΔH^0) and entropy change (ΔS^0) can be calculated from the slope (according to Eq. 5) and the intercept of the plot of $\ln K_0$ versus $1/T$ (Fig. 7). The negative values in the free energy change ΔG^0, as presented in Table 2, indicate the spontaneous nature of the adsorption process. The positive ΔH^0 value
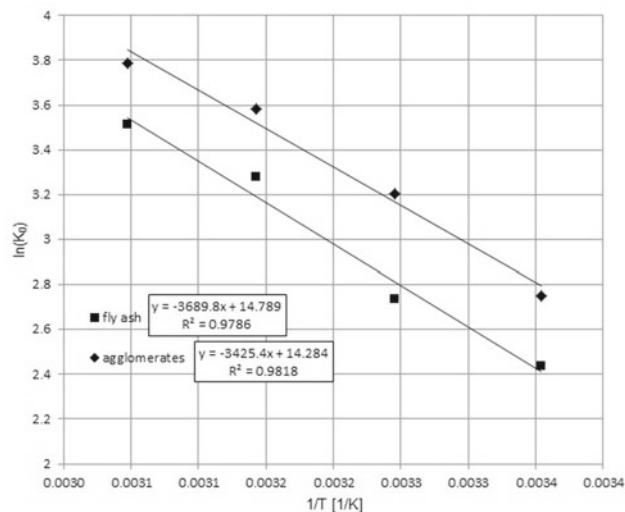

Figure 7. Plot of $\ln(K_0)$ vs. $1/T$ for estimation of thermodynamic parameters for As(III) adsorption onto fly ash and fly ash agglomerates

Table 1. Isotherm parameters and R^2 values for arsenic(III) adsorption onto fly ash and fly ash agglomerates (2.5–5.0 mm)

	Fly ash			Fly ash agglomerates		
	25 g/L	50 g/L	100 g/L	100 g/L	250 g/L	500 g/L
Freundlich						
k_f [mg/g]	0.8521	1.038	1.656	0.0780	0.1044	0.2307
$1/n$	0.4660	0.3914	0.2795	0.5105	0.5308	0.6275
n	2.15	2.55	3.58	1.96	1.88	1.59
R^2	0.9605	0.9396	0.9042	0.9961	0.9926	0.9964
Langmuir						
Q_0 [mg/g]	13.30	9.99	5.80	2.38	2.08	1.84
b [L/mg]	0.0190	0.0338	0.1813	0.0059	0.0187	0.1287
R^2	0.9913	0.9948	0.9900	0.9268	0.9730	0.8312

Table 2. Thermodynamic parameters for arsenic(III) adsorption onto fly ash and fly ash agglomerates (2.5–5.0 mm)

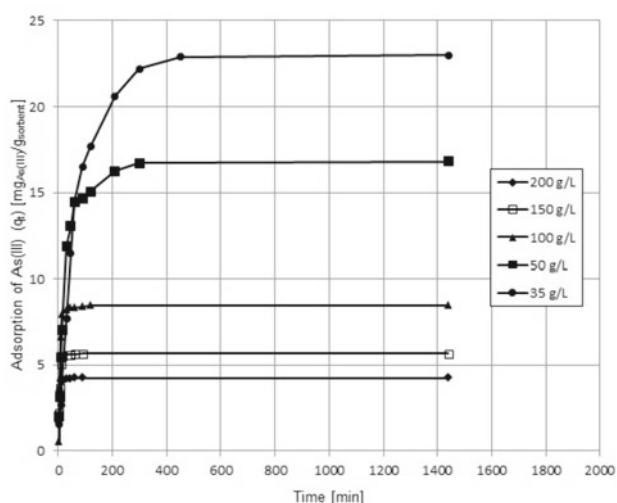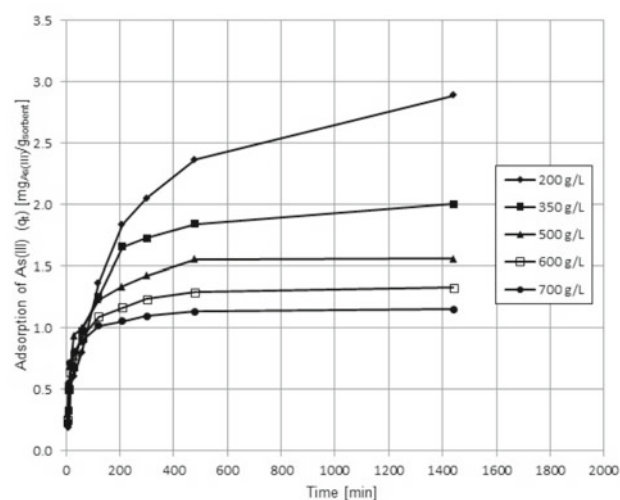| Adsorbent	T [K]	$\ln K_0$	ΔG^0 [kJ/mol]	ΔH^0 [kJ/mol]	ΔS^0 [kJ/mol · K]
Fly ash	298	2.4385	− 6.04	30.6	0.123
	308	2.7345	− 7.01		
	318	3.2780	− 8.67		
	328	3.5141	− 9.59		
Fly ash agglomerates (2.5–5.0 mm)	298	2.7498	− 6.81	28.2	0.119
	308	3.2059	− 8.21		
	318	3.5834	− 9.48		
	328	3.7863	− 10.30		

indicates the endothermic nature of the adsorption process. The positive ΔS^0 value suggests an increase in the randomness at the solid/solution interface during the adsorption process[1, 4, 21].

Other authors pointed out that an adsorption process can be exothermic or endothermic. Adsorption of arsenic(III) by macrofungus (*Inonotus hispidus*) biomass[1] and iron oxide-coated cement (IOCC)[4] showed an exothermic nature. The process is endothermic for the adsorption of arsenic(III) by a nanocrystalline hybrid surfactant–akaganeite sorbent[25] and synthetic siderite[14]. Adsorption on the fly ash adsorbent can be exothermic as well as endothermic, for example for chromium, lead[28] and boron[21] is exothermic, while for cadmium and nickel – endothermic[29].

Adsorption kinetics

In order to investigate the mechanisms of sorption and potential rate controlling step, such as mass transport and chemical reaction processes, some various kinetic and diffusion models can be used to test the experimental data. The sorption process can be described by four steps: (i) transport in the bulk of the solution, (ii) diffusion across the film surrounding the sorbent particles, (iii) diffusion in the liquid contained in the pores and in the sorbate along the pores walls, (iv) sorption and desorption within the particle and on the external surface[27]. Figure 8 and Figure 9 show the kinetics of arsenic(III) adsorption onto fly ash and fly ash agglomerates, respectively.

The kinetic data were elaborated using three different kinetic models: the pseudo-first order, the pseudo-second order and the intraparticle diffusion equation.

Figure 9. Adsorption kinetics of As(III) on fly ash agglomerates (pH~9; T = 25°C; initial concentration of As(III): 1000 mg/L)

The linear form of the pseudo-first order rate equation[6, 15, 16, 27, 30] is given as:

$$\ln(q_m - q_t) = \ln q_m - k_1 \cdot t \qquad (6),$$

where: q_m and q_t [mg$_{As(III)}$/g$_{adsorbent}$] are the amounts of solute adsorbed at equilibrium and at any time t [min], respectively, k_1 is the rate constant of the equation [1/min]. The adsorption rate constants (k_1) was determined experimentally by plotting of $\ln(q_m - q_t)$ versus t.

The linear form of the pseudo-second order model[2, 7, 9, 12, 14, 15, 19, 27] is as follows:

$$\frac{t}{q_t} = \frac{1}{k_2 \cdot q_m^2} + \frac{t}{q_m} \qquad (7)$$

where: k_2 is the adsorption rate constant [g/mg · min], q_m is the amount of adsorbate at equilibrium [mg$_{As(III)}$/g$_{adsorbent}$], q_t is the amount of an adsorbate at any time and $k_2 \cdot q_m^2 = h$ denotes the initial adsorption rate [mg/g · min].

The intraparticle diffusion model[27, 31, 32] is given as:

$$q_t = a + k_d \cdot t^{1/2} \qquad (8)$$

where: q_t [mg$_{As(III)}$/g$_{adsorbent}$] is the amounts of arsenic(III) adsorbed at time t [min], a is the intraparticle diffusion constant [mg/g] and k_d is a measure of diffusion coefficient [mg/g · min$^{1/2}$]. In the intraparticle diffusion process, a plot of q_t versus $t^{1/2}$ should give a straight line passing through the origin, otherwise when the intercept is not zero, some degree of the external the film mass transfer or boundary layer control exists. The larger the intercept, the greater the layer effect[32].

The parameters of the examined models were shown in Table 3, along with correlation coefficients (R^2). The best fit for the experimental data of this adsorption study

Figure 8. Adsorption kinetics of As(III) on fly ash (pH~11; T = 21°C; initial concentration of As(III): 1000 mg/L)

Table 3. Kinetics study calculations

Adsorbent	Solid/liquid ratios	h [mg/g · min]	k_2 [mg/g · min]	q_m [mg/g$_{solid}$]	R^2
PSO model					
Fly ash	35 g/L	0.4834	0.0008	24.06	0.9979
	50 g/L	1.044	0.0036	17.04	0.9998
	100 g/L	1.190	0.0163	8.532	0.9990
	150 g/L	2.245	0.0653	5.863	0.9980
	200 g/L	2.472	0.1306	4.350	0.9981
Fly ash agglomerates (2.5–5.0 mm)	200 g/L	0.198	0.0018	3.354	0.9992
	350 g/L	0.0293	0.0062	2.182	0.9993
	500 g/L	0.0603	0.0239	1.588	0.9996
	600 g/L	0.0572	0.0315	1.347	0.9998
	700 g/L	0.0752	0.0555	1.164	0.9999

Adsorbent	Solid/liquid ratios	k_1 [1/min]	q_m [mg/g$_{solid}$]	R^2
PFO model				
Fly ash	35 g/L	0.0050	11.5	0.8306
	50 g/L	0.0044	4.75	0.7488
	100 g/L	0.0031	0.523	0.4083
	150 g/L	0.0645	1.26	0.8321
	200 g/L	0.0594	0.470	0.7227
Fly ash agglomerates (2.5–5.0 mm)	200 g/L	0.0013	2.65	0.8671
	350 g/L	0.0010	1.13	0.7521
	500 g/L	0.0047	0.646	0.8495
	600 g/L	0.0045	0.570	0.9566
	700 g/L	0.0044	0.413	0.9088

Adsorbent	Solid/liquid ratios	k_d [g/mg · min]	a [mg/g$_{solid}$]	R^2
intraparticle diffusion model				
Fly ash	35 g/L	0.678	5.40	0.6637
	50 g/L	0.369	7.64	0.4579
	100 g/L	0.101	6.00	0.1603
	150 g/L	0.341	3.13	0.5392
	200 g/L	0.207	2.74	0.4069
Fly ash agglomerates (2.5–5.0 mm)	200 g/L	0.0453	0.649	0.8228
	350 g/L	0.0291	0.678	0.6710
	500 g/L	0.0331	0.643	0.6888
	600 g/L	0.0269	0.584	0.6623
	700 g/L	0.0233	0.719	0.5556

was achieved by the application of a pseudo-second order (PSO) chemisorption model. Adsorption rate constant (k_2) can be determined from the slope on the graph of t/q_t against t and its abscissa. A high correlation coefficient ($R^2 > 0,99$) confirms that the kinetics of arsenic adsorption onto fly ash and fly ash agglomerates follow this model. The initial adsorption rate h, for arsenic(III) adsorption onto fly ash as well as fly ash agglomerates, increases when the concentration of arsenic(III) in solution and per unit mass of adsorbent increases. The adsorption rate constant (k_2) and the amount of an adsorbate taken at equilibrium (q_m) are higher when the adsorption process was carried onto the non-agglomerated fly ash. From these results, it can be said that the pseudo-second-order kinetic model provided a good correlation for the arsenic(III) adsorption onto fly ash and fly ash agglomerates. Also, the literature data confirm this finding[1, 4, 14, 15, 19].

CONCLUSIONS

In order to optimize the adsorption conditions for arsenic(III) removal from aqueous solution by fly ash and fly ash agglomerates, isotherm, kinetic and thermodynamic studies have been completed.

Fly ash (13.5 mg$_{As(III)}$/g$_{sorbent}$) as well as fly ash agglomerates (5.7 mg$_{As(III)}$/g$_{sorbent}$) are good adsorbents of arsenic(III). Although fly ash can adsorb more arsenic(III) ions than the fly ash agglomerates. However, fly ash agglomerates are easier to use, because this material is less dusty and easier to separate from solution. Isotherm studies showed good fit with the Langmuir (fly ash) and the Freundlich (fly ash agglomerates) isotherm models. The experimental data of arsenic adsorption kinetic onto fly ash and fly ash agglomerates can be fitted the by pseudo-second-order model (PSO). The temperature dependence of arsenic(III) adsorption was investigated and the values of the calculated thermodynamic parameters: ΔG^0 (from –10.3 to –6.04 kJ/mol), ΔH^0 (about 30 kJ/mol) and ΔS^0 (about 0.12 kJ/mol · K) showed endothermic and spontaneous nature of an adsorption process.

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