

BATCH SORBER DESIGN USING EQUILIBRIUM AND CONTACT TIME DATA FOR THE REMOVAL OF LEAD

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Abstract. The two-stage batch sorption design of the sorption of lead ions onto peat has been studied based on a Langmuir isotherm type equation obtained from different contact time studies. A model has been developed to predict the optimised minimum operating time to determine a specified amount of lead removal. The sorption capacity at any contact time has been evaluated with an equation also based on the mathematical form of the Langmuir equation.

Keywords: batch sorption, contact time optimisation, Langmuir equation, lead ions, two-stage

1. Introduction

Health problems due to lead in water are well documented (Freedman *et al.*, 1990; Goldstein, 1990). It is therefore important to consider the removal of lead from any processed waters, including both potable waters and wastewaters. Lead contained in wastewaters often ends up being recycled back into drinking water sources.

Several workers have studied the removal of lead by sorption processes, including the sorption of lead onto used tea leaves (Tan and Abddul-Rahman, 1988), the sorption of lead onto china clay and wollastonite (Yadava *et al.*, 1991) and onto modified groundnut (*Arachis hypogea*) husks (Okieimen *et al.*, 1991). However, there is only limited data recorded in the literature on the design of sorption systems for the removal of lead ions. Fixed bed studies have been carried out (McKay and Bino, 1987; Kuennen *et al.*, 1992) but there is no design data for batch contact systems.

Most previous optimisation models for batch adsorbers (Helferich, 1962; Lerch and Rathowsky, 1967; McKay and Bino, 1987; Ozer *et al.*, 1997) are based on minimising the mass of adsorbent required to remove a certain amount of metal ions from a fixed volume of wastewater. This optimised parameter is a critical factor when using expensive adsorbents, such as activated carbons, zeolites and silica, because it makes maximum use of the adsorbent but gives little consideration to contact or hold-up time. In recent years, many cheap sorbent materials have been identified such as the removal of cadmium using 'waste' hydroxide (Namasivayam and Ranganathan, 1995), ferric oxide (Dzomback and Morel, 1986), ceric oxide



(Mishra and Singh, 1995) and beech leaves (Salim *et al.*, 1992); the removal of copper using fly ash (Panday *et al.*, 1985), peat (Ho *et al.*, 1994) and chitosan (Findon *et al.*, 1993); the removal of chromium using leaf mould (Sharma and Forster, 1994), 'spent' bleaching earth (Low and Lee, 1996) fly ash (Dasmahapatra *et al.*, 1996; Gupta *et al.*, 1988), and mercury on kaolinite (Singh *et al.*, 1996). Peat has been used successfully to treat a wide range of metal ion bearing wastewaters (Viraraghavan and Dronamraju, 1993; Allen *et al.*, 1992; Sharma and Forster, 1993; Ho *et al.*, 1995; Gosset *et al.*, 1986) which include cadmium, copper, chromium, lead, mercury, nickel and zinc.

In many countries, however, industries find that space management is a major challenge and optimising the rate of treatment of a fixed volume of wastewater is crucial. Therefore using these cheaper sorbents and minimizing the contact time to achieve a desired fraction/percentage of metal ion removal, with a fixed mass of sorbent will enable the treatment of more batches of polluted wastewater per day. Thus, the process plant equipment items can be reduced in size, with a resultant decrease in capital cost.

The present article presents a novel model to predict the optimum contact time for the removal of lead ions by peat in a two stage agitated batch sorption system. The model is based on predicting the minimum contact time required to remove a fixed percentage of lead ions from a given volume of waste water effluent of specified Pb concentration, using a fixed mass of peat. In the more conventional studies mentioned previously, the optimisation emphasis for expensive sorbents is based on maximum sorbent utilisation rather than minimum contact time and reduced plant size.

2. Materials and Methods

The experiments were conducted with sphagnum peat moss obtained from Germany. The peat, as supplied, was dried at a temperature of 90 ± 2 °C for 24 hr. The sample was broken up using a hammer mill and then screened to obtain a discrete particle size range of 500–710 μm . The peat surface area was found to be $26.5 \text{ m}^2 \text{ g}^{-1}$ by BET analysis using nitrogen. The absolute peat density using hexane was 1225 kg m^{-3} and the equilibrium moisture content was 14.0% by weight. The ash content was 1.9% by weight and elemental analysis was determined to be: C = 51.2%; H = 5.1%; N = 1.1%; S = 0.6%.

The lead nitrate used in the experiments was supplied by Aldrich Chemicals. Stock solutions of lead nitrate (analytical grade $\text{Pb}(\text{NO}_3)_2$ (3.26 g dm^{-3})) were prepared in deionised, distilled water. All solutions used in this study were diluted with distilled water as required.

Equilibrium isotherms were determined by shaking a fixed mass of peat (0.5 g and particle size 500–710 μm) with 125 mL lead ion solution in screw cap plastic bottle at room temperature (20 ± 2 °C). A range of lead ion concentrations, from

100 to 500 mg dm⁻³ at increments of 50 mg dm⁻³, were used. Shaking, using a Griffin shaking water bath, was carried out for five days, and then the solution was filtered and analysed using inductively coupled plasma atomic emission spectroscopy (ICP-AES).

A series of contact time experiments were carried out in an agitated (400 rpm) batch adsorber vessel at different initial metal ion concentrations. Samples were taken, and metal ion concentrations were determined at a series of fixed contact times using contact times from 10 to 240 min and lead ion concentrations from 100 to 500 mg dm⁻³. All the batch contact time investigations were carried out using a baffled agitated 2 dm³ sorber vessel. Samples were withdrawn at suitable time intervals, filtered through a 0.45 μm membrane filter to remove particulates and then these filtered solution samples were analysed using an inductively coupled-atomic emission spectrophotometer. Standards were used to regularly check the instrument's calibration setting and triplicate measurements of samples were made. The methodology was the one recommended by the USEPA (1980) for metal ions in solution. A 6.8 g sample of peat (500–710 μm) was added to each 1.7 dm³ volume of lead ion solution using an agitation speed of 400 rpm at 24±2 °C.

3. Results and Discussion

Batch-type processes are usually limited to the treatment of small volumes of wastewaters. The efficiency of solute removal can be improved if the solution is treated using a number of batch stages rather than a single-stage batch process. In a batch sorption process, sorbent is contacted with the solution in a tank for a period of time at fixed operating conditions. The sorbent is separated from the mixture by settling, filtration or centrifugation, and then regenerated or discarded. The time required to approach equilibrium depends on concentration of sorbate, dose and particle size of sorbent, degree of agitation, temperature and pH value. For batch operations, the sorbent is usually applied as small particle size material, less than 1000 microns, to increase the sorbent external surface area and reduce the diffusion resistance inside its pores. But in fixed bed adsorbers, a small adsorbent particle size can cause problems due to high pressure drop. Agitation of the solution/particle system improves contact of sorbent with solution and decreases the mass transfer resistance at the particle surface. Higher pH values of the solution improve the ion-exchange potential in the system when the sorption rate is largely controlled by ion-exchange reactions rather than complexing reactions. Endothermal sorption reaction can also be improved by using higher temperatures of reaction.

Sorption equilibrium can be represented by the Langmuir isotherm as given below

$$q_e = \frac{X_{m,e} a_{L,e} C_e}{1 + a_{L,e} C_e} \quad (1)$$

where

- q_e = the sorption capacity at the equilibrium solute concentration C_e , (mg metal ion g^{-1} peat);
 C_e = the concentration of sorbate in solution, (mg lead dm^{-3});
 $X_{m,e}$ = the maximum sorption capacity corresponding to complete mono-layer coverage, (mg lead g^{-1} peat);
 $a_{L,e}$ = a Langmuir constant related to the energy of sorption, ($\text{dm}^3 \text{mg}^{-1}$).

It is important to develop an equation which accurately represents the results of the capacity of the batch adsorber from the analysis of the isotherm data and which can be used for design purposes. An isotherm is shown in Figure 1 for the sorption of lead ions onto peat. The experimental data (crosses) are plotted in the form of lead ion sorbed per unit mass of peat, q_e , against the concentration of lead ion remaining in solution, C_e . These data were correlated using Equation (1) and the correlation coefficient, r^2 , was 0.999 for the agreement between the experimental data and the theoretical equilibrium data. The maximum sorption capacity of lead, $X_{m,e}$, and Langmuir constant, $a_{L,e}$, were determined from the slope and intercept of the Langmuir equation and were found to be 122 mg lead g^{-1} peat and 0.374 $\text{dm}^3 \text{mg}^{-1}$ peat, respectively. Figure 1 also shows the experimental contact time data (symbols ●, ■, ○, ▲, ◆) for initial lead ion concentrations of 101, 209, 309, 410 and 504 mg dm^{-3} with the operating lines which are generated using a slope of solution volume/peat mass ratio, that is $-L/S$. The final solution concentration, C_e , for a batch adsorber can be predicted from the point of intersection of the operating line on the sorption isotherm or using the mass balance Equation (2) for a single stage batch sorber.

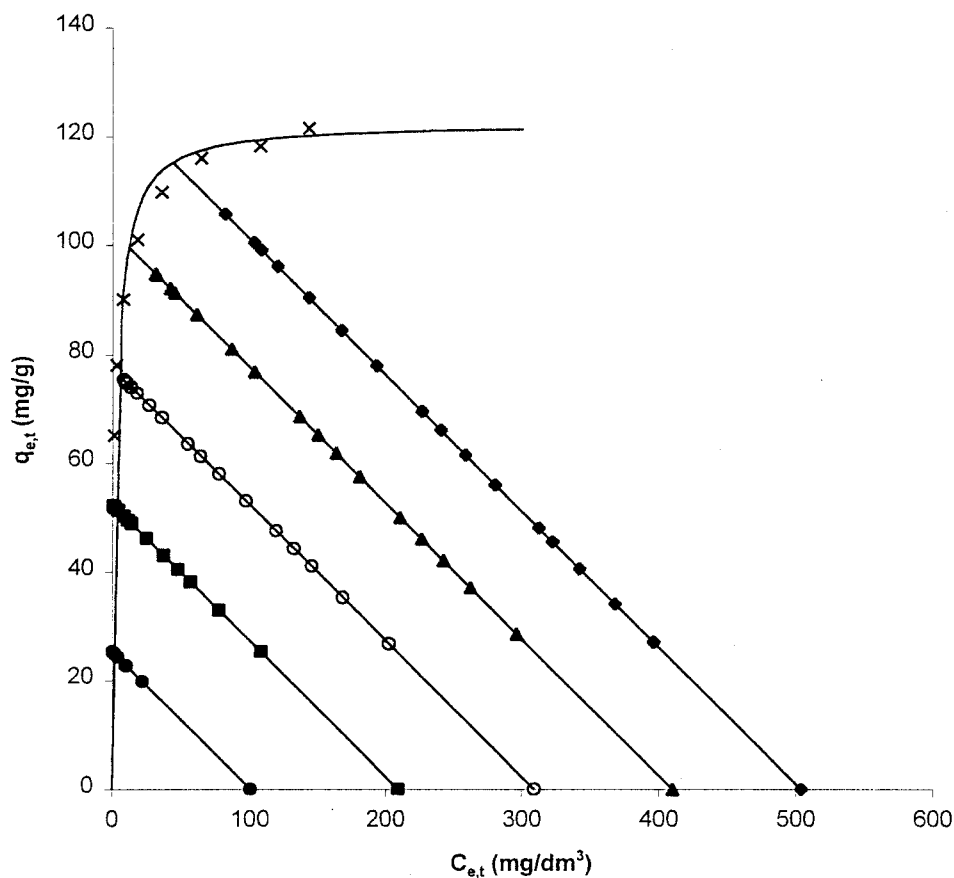
$$S(q_o - q_e) = L(C_o - C_e) \quad (2)$$

with

- q_o = the sorption capacity of peat at the start of the process (mg lead on 1g peat at time 0 (= 0 for fresh peat));
 q_e = the sorption capacity of lead on peat (mg lead g^{-1} peat at equilibrium);
 L = the volume of solution (dm^3);
 S = the mass of peat (g);
 C_o = the initial lead ion concentration in solution (mg lead dm^{-3});
 C_e = the equilibrium lead ion concentration in solution (mg lead dm^{-3}).

Rearranging the mass balance Equation (2) enables the equation for the operating line to be expressed in Equation (3) as:

$$-\frac{L}{S} = \frac{q_e}{C_o - C_e} \quad (3)$$



- x Equilibrium Data
- $C_o = 101 \text{ mg/dm}^3$, Experimental data
- $C_o = 209 \text{ mg/dm}^3$, Experimental data
- $C_o = 309 \text{ mg/dm}^3$, Experimental data
- ▲ $C_o = 410 \text{ mg/dm}^3$, Experimental data
- ◆ $C_o = 504 \text{ mg/dm}^3$, Experimental data
- Pb, Langmuir
- $C_o = 101 \text{ mg/dm}^3$, Operating Line
- $C_o = 209 \text{ mg/dm}^3$, Operating Line
- $C_o = 309 \text{ mg/dm}^3$, Operating Line
- $C_o = 410 \text{ mg/dm}^3$, Operating Line
- $C_o = 504 \text{ mg/dm}^3$, Operating Line

Figure 1. Langmuir equilibrium isotherm for the sorption of lead showing operating lines of initial lead concentrations.

Furthermore, the equilibrium sorption capacity, q_e , can be obtained from the operating line and the Langmuir equation (Figure 1) for any initial lead ion concentration, C_o , and peat mass to solution volume ratio, as shown by Equation (4).

$$q_e = \frac{-L}{S}(C_o - C_e). \tag{4}$$

The schematic diagram for a two-stage sorption system is shown in Figure 2. The solution to be treated contains L (dm^3) solution and the lead ion concentration is

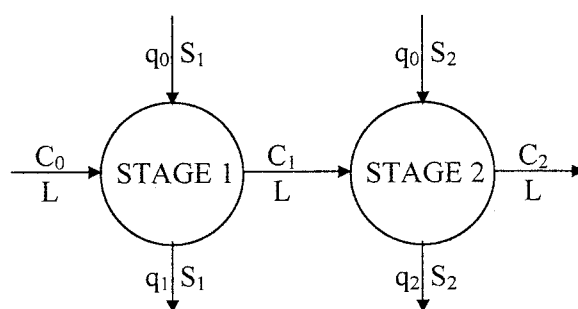


Figure 2. Schematic for a two-stage countercurrent batch sorption process.

reduced for each stage from C_{n-1} to C_n (mg dm^{-3}). Initially at $t = 0$, the amount of sorbent added is S g of peat with the lead ion concentration on the peat q_0 , (usually $q_0 = 0$ mg lead g^{-1} peat) and the lead ion concentration on the peat increases from q_0 to q_n mg g^{-1} of peat. The general mass balance equation for an n -stage batch sorber system is shown by Equation (5).

$$L_i(C_{o,i} - C_{e,n}) = S(q_{e,n} - C_0) . \quad (5)$$

The parameters C_e and q_e are based on equilibrium values at the n th stage. Usually, the contact time for sorption systems to reach complete equilibrium saturation is relatively long then the times could result in unreasonably long hold-up times. It is therefore better to replace these values by more realistic time dependent liquid phase concentrations, $C_{e,t}$, and sorbed phase concentrations, $q_{e,t}$. For multi-stage sorbers, the question that arises is how the mass balance equation can be solved and optimised to minimise total batch contact time. Essentially, analytical solutions are required to predict $C_{e,t}$ and $q_{e,t}$. Such solutions can be obtained by correlating the experimentally obtained contact time points, $C_{e,t}$ and $q_{e,t}$, on the operating lines in Figure 1. All the points at one contact time, for example 10 min, are taken and correlated. It was found that a Langmuir type Equation (6) gave excellent correlation of these 'constant time' lines:

$$q_{e,t} = \frac{X_{m,t} a_{L,t} C_t}{1 + a_{L,t} C_{e,t}} \quad (6)$$

The values for the twelve sample times from 10 to 240 min are given in Table I. The correlation coefficients are extremely high, all are above 0.997.

Table I shows the effective time dependent Langmuir equation parameters for different fixed agitation/contact times. Figure 3 shows these isotherm plots obtained for varying contact times. The corresponding linear plots of the values of $X_{m,t}$ and $a_{L,t}$ against time were regressed to obtain expressions for these values in terms of the agitation time with high correlation coefficients. It can be further

TABLE I
Fixed time dependent 'Langmuir equation' parameters

Time (min)	$X_{m,t}$ (mg g ⁻¹)	$a_{L,t}$ (g mg ⁻¹ min)	r^2
10	48.9	0.502	0.999
15	56.9	0.313	0.999
20	62.1	0.398	1.000
25	66.5	0.349	1.000
30	70.1	0.371	1.000
45	78.5	0.450	0.999
60	84.8	0.443	0.999
90	91.0	0.591	0.999
120	97.0	0.566	0.999
150	99.6	0.685	0.999
180	101.0	0.838	0.999
240	106.0	0.673	0.997

shown that $X_{m,t}$ and $a_{L,t}$ can be expressed as a function of time with correlation coefficients 0.999 and 0.953 as follows:

$$X_{m,t} = \frac{t}{8.95 \times 10^{-3}t + 0.152} \quad (7)$$

$$a_{L,t} = t1.27t + 34.3 . \quad (8)$$

Substituting Equations (7) and (8) (into (6)), means Equation (6) can be represented as follows:

$$q_{e,t} = \frac{\left(\frac{t}{8.95 \times 10^{-3}t + 0.152}\right) \left(\frac{t}{1.27t + 34.3}\right) C_{e,t}}{1 + \left(\frac{t}{1.27t + 34.3}\right) C_{e,t}} \quad (9)$$

This equation can then be used to predict the amount of lead ion sorbed for any sorption contact time and the time dependent Langmuir type equation parameters on a fixed time basis.

A three dimensional plot of Equation (9) is shown in Figure 4.

When fresh peat is used at each stage ($q_0 = 0 \text{ mg g}^{-1}$) and the time dependent Langmuir expression is used to describe the final state in a multi-stage sorption

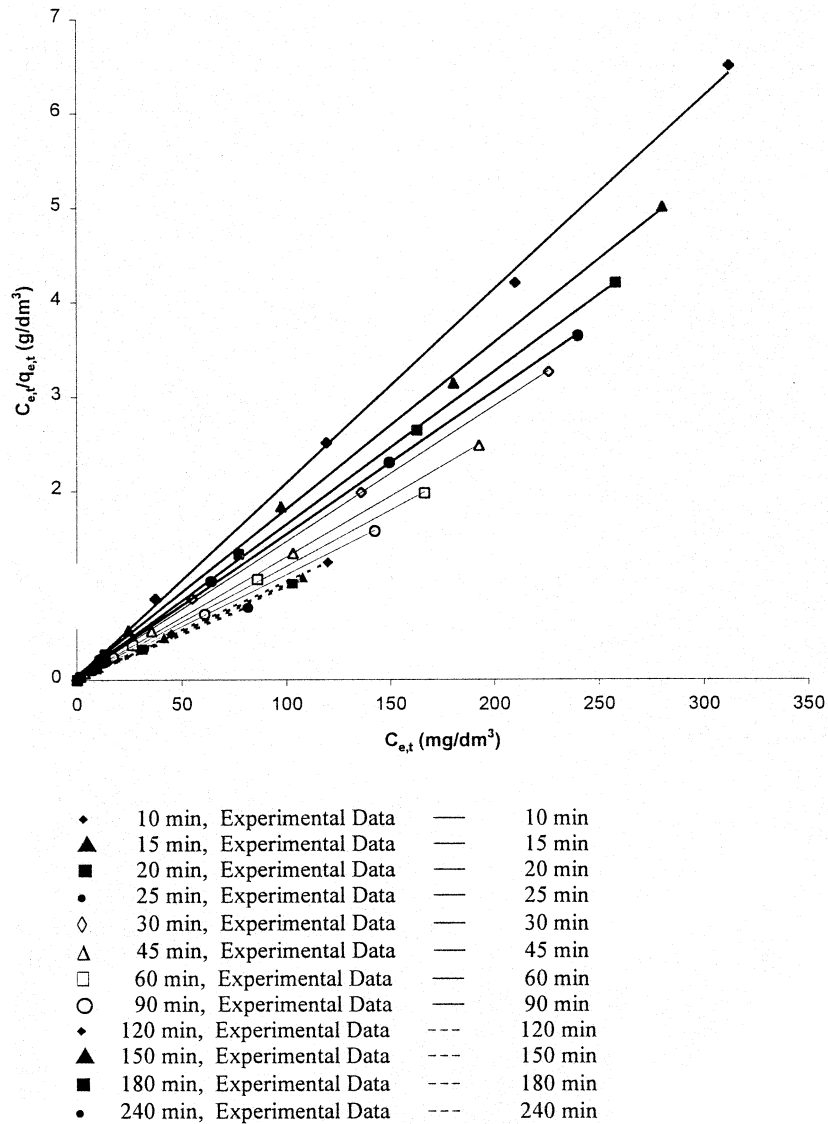


Figure 3. Langmuir style equation plots for the sorption of lead ion onto peat at varying fixed contact times.

system, the mass balance equation can be obtained by combining Equations (5) and (9):

$$C_{n-1} = C_n - \frac{S \left(\frac{t}{8.95 \times 10^{-3}t + 0.152} \right) \left(\frac{t}{1.27t + 34.3} \right) C_n}{L \left[1 + \left(\frac{t}{1.27t + 34.3} \right) C_n \right]} \quad (10)$$

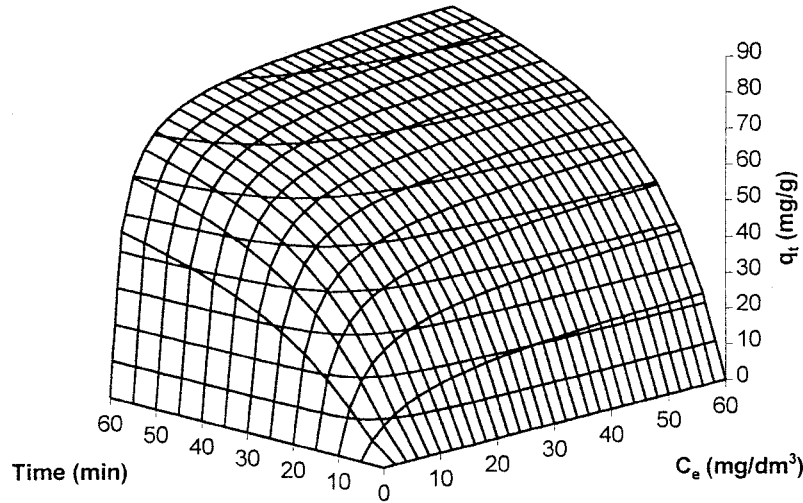


Figure 4. Relationship between time, C_e and q_t on the sorption of lead ion onto peat.

The total amount of lead ion removal can be calculated analytically as follows:

$$\sum_{n=1}^n (C_{n-1} - C_n) = \sum_{n=1}^n \frac{S \left(\frac{t}{8.95 \times 10^{-3}t + 0.152} \right) \left(\frac{t}{1.27t + 34.3} \right) C_n}{L \left[1 + \left(\frac{t}{1.27t + 34.3} \right) C_n \right]} . \quad (11)$$

The lead ion removal, R_n , in each stage can be evaluated from Equation (12) as follows:

$$R_n \frac{100(C_{n-1} - C_n)}{C_0} = \frac{100S \left(\frac{t}{8.95 \times 10^{-3}t + 0.152} \right) \left(\frac{t}{1.27t + 34.3} \right) C_n}{LC_0 \left[1 + \left(\frac{t}{1.27t + 34.3} \right) C_n \right]} . \quad (12)$$

The total removal of lead ions can be determined analytically from Equation (13):

$$\sum_{n=1}^n R_n = \frac{100S}{LC_0} \sum_{n=1}^n \frac{\left(\frac{t}{8.95 \times 10^{-3}t + 0.152} \right) \left(\frac{t}{1.27t + 34.3} \right) C_n}{1 + \left(\frac{t}{1.27t + 34.3} \right) C_n} . \quad (13)$$

Equations (12) and (13) can be used for predicting the removal of lead at any specified initial lead concentration and batch contact sorption time for any n -stage sorber system. A typical example is considered for the case of a two stage counter-current batch sorption, when the solution to be treated contains 5 m³ solution, the amount of peat added is 10 kg of peat in each of the two stages and the initial lead ion concentration is 200 mg dm⁻³ in the first stage. A series of contact times from 10 min up to 55 min in 5-min increments has been considered in stage 1 of a two-stage batch sorber system, that is ten 5-min increments. Therefore, in Figures 5 and

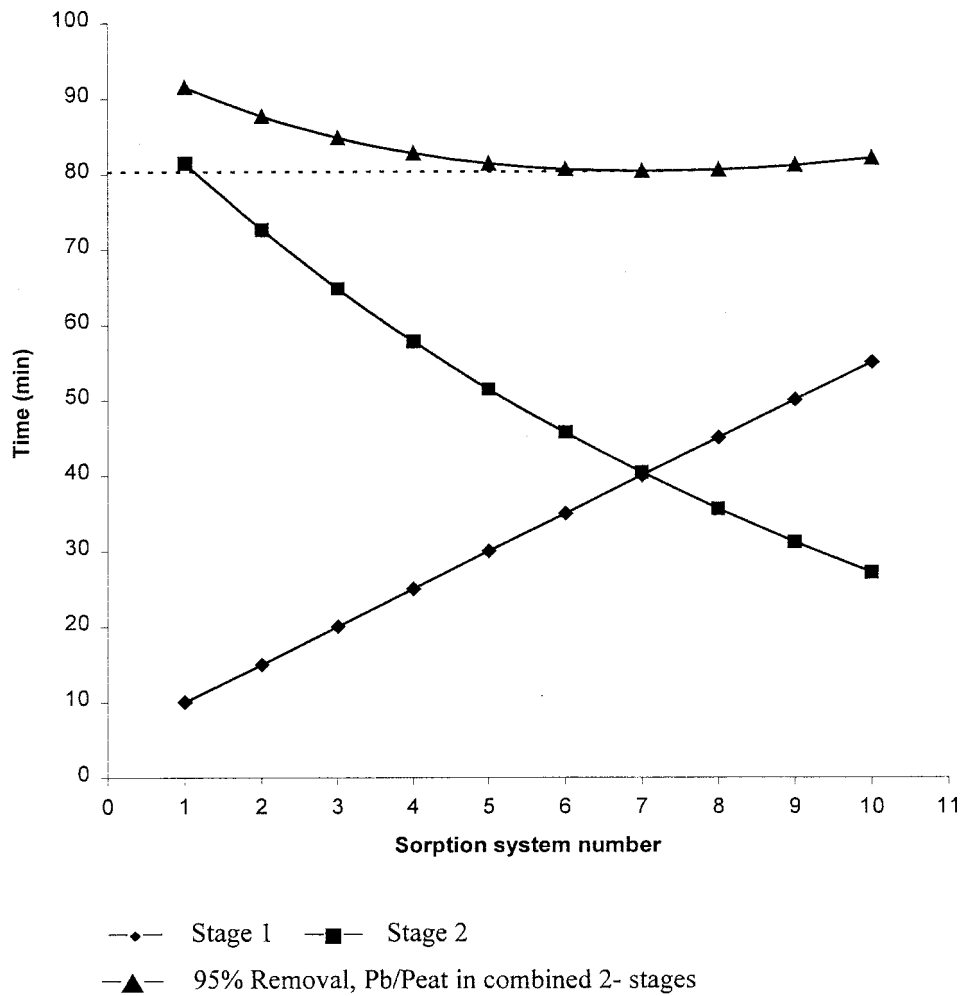


Figure 5. Comparison of 95% lead ion removal time of each stage in two-stage process.

6 each system number (1–10) is based on a 5 min contact time interval in stage 1 of the two stage system. In the first sorber for example, system number 4 implies that the first adsorber contact time is 10 min + (4–1) 5 min = 25 min, since system number 1 represents 10 min contact time in sorber number 1. Therefore the contact time in the second sorber, t_2 , is the time required, T minutes, to achieve a fixed total % lead ion removal minus the contact time in the first adsorber stage t_1 ; therefore:

$$T = t_1 + t_2 . \quad (14)$$

For N systems, t_1 , becomes:

$$t = 10 + (N - 1)5 \dots \text{min} . \quad (15)$$

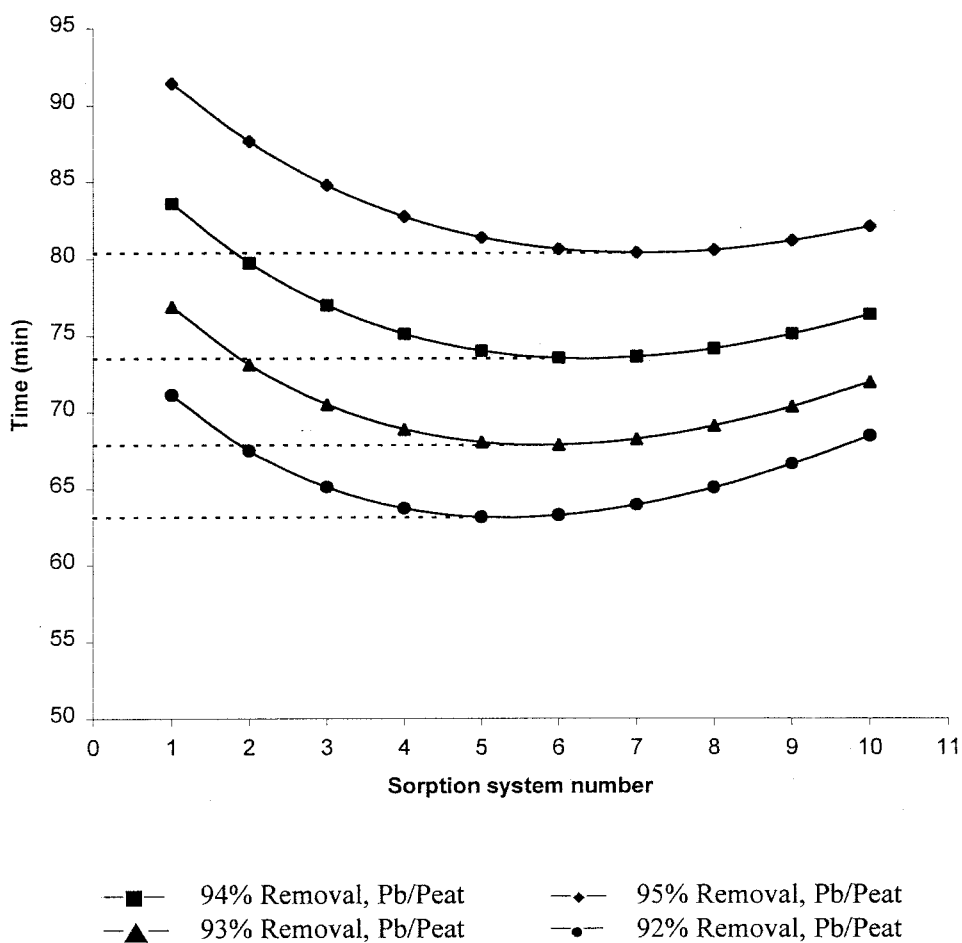


Figure 6. Minimum contact time for various percentage lead ion removal in a two-stage process.

And the total batch contact time, T , is:

$$T = 10 + (N - 1)5 + t_2 \tag{16}$$

The total contact time is calculated for each system number, $N = 1$ to $N = 10$ (based on the fixed t_1 values), for a fixed percentage dye removal. The T values are plotted against the system number, N , as shown in Figures 5 and 6, and the minimum contact time can be determined.

Figure 5 shows a comparison of the time for 95% lead ion removal for each stage and the total reaction time of the two-stage countercurrent batch sorption process. Figure 5 shows the reaction time for a number of two-stage sorption systems in series for each individual stage and for the combined total of the two stages. Table II shows a series sorption systems $N = 1-10$ for testing and the minimum total contact time for various percentage lead ion removals in the two-stage

TABLE II

Sorption times (min) to achieve different % Pb removals for a series of two stage sorption systems

System no.	Stage 1	Stage 2			
	Contact time	95% Removal time (min)	94% Removal time (min)	93% Removal time (min)	92% Removal time (min)
1	10	81.4	73.6	66.9	61.1
2	15	72.6	64.7	58.1	52.5
3	20	64.8	56.9	50.5	45.1
4	25	57.7	50.1	43.9	38.7
5	30	51.4	44.0	38.0	33.1
6	35	45.6	38.5	32.9	28.3
7	40	40.4	33.6	28.2	23.9
8	45	35.6	29.1	24.1	20.1
9	50	31.1	25.1	20.4	16.6
10	55	27.1	21.4	16.9	13.4

process can be evaluated from Equation (11) for each system. From the design criteria, the percentage lead ion removal by the sorber system is defined. In the two stage system in Table II, four examples have been shown, namely, 95, 94, 93 and 92% lead removal. The total contact time, T , to achieve this fixed percentage lead removal can be determined based on the fixed contact time assigned to Stage 1, t_1 . The data can then be plotted for the 10 systems at a fixed percentage dye removal for Stage 1, Stage 2 and Stage 1 + Stage 2 as shown by the three curves in Figure 5. Figure 5 shows the minimum contact time for various percentage lead ion removal values in a two-stage process. The minimum contact time for 95% lead ion removal can be found, using Equation (11) or by plotting graphs analogous to Figure 5. For 95% lead ion removal, the minimum contact time is 80.4 min as shown by system 7 with reaction times of 40 min for stage 1 and 40.4 min for stage 2; for 94% lead ion removal, the minimum time is 73.5 min as shown by system 6 with reaction times of 40 min for stage 1 and 38.5 min for stage 2; for 93% lead ion, the total minimum removal time is 67.9 min; and for 92% lead ion removal, the time is 63.1 min. The data for Stage 1 + Stage 2 at four percentage lead removals are presented in Figure 6. The minimum total contact time to achieve each percentage lead removal is shown by the dashed line.

4. Conclusions

An optimisation model has been developed to minimise the total contact time for the removal of lead ions in a two stage agitated batch sorber process. The minimum time to achieve a fixed percentage of lead removal from wastewater by sorption onto peat can be readily predicted. The relationship between time and sorption capacity is obtained by developed time dependent Langmuir style analytical equations which can be readily incorporated into the removal capacity equations. The model is novel since it minimises contact time rather than maximizing sorbent utilisation.

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