

# Removal of lead ions from aqueous solution using sphagnum moss peat as adsorbent

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## Abstract

The use of sphagnum moss peat for the removal of lead from aqueous solution has been studied in batch experiments. Investigations include the effect of pH and temperature, and kinetics of adsorption. The adsorption equilibria data were found to follow Langmuir models. It was found that the efficiency of lead removal using sphagnum moss peat as adsorbent depended very little either on the reaction temperatures (10 to 40°C) or on the initial pH values (4.0 to 6.0). Kinetic data suggested that the adsorption process was endothermic and pore diffusion was not the only rate-determining step.

## Introduction

Lead is a toxic element which is conservative and which has cumulative characteristics. As such, it is classified by the EC Directive on Dangerous Substances as a List II material (Council of European Communities, 1976). The majority of lead discharges to the environment come from atmospheric and particulate sources. However, there are a range of industries which generate waste waters containing significant concentrations of lead; for example, the lead-acid battery industry (Broom et al., 1994; Squires, 1992), smelting (Hallowell et al., 1973; Carter and Scheiner, 1991), printed wiring board manufacturing (Strilko, 1992) and paper mills (Sharif Fazeli et al., 1991). Mine drainage can also give rise to inputs of lead to the aquatic environment (Larsen et al., 1973; Beelman, 1993). Excessive amounts of lead in the human body can cause hypertension and brain damage. It is, therefore, essential that there are technologies for controlling the concentrations of lead in aqueous emissions.

The technologies which have been used range from the use of granular activated carbon (Reed and Arunachalam, 1994; Reed and Berg, 1993) to chemical treatments (Carter and Scheiner, 1991) and cross-flow filtration (Broom et al., 1994). However, these are processes which are more applicable to developed countries and there is a need to examine technologies which can be used in developing countries. One approach is to use alternative adsorbents. These are low-cost, often naturally occurring, products which have good adsorbent properties. A range of products have been examined previously. These include clays (Farrah et al., 1980), pine bark (Teles de Vasconcelos and Gonzalez Beca, 1994) and penicillium biomass (Niu et al., 1993). Good results have also been reported for peat (McLellan and Rock, 1988). This paper reports the results of a further study into the removal of lead from aqueous solutions by sorption onto sphagnum moss peat in which the effects of contact time, temperature and pH were examined.

## Materials and methods

### Materials and analytical methods

Sphagnum moss peat, which is commercially available in Britain and Ireland, was dried in an oven for 24 h at 105°C and then screened through a 14 mesh sieve (1.18 mm aperture; B.S. 410/43) to remove any large non-peat solids.

Analytical grade reagents were used in all cases. A stock lead(II) solution (1 000 mg/l) was prepared in distilled water using lead nitrate. All working solutions were prepared by diluting this stock solution with distilled water.

The concentration of lead(II) was determined by acetylene-air atomic absorption spectrophotometry (Instrumentation Laboratory aa/ae spectrophotometer 751) using a single element hollow cathode lamp.

### Experimental procedures

Batch sorption experiments were carried out at the desired temperature (10, 20, 30 and 40°C) on a rotary shaker at 100 r/min using capped 250 ml conical flasks.

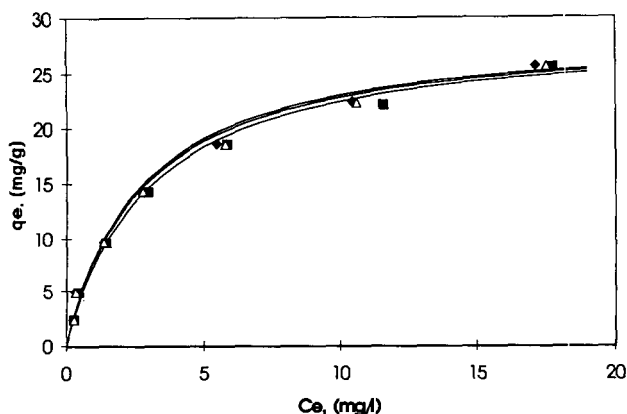
In the adsorption isotherm tests, peat (0.40 g) was thoroughly mixed with aqueous solutions of lead (100 ml). The initial pH adjustments were carried out either by sulphuric acid or sodium hydroxide solution and recorded as pH<sub>in</sub>. Three different values of pH<sub>in</sub> were used; 4.0, 5.0 and 6.0; with initial lead(II) concentrations (C<sub>0</sub>) ranging from 10 mg/l to 120 mg/l. After shaking the flasks for 4 h, the reaction mixtures were filtered through filter paper (Whatman No. 1) and the filtrates analysed for pH (recorded as pH<sub>fin</sub>), and the concentration of lead(II) (Gaid et al., 1994).

In the sorption kinetic experiments, peat (1.2 g) was thoroughly mixed with 350 ml lead(II) solution (100 mg/l). The peat suspensions were shaken for 3 h at the desired temperature (10, 20, 30 and 40°C) using capped 500 ml conical flasks and 2 ml samples were collected and filtered through a 0.45 µm membrane filter every 3 min for the first 15 min, every 5 min for the next 15 min, every 10 min for next half hour every 15 min for the next hour and then at 30 min intervals for the final hour. The filtrates were analysed using atomic absorption spectrophotometry.

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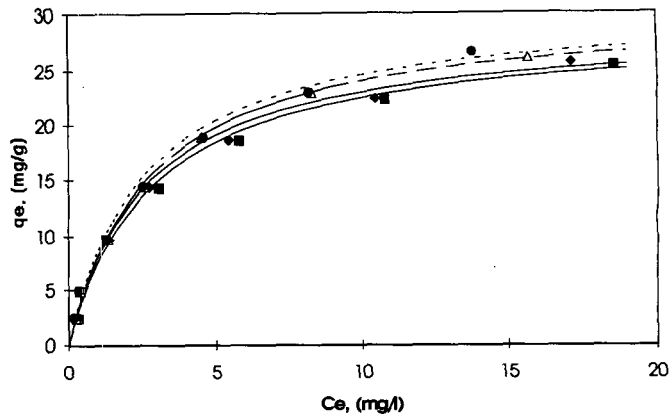
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**Figure 1**  
Typical adsorption isotherms at different pH values at 20°C

- pH = 4
- ◆ pH = 5
- △ pH = 6



**Figure 2**  
Typical adsorption isotherms at different temperatures at pH 5

- 10°C
- ◆ 20°C
- △ 30°C
- 40°C

## Results and discussion

### Adsorption isotherms

Figure 1 shows typical isotherms for the lead-peat system at different pH values and 20°C and Fig. 2 shows the effect of temperature at pH 5. In both cases the adsorption increased initially with concentration, but then reached saturation.

The data showed good compliance with the Langmuir equation.

$$\frac{C_e}{q_e} = \frac{1}{X_m k} + \frac{C_e}{X_m}$$

where  $X_m$  and  $k$  are Langmuir constants, related to the capacity and energy of adsorption respectively,  $q_e$  is adsorption density and  $C_e$  is equilibrium concentration.

The linearised form of the data enabled values of  $X_m$  and  $k$  to be calculated. These are presented in Table 1. It can be seen that the maximum adsorption capacity ( $X_m$ ) increased with increasing

reaction temperature for all the pH values. The highest values of adsorption capacity were found to be 30.7 mg/g of peat at an initial adjusted pH value of 5.0 and a temperature of 40°C. The results showed that the higher temperatures produced some improvement in the lead removal by adsorption on moss peat. The data also show that the changes in the initial pH value had a relatively insignificant effect on  $X_m$ .

The effect of isotherm shape can be used to predict whether an adsorption system is 'favourable' or 'unfavourable' both in fixed-bed systems (Weber and Chakravorti, 1974) as well as in batch processes (Poots et al., 1978). According to Hall et al. (1966) the essential features of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor or equilibrium parameter  $K_R$  which is defined by the following relationship:

$$K_R = \frac{1}{1 + kC_0}$$

This parameter indicates the shape of the isotherm accordingly:

Values of $K_R$	Type of isotherm
$K_R > 1$	Unfavourable
$K_R = 1$	Linear
$0 < K_R < 1$	Favourable
$K_R = 0$	Irreversible

The values of  $K_R$  at pH 4.0, 5.0 and 6.0, and 10, 20, 30 and 40°C are given in Table 2. The  $K_R$  values indicate that adsorption is more favourable for the higher initial lead concentrations than for the lower ones but the effects of temperature and pH on adsorption were slight, especially with the conditions used in this experiment.

### Effect of contact time and temperature

The removal of lead from water by adsorption on peat increased with time until equilibrium was accomplished (i.e. 150 min) and thereafter it became constant. At an initial concentration of 100 mg/l and a pH of 5.0, the amount adsorbed increased slightly from 22.0 (88.9% removal) to 22.7 mg/g (90.8% removal) with

**TABLE 1**  
LANGMUIR CONSTANTS OF LEAD-PEAT BINDING

pH <sub>in</sub>	Temp (°C)	$X_m$ (mg/g)	$k$ (l/mg)	Correlation ( $r^2$ )
4.0	10	27.3	0.349	0.993
4.0	20	28.8	0.351	0.994
4.0	30	29.5	0.344	0.996
4.0	40	30.4	0.323	0.992
5.0	10	28.7	0.361	0.993
5.0	20	28.9	0.391	0.994
5.0	30	30.4	0.372	0.997
5.0	40	30.7	0.398	0.992
6.0	10	27.6	0.459	0.994
6.0	20	28.8	0.379	0.992
6.0	30	29.5	0.416	0.994
6.0	40	30.7	0.384	0.987

TABLE 2 K <sub>R</sub> VALUES BASED ON THE LANGMUIR EQUATION				
pH <sub>in</sub>	Temp (°C)	C <sub>0</sub> = 10 mg/l (K <sub>R</sub> value)	C <sub>0</sub> = 60 mg/l (K <sub>R</sub> value)	C <sub>0</sub> = 120 mg/l (K <sub>R</sub> value)
4.0	10	0.223	0.0456	0.0233
4.0	20	0.222	0.0454	0.0232
4.0	30	0.225	0.0461	0.0236
4.0	40	0.226	0.0464	0.0237
5.0	10	0.217	0.0441	0.0225
5.0	20	0.204	0.0409	0.0209
5.0	30	0.212	0.0428	0.0219
5.0	40	0.201	0.0406	0.0205
6.0	10	0.179	0.0350	0.0178
6.0	20	0.209	0.0421	0.0215
6.0	30	0.194	0.0385	0.0196
6.0	40	0.207	0.0416	0.0213

an increase in solution temperature from 10 to 40°C (Fig. 3). However, the amount of lead removed by adsorption depended only to a small extent on the reaction temperature. This slight increase in the adsorption capacity suggests that the adsorption process could be endothermic. Alternatively, it could be due to the desolvation of the adsorbing species, changes in the size of the porosities, or an enhanced rate of intraparticle diffusion of adsorbate as well as chemisorption.

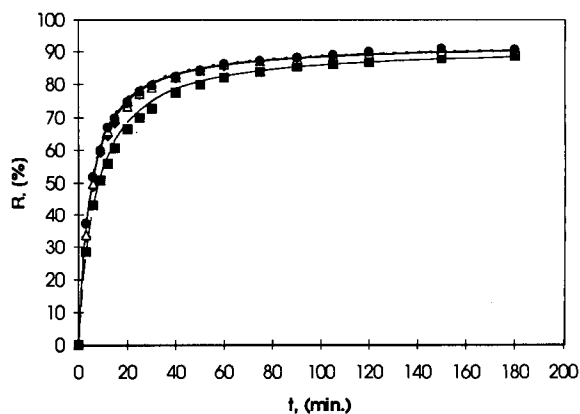
#### Effect of pH

The removal of metal ions from aqueous solution by adsorption is normally related to the pH of the solution which affects the surface charge and the degree of ionisation and species of adsorbate. Mittal and Venkobachar (1993) have investigated metal removal by sorbents in the pH range of 4.0 to 6.0 and reported that the maximum metal uptake was at pH 6.0. Hung and Wu (1977) have reported an abrupt increase in the adsorption of lead in the pH range of 5.0 to 6.0. Srivastava et al. (1989) have also pointed out that the removal of lead was almost the same, being in the pH range of 4.0 to 6.0. In this current study, using a pH range of 4.0 to 6.0, at four different temperatures, it was found that an increase in pH from 4.0 to 5.0 caused an increase in removal efficiency of 86.1 to 89.2% at 10°C, of 88.4 to 89.5% at 20°C, of 89.9 to 91.7% at 30°C and 90.9 to 91.8% at 40°C (Fig. 4). A further increase in pH to 6.0 caused a decrease in the removal efficiencies to 88.7, 89.4, 91.6 and 91.9% respectively. At the pH optimum, 5.0, the maximum amount of lead which could be absorbed was 30.7 mg/g. This figure can be compared with the value of 16.4 mg/g for copper determined in an earlier study (Ho et al., 1994).

Table 3 shows that, as the initial lead concentration (C<sub>0</sub>) increased, the resultant equilibrium pH decreased. As the reaction temperatures were raised to 40°C the resultant equilibrium pH values again decreased. It is, therefore, suggested that as more lead ions are adsorbed onto the peat, more hydrogen ions are released from the peat into the solution.

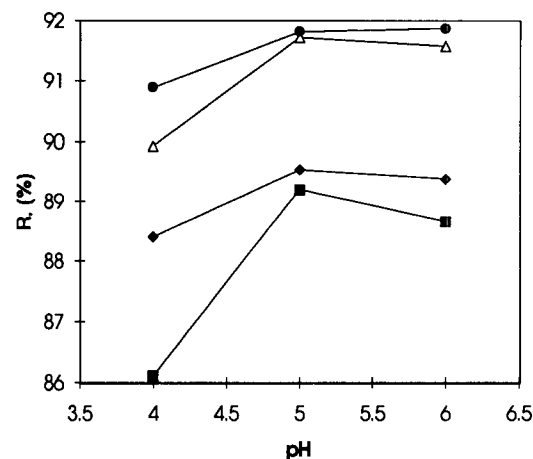
#### Removal of lead

The removal of lead process by various adsorption processes has been studied previously for a range of adsorbents (Table 4).



**Figure 3**  
The effect of contact time (*t*) and temperature on the removal efficiency (*R*) at an initial lead concentration of 100 mg/l

- 10°C
- ◆ 20°C
- △ 30°C
- 40°C



**Figure 4**  
Effect of pH on the removal efficiency (*R*) at an initial lead concentration of 100 mg/l

- 10°C
- ◆ 20°C
- △ 30°C
- 40°C

Yadava et al. (1989) have reported that the Langmuir maximum adsorption of lead ion by fly-ash was 0.368 mg/g at 20°C and a fly-ash dose of 20 g/l. These authors also reported (1991) that the Langmuir maximum adsorption of lead by China clay was 0.415 mg/g at 20°C and a China clay dose of 20 g/l, and with wollastonite it was 0.308 mg/g at 20°C and a dose of 20 g/l. Farrah et al. (1980) also examined the Langmuir maximum adsorption of lead. Using three different Na<sup>+</sup>-form clays, they found values of 3.93 mg/g for kaolin, 14.1 mg/g for illite and 71.8 mg/g for montmorillonite at 20°C and pH 5.0. McLellan and Rock (1988) reported that lead removal, using sphagnum moss peat, was 19.9 mg/g for an initial ion concentration of 50 mg/l and 40.0 mg/g for the initial ion concentration of 500 mg/l. Srivastava et al. (1989), in a study of the adsorption of lead ion by waste slurry, found that the maximum

**TABLE 3**  
**EQUILIBRIUM pH AT DIFFERENT INITIAL LEAD CONCENTRATIONS AND TEMPERATURES**

Initial lead concn. (mg/l)	10°C			20°C			30°C			40°C		
	pH4	pH5	pH6	pH4	pH5	pH6	pH4	pH5	pH6	pH4	pH5	pH6
0.0	3.6	4.1	4.1	3.8	4.0	4.1	3.7	3.9	4.0	3.5	3.7	3.9
10	3.6	3.9	4.0	3.7	3.9	4.0	3.6	3.9	3.9	3.5	3.7	3.8
20	3.6	3.9	3.9	3.7	3.8	3.9	3.6	3.8	3.8	3.4	3.6	3.7
40	3.5	3.7	3.8	3.5	3.7	3.7	3.5	3.6	3.7	3.3	3.5	3.5
60	3.4	3.6	3.6	3.4	3.5	3.6	3.4	3.5	3.6	3.2	3.4	3.4
80	3.3	3.5	3.5	3.3	3.5	3.5	3.3	3.4	3.5	3.2	3.3	3.3
100	3.3	3.4	3.4	3.3	3.4	3.4	3.2	3.4	3.4	3.1	3.2	3.2
120	3.2	3.3	3.3	3.2	3.3	3.4	3.2	3.3	3.3	3.0	3.1	3.2

**TABLE 4**  
**MAXIMUM CAPACITIES FOR ADSORPTION OF LEAD**

$X_m$ (mg/g)	Capacity (mg/g)	pH	Dose (g/l)	Material	Reference
0.368	-	-	20	Fly-ash	Yadava et al. (1989)
0.415	-	-	20	China clay	Yadava et al. (1991)
0.308	-	-	20	Wollastonite	
3.93	-	5.0	-	Kaolin clay	Farrah et al. (1980)
14.1	-	5.0	-	Illite clay	
71.8	-	5.0	-	Montmorillonite clay	
-	(i) 19.9	-	10	Sphagnum moss peat	McLellan and Rock (1988)
-	(ii) 40.0	-	10	Sphagnum moss peat	
-	20.0	-	26.57	Peat (Rastunsuo)	Tummauori and Aho (1980)
1380	-	-	-	Waste slurry	Srivastava et al. (1989)
49.9	-	-	10	Moss ( <i>Calymperes delessertii</i> <i>Besch</i> )	Low and Lee (1987)
30.7	-	5.0	4	Sphagnum moss peat	This work

(i) Initial lead ion concentration was 50 mg/l  
(ii) Initial lead ion concentration was 500 mg/l

**TABLE 5**  
**INTRAPARTICLE DIFFUSION CONSTANTS**

Temp (°C)	$K_t$	b	Correlation ( $r^2$ )
10	25.6	0.309	0.986
20	32.3	0.265	0.954
30	33.9	0.253	0.944
40	35.2	0.242	0.957

adsorption was 1 380 mg/g at 27°C. Low and Lee (1987) reported that the maximum sorption capacity for lead, on *Calymperes delessertii Besch* was 49.9 mg/l when the moss dose was 10 g/l. Tummauori and Aho (1980) also investigated the adsorption of lead ion solution by peat. Their results showed that the capacity of lead was 20.0 mg/g for the peat from Rastunsuo when the peat dose was 26.6 g/l. Although the comparison of the adsorption characteristics of different adsorbents tested under different conditions can be misleading, it is a procedure which is frequently done (Gaid et al., 1994). The results in Table 4 show that, in general:

- The current study produced data which were comparable with earlier work.
- The biosorbents (peat, slurry and moss) were more effective than the inorganic materials, with the exception of montmorillonite clay.

### Intraparticle diffusion

It is clear from Fig. 3 that the removal of Pb(II) at different initial temperatures takes place in two phases. The first phase of solute uptake; the "immediate solute removal" which is achieved relatively quickly, is followed by the "subsequent removal of solute" which extends over a longer period of time. These can be linearised by using the logarithmic expression;

$$R = K_i \cdot t^b \quad (1)$$

where R is the per cent lead removed, t is the contact time (min.), b is the slope of the linear plot and K is a constant coefficient.

The term b depicts the adsorption mechanism (Srivastava et al., 1987) and the term  $K_i$  may be taken as a rate factor (i.e. per cent solute removal per min). Higher values of  $K_i$  indicate an enhancement in the rate of removal, whereas larger b values indicate a better sorption mechanism, which may be due to an improved bonding between solute ions and adsorbent particles.

Regression analysis shows that the data, for the time scale from 6 to 50 min, do fit such an expression with correlation coefficients of 0.954 to 0.986. The adsorption data, as shown in Table 5, indicate that an increase in initial temperature causes some decrease in the slope, b, and an increase in the constant  $K_i$ . The larger values of b, indicating stronger bonding between adsorbate and adsorbent, also justify the concept of subsequent removal of solute by diffusion through peat particles, when temperature is lower at the solid-liquid interface.

Srivastava et al. (1987) have reported that, according to Weber and Morris (1963), the uptake of solute on adsorbents may involve two processes. One is transport of the adsorbate from solution to the particle/solution interface; the other is adsorption onto the accessible surface of the adsorbent particle. If intraparticle diffusion is the rate-limiting step, the value of b in the kinetic plots should be 0.5, while smaller values indicate the involvement of both the processes to an almost equal extent. In these investigations the values of b were found to range from 0.187 to 0.245, which indicates that pore diffusion is not the only rate-determining step in peat/lead adsorption. Transport of adsorbate from solution to the particle solution interface and the lead-binding reaction are also likely to be involved.

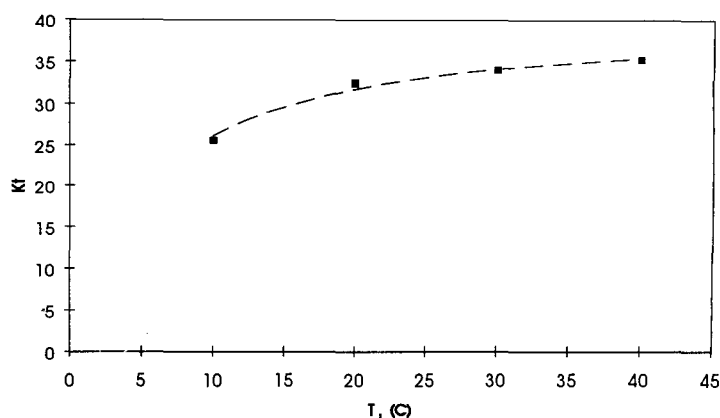
The variation of  $K_i$  against T shown in Fig. 5, can be expressed as:

$$K_i = \frac{T}{13.2 + 0.025T} \quad (2)$$

The variation of b against T, shown in Fig. 6 can be expressed as:

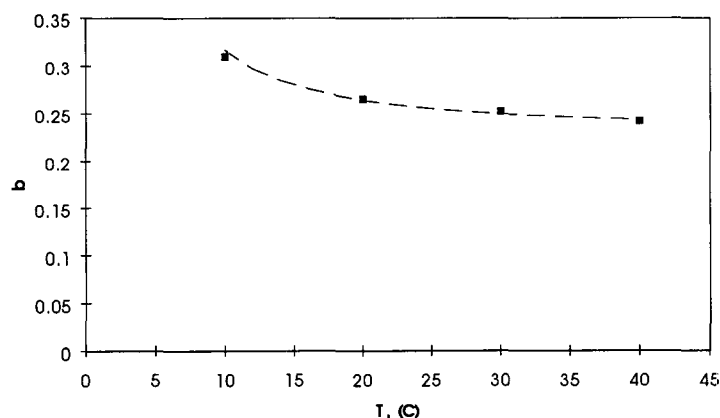
$$b = \frac{T}{4.42T - 12.6} \quad (3)$$

Combining Eqs. (1), (2) and (3) gives an empirical model for the



**Figure 5**

Variation in the rate factor ( $K_i$ ) with temperature at an initial lead concentration of 100 mg/l and a pH value of 5



**Figure 6**

Variation in the intraparticle diffusion constant (b) with temperature at an initial lead concentration of 100 mg/l and pH value of 5

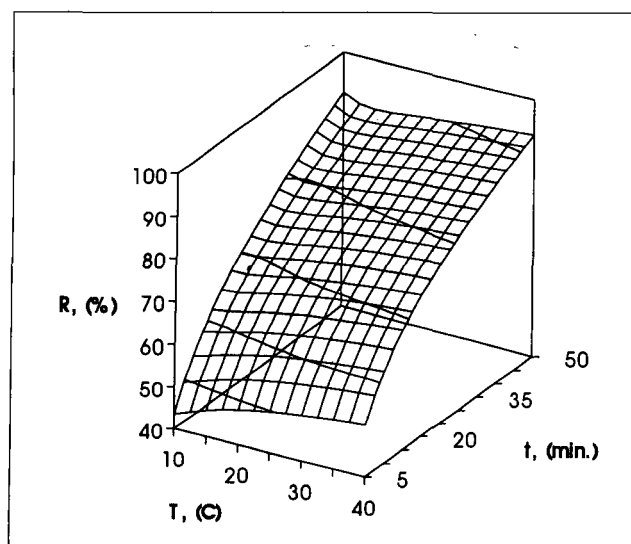
percentage removal at any time t:

$$R = \left[ \frac{T}{13.2 + 0.025T} \right] \cdot t^{T/(4.42T - 12.6)} \quad (4)$$

where T is initial temperature in °C and t is time in min. The results are shown in Fig. 7.

### Conclusions

The use of sphagnum moss peat in the removal of lead from aqueous solution was studied in batch experiments. The adsorption data can be described by Langmuir isotherms. The maximum adsorption capacity for sphagnum moss peat was 30.7 mg/g of peat, at a peat dose of 4 g/l, an initial adjusted pH of 5.0 and a temperature of 40°C. It is evident that sphagnum moss peat, a commercially available natural material, is a good adsorbent for lead(II) and it can be suggested for the removal of lead(II) from waste water. It was found that the efficiency of removal was only slightly dependent on the reaction temperature (10 to 40°C) and initial pH (4.0 to 6.0). Finally, kinetic data suggested that the adsorption process was endothermic and transport of adsorption from solution onto the



**Figure 7**

The relationship between the removal efficiency ( $R$ ), the reaction temperature ( $T$ ) and the contact time ( $t$ )

particle solution interface and the lead binding were both responsible for determining the rate.

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