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Adsorption of landfill leachates onto activated carbon Equilibrium and kinetics

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Abstract

The adsorption of stabilized leachates generated in a municipal landfill onto three commercial activated carbons has been investigated. Norit 0.8, Chemviron AQ40 and Picacarb 1240 have been used as adsorbents. Equilibrium experiments have been conducted to obtain the experimental isotherm profiles. Isotherms have been plotted based on the adsorption of general parameters, for instance chemical oxygen demand, total carbon, absorption at 410 nm and absorption at 254 nm. Different literature models and error functions have been used to adequately fit the experimental data. As a rule of thumb, three-parameter models do adjust experimental results better than two-parameter models. Norit 0.8 shows better adsorption characteristics than the rest of activated carbons, both in terms of contaminant level reduction of per unit mass of absorbent and in terms of the process kinetics.

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Keywords: Landfilling; Leachates; Adsorption; Activated carbon; Isotherm; Kinetics

1. Introduction

Comparative studies for urban solid wastes management and disposal have reported landfilling as the most suitable technology to deal with this type of wastes [1]. However, in spite of the economical benefits of the method and easiness of implementation, generation of toxic leachates might cause serious problems to the surroundings. Leachate from landfills can be a major hazard to health if the landfill is not properly operated and taken care of. These leachates can seep into the ground, moving into the nearby underground water supplies of a community with all the negative consequences implicated in the process.

Among technologies aimed at the reduction of the leachate hazardous nature, both secondary and tertiary treatments have been studied and reported in the specialised literature. Aerobic and anaerobic biodegradation processes have been applied to effectively reduce the contaminant load of leachates generated from young landfill sites [2,3]. Nevertheless, the main contaminant fraction of leachates from stabilized tips is composed by biologically refractory substances (i.e. humic substances), preventing, therefore, the adequate performance of secondary technologies [4]. As a consequence, tertiary treatments seem to be the alternative technologies to deal with leachates from landfills in the methanogenic phase. Thus, in order to fulfil the standards of quality for aqueous discharges or/and reutilisation of contaminated wastewaters, oxidation systems (UV, O₃, Fenton's reagent, advanced oxidation processes), membrane based technologies and adsorption processes have been investigated [5–7].

Among tertiary treatments, adsorption onto activated carbon (AC) has been reported as one of the most effective methods to remove high molecular weight compounds (present in stabilized leachates) from aqueous matrix. However, few works can be found on the use of AC to process sanitary landfill leachates, either as a single stage or in combination with other complementary technologies. Hence, Morawe et al. [7] investigated the performance of a two-column system in series as a post-treatment for a biologically treated leachate. With this system, chemical oxygen demand (COD) reductions up to 90% were observed (i.e. from roughly 900–80 ppm). Additionally,

Abbreviations: ARE, average relative error; CFEF, composite fractional error function; DMPSD, derivative Marquardt's percent standard deviation; Err², sum of error squares; SAE, sum of absolute errors

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Nomenclature

- A(t) adjustable parameter in Temkin equation (g or absorbance unit g⁻¹ AC)
- b adjustable parameter in Langmuir equation ((g or absorbance unit L^{-1})⁻¹).
- $B_{\rm T}$ adjustable parameter in Temkin equation ((g or absorbance unit $L^{-1})^{-1}$)
- C concentration in the liquid phase (g or absorbance unit L^{-1})
- k_{ads} kinetic parameter of modified Lagergren equation (h^{-1}) .
- $K_{\rm F}$ adjustable parameter in Freundlich equation ((g or absorbance unit g⁻¹ AC)(g or absorbance unit L⁻¹)ⁿ⁻¹)
- $m_{\rm AC}$ mass of activated carbon (g)
- *n* adjustable parameter in Freundlich equation
- Q concentration in the solid phase (g or absorbance unit $g^{-1} AC$)
- $Q_{\rm m}$ adjustable parameter in Langmuir equation (g or absorbance unit g⁻¹ AC)
- t_0 kinetic parameter of modified Lagergren equation (h)
- *V* liquid phase volume (L)

Greek letters

- $\begin{array}{ll} \alpha_R & \quad \mbox{adjustable parameter in Redlich-Peterson} \\ & \quad \mbox{equation (g or absorbance unit } g^{-1} \ AC) \end{array}$
- γ adjustable parameter in Redlich–Peterson equation
- δ_T adjustable parameter in Toth equation ((g or absorbance unit g^{-1} AC)(g or absorbance unit $L^{-1})^{\lambda-1})$
- ξ adjustable parameter in Sips equation
- λ adjustable parameter in Toth equation
- σ adjustable parameter in Toth equation (g or absorbance unit L⁻¹)
- $\Psi_{\rm D}$ adjustable parameter in Dubinin–Radushkevich equation
- ω_S adjustable parameter in Sips equation ((g or absorbance unit $L^{-1})^{-1}$)
- Ω_S adjustable parameter in Sips equation (g or absorbance unit g^{-1} AC)

Ramírez et al. [8] utilised and compared two combined processes, for instance coagulation-flocculation-activated carbon and Fenton's oxidation-activated carbon, claiming the improvement of the adsorption process after the chemical oxidation stage due to the generation of smaller and more adsorbable molecules. Moreover, Fettig et al. [9] also utilised a pre-oxidation step before running the adsorption step onto AC. However, in the lat-

| Table 1 | |
|---------|--|
|---------|--|

Leachate characterization from Badajoz landfill site

| 3600 | |
|------|--|
| 400 | |
| 250 | |
| 870 | |
| 300 | |
| 8.20 | |
| | 3600 400 250 870 300 8.20 |

Average values. Units in $mg L^{-1}$.

ter case, these authors reported an increase in the non-adsorbable fraction of leachates after the oxidation pre-treatment with ozone.

In addition to the lack of studies based on the usage of AC in landfill leachate remediation, equilibrium data of the system leachate-AC are rarely mentioned or studied rigorously. Isotherm data provide a fundamental tool at the time of design and scale-up of adsorbers. Similarly, kinetic results are valuable information to weigh up the suitability and effectiveness of the adsorption process. Again, few works can be found in the literature focused on the latter subject.

Consequently, in the present research, an analysis of the equilibrium adsorption of a stabilized leachate onto three commercial activated carbons is presented. Also, the kinetics of the adsorption process is investigated by using different amounts of activated carbon. Experimental data based on measurements of chemical oxygen demand (COD), colour at 410 nm (Abs₄₁₀), total carbon (TC) and absorbance at 254 nm (Abs₂₅₄) have been fitted to different models previously reported in the literature and the results discussed after the fitting process.

2. Experimental

Leachates were collected from the landfill site of Badajoz (South West of Spain). Table 1 summarizes the main characteristics of the leachates used in this study (average values are shown). From Table 1 it is observed that this effluent presents a low value of COD and BOD₅. The rates BOD₅/COD and TOC/COD situated in the range 0.1–0.3 and 0.2–0.4, respectively, and pH above 7 indicates the stabilized nature of the leachate. Among metallic species analysed, Al, Fe, Cr, Mn and Ni show the highest concentrations with other metals detected at trace levels. A periodic characterisation of these leachates can be found elsewhere [10].

Chemical oxygen demand (COD) was determined in a Dr. Lange spectrophotometer, the method based on the standard dichromate reflux method [11]. Total carbon (TC) concentrations were obtained by means of a DC-190 Dorhman analyzer. Absorbance of samples at 254 and 410 nm was determined by means of a U2000 model HITACHI spectrophotometer.

The commercial activated carbons used in this study were Norit 0.8, Picacarb 1240 and Chemviron AQ40. The main characteristics of manufactured ACs are detailed in Table 2.

The bottle point isotherm technique was employed to determine the equilibrium capacity of the commercial activated carbons investigated. Experiments were conducted at the original pH of the leachate. Accordingly, experiments were carried out in glass vials (25 cm^3 capacity) sealed by Teflon caps.

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