Adsorption of amoxicillin on two types of activated carbon

Mansouri Hayet*, Souissi Najar Souad, Ouederni Abdelmottaleb
Engineering Process and industrial systems, National School of engineers of Gabes,
Street Omar Ibn Elkhatab 6029
Gabes, Tunisia
hhayetmansouri@yahoo.fr

Abstract—This work investigates the adsorption of amoxicillin (AMX), widely consumed pharmaceutical, onto prepared and commercial activated carbons. Batch adsorption experiments of pure components in water were carried out to measure both adsorption equilibrium and kinetics. The influence of different operating parameters was investigated including initial concentration and size particles as results. Thermodynamic study suggests that the adsorption for both activated carbons is an exothermic process.

Keywords—Adsorption; kinetic; Amoxicillin; Activated carbons; thermodynamic.

I. INTRODUCTION

Groundwater contamination by pharmaceutical ingredients (analgesic, antibiotics, antidepressants, antiidiabetics, contraceptives, growth regulators, impotence drugs, painkillers, and tranquilizers) is an environmental problem of widespread concern [1] [2] [3]. Since these compounds are believed to have severe potential effects on the ecosystem and human health because of their continuous input to and persistence in aquatic environments [4]. Therapeutic groups most commonly detected in water are: anti-inflammatories and analgesics (paracetamol, ibuprofen …) and antibiotics (tetracyclines, macrolides, β-lactams, penicillins, quinolones, sulfonamides…) [5]. The selected compound in this study is amoxicillin (AMX) which is commonly found in wastewaters. Amoxicillin is the only phenolic penicillin and amoderate-spectrum β-lactam antibiotic used in humans and food-producing animals to treat several diseases. β-lactam antibiotics presents a structure based on a β-lactam ring responsible for the antibacterial activity and variable side chains that account for the major differences in their chemical and pharmacological properties [6]. The purpose of this work is to study the adsorption of amoxicillin from aqueous solutions onto activated carbons one prepared from olive stones by chemical activation (CACH) and a commercial one (CAC) for comparaison.

II. METHODS

A. Characterisation of activated carbons

Textural characterization was carried out by measuring the N2 adsorption isotherms at 77 K in an automatic apparatus (micromeritics ASAP 2010 M). Before the experiments, the samples were outgassed under vacuum at 120°C overnight. The N2 isotherms were used to calculate the specific surface area, \( S_{BET} \), total pore volume, \( V_T \).

B. Adsorption of amoxicillin on activated carbons

Batch equilibrium studies were carried out by adding a fixed amount of activated carbon (0.025 g) into 50 ml Erlenmeyer flasks containing 50 mL of different initial concentrations (10–500 mg/l) of AMX solution. The flasks were agitated in an isothermal bath shaker at 400 rpm and 30 °C for 24 h. The initial and equilibrium AMX concentrations were determined by absorbance measurement using UV/VIS spectrophotometer (Shimadzu, Model UV 1700) at 229 nm. It was then computed to AMX concentration using standard calibration curve. The amount of adsorption at equilibrium, \( q_e \) (mg/g), was calculated by:

\[
q_e = \frac{(C_0 - C_e)V}{m} \tag{1}
\]

Where \( C_0 \) and \( C_e \) (mg/l) are the liquid-phase concentrations of AMX at initial and equilibrium, respectively. \( V \) (l) is the volume of the solution and \( m \) (g) is the mass of dry adsorbent used.

III. RESULTS

C. Textural characterization of activated carbons

The textural characteristics of activated carbon obtained from N2 adsorption analysis are summarized in Table 1, indicating that the adsorbents CACH and CAC had a large specific surface area (1102 and 1370 m² g⁻¹) respectively.

<table>
<thead>
<tr>
<th>Sample</th>
<th>( S_{BET} ) (m²/g)</th>
<th>( V_{st} ) (cm³/g)</th>
<th>( V_{mv} ) (cm³/g)</th>
<th>( D_a ) (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CACH</td>
<td>1102</td>
<td>0.56</td>
<td>0.388</td>
<td>17</td>
</tr>
<tr>
<td>CAC</td>
<td>1370</td>
<td>1.318</td>
<td>0.971</td>
<td>38.47</td>
</tr>
</tbody>
</table>

TABLE 1: PHYSICAL CHARACTERISTICS OF ACTIVATED CARBONS
Kinetic study

For kinetic studies, 0.50 g of activated carbon was contacted with 1 L of AMX concentrations 50–100 mg/L using batch reactor at 30 °C. The agitation speed was kept constant at 400 rpm. At predetermined intervals of time, solutions were analyzed for the final concentration of AMX. The amount of adsorption $q_t$ (mg/g), at time $t$ (h), was calculated by (1).

$$q_t = \frac{C_0 - C_t}{m}$$

(a) Effect of contact time and AMX concentration

The adsorption of AMX by CACH and CAC was studied at different initial AMX concentrations (15, 30 and 60 mg/l). Fig. 3 shows the result for effect of initial concentration on adsorption of AMX. As can be seen from Fig.2, the amount of the adsorbed AMX increased from 25 to 46 and 50 mg g$^{-1}$ for respectively CACH and CAC with the increase of initial concentration from 15 to 60 mg l$^{-1}$. A similar result was found by [7] who found that the adsorption of 2, 4-dichlorophenoxyacetic acid depends on the initial concentration. Adsorption equilibrium was achieved in approximately 30mn. However, the experimental data were measured at 12 h to be sure that full equilibrium was attained.

(b) Effect of particle size

Experiments were also carried out to evaluate the effect of activated carbon particle size in the adsorption process. Three different particle sizes were tested: diameters less than 125 μm, 125-250 μm and 250-600 μm. Figure 3 shows the results obtained. The amount adsorbed increased from 14 to 53 mg g$^{-1}$ with the decrease in the particle size. Similarly, greater removal efficiency was achieved with smaller particle sizes (30 to 63%).

E. Adsorption Isotherm

Fig. 4 pictures the obtained results to the equilibrium isotherm of amoxicillin on the two activated carbons as described before. It reveals that the CAC adsorbs more AMX than CACH.
In order to confirm the adsorption nature in this study, thermodynamic parameters were determined. Entropy (ΔS°), enthalpy (ΔH°) and Gibbs free energy (ΔG°) were calculated. Table 3 presents the values of thermodynamic parameters. The enthalpy change is negative, suggesting that the adsorption process is exothermic.

<table>
<thead>
<tr>
<th>T (K)</th>
<th>ΔG° (kJ mol⁻¹)</th>
<th>ΔH° (kJ mol⁻¹)</th>
<th>ΔS° (J mol⁻¹ K⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CACH</td>
<td>CAC</td>
<td>CACH</td>
<td>CAC</td>
</tr>
<tr>
<td>303</td>
<td>10.463</td>
<td>10.445</td>
<td>-14.147</td>
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<tr>
<td>313</td>
<td>11.120</td>
<td>11.891</td>
<td>-12.034</td>
</tr>
<tr>
<td>323</td>
<td>11.733</td>
<td>12.097</td>
<td>-81.011</td>
</tr>
<tr>
<td>333</td>
<td>12.885</td>
<td>12.59</td>
<td>-74.221</td>
</tr>
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</table>

References


