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Review

A kinetic, equilibrium and thermodynamic study of L-phenylalanine adsorption using activated carbon based on agricultural waste (date stones)

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Abstract

The main purpose of this work is to produce low cost activated carbons from date stones wastes for the adsorption of L-phenylalanine. The activated carbons were prepared by chemical activation with KOH (ACK) and $ZnCl_2$ (ACZ) and characterized by scanning electron microscopy, N_2 adsorption–desorption isotherms and FT-IR spectroscopy. Both The activated carbons ACK and ACZ have high specific surface areas and large pore volumes, favorable for the adsorption. Batch experiments were conducted to determine the adsorption capacities. A Strong dependence of the adsorption capacity on pH was observed, the capacity decreases with increasing pH up to optimal value of 5.7. The adsorption follows a pseudo-second order kinetic model. Additionally, the equilibrium adsorption data were well fitted to the Langmuir isotherm, and the maximum adsorption capacities of L-phenylalanine onto ACK and ACZ were 188.3 and 133.3 mg g^{-1} at pH 5.7, respectively. The thermodynamic study revealed that the adsorption of L-phenylalanine onto activated carbons was exothermic in nature. The proposed adsorption mechanisms take into account the hydrophobic and electrostatic interactions which played the critical roles in the L-phenylalanine adsorption.

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Keywords: Activated carbon; L-phenylalanine; Size distribution; Sorbent surface; Adsorption isotherm; Thermodynamic

1. Introduction

Adsorption of amino acids onto solid surfaces has received much attention because of its scientific importance and applications in the separation and purification processes (Han & Yun, 2007; Hong & Bruening, 2006; Kostova & Bart, 2007; O'Connor et al., 2006; Sánchez-Hernández, Bernal, del Nozal, & Toribio, 2016). Amino acids are biomolecules of great relevance that are widely used in many industries such as food, cosmetic, medicine, biochemistry and others (Bourke & Kohn, 2003; Hartmann, 2005; Infante et al., 2004; Oshima, Saisho, Ohe, Baba, & Ohto, 2009; Palit & Moulik, 2001). They are non-toxic and are used as building blocks for the production of pharmaceutical and agrochemical compounds. In addition, they are interesting molecules as adsorbates because of their molecular size and zwitterionic nature (O'Connor et al., 2006). Similar to many amino acids, L-phenylalanine is essential for animals and the human body. It is extensively used as ingredient in food or feed additive, in infusion fluids, neutraceutical and pharmaceutical (Pimentel, Alves, Costa, Fernandes, et al., 2014; Pimentel, Alves, Costa, Torres, et al., 2014; Zhou, Liao, Wang, Du, & Chen, 2010). Generally, the amino acids have been studied by adsorption on well-ordered surfaces of solids. On the other hand, most of the current methods employed for the removal of L-phenylalanine from protein hydrolysates are based on the adsorption on activated carbon, polymeric resins, zeolites and ion exchangers (Lopes, Delvivo, & Silvestre, 2005; Outinen et al., 1996; Shimamura et al., 2002). These studies give information for practical researches on the purification and separation of amino acids. Over the last years, several studies have been reported for the adsorption of amino acids on porous solids (Casado et al., 2012; El Shafei, 2002; El Shafei & Moussa, 2001; Ghosh, Badruddoza, Uddin, & Hidajat, 2011;

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Goscianska, Olejnik, & Pietrzak, 2013b, 2013c; Jiao, Fu, Shuai, & Chen, 2012; Long et al., 2009; Mei, Min, & Lü, 2009; Palit & Moulik, 2001; Silvério, Dos Reis, Tronto, & Valim, 2008; Titus, Kalkar, & Gaikar, 2003; Wu, Zhao, Nie, & Jiang, 2009). However, their adsorption capacity is still low because of the small pore volume or wide pores of these adsorbents, which are consequently inappropriate to the molecular size of amino acids. Moreover, the major drawback of the adsorption process is the high cost for the production and regeneration of adsorbents. Such inconvenient resulted in growing research on inexpensive adsorbents (Alves, Franca, & Oliveira, 2013a, 2013b; Clark, Alves, Franca, & Oliveira, 2012; Goscianska, Nowicki, & Pietrzak, 2014; He, Lin, Long, Liang, & Chen, 2015; Sebben & Pendleton, 2015).

In this study, porous activated carbon-based materials obtained from date stones (seeds) are potential adsorbents for L-phenylalanine amino acid. This is mainly due to their physical and chemical characteristics such as highly developed porous structure, good thermal stability, low cost and more accessibility. Date stones are among the most common agricultural by products available in palms growing in the Mediterranean countries like Algeria, which is one of the largest producers in the world. Algeria produces more than 400 different varieties of dates with an annual production of about 400,000 tons (Chandrasekaran & Bahkali, 2013). Date stones constitute roughly 10% of the date weight and this lignocellulosic-based agricultural waste is a good precursor for preparing activated carbon because of its excellent natural structure and low ash content (Bouchenafa-Saib, Grange, Verhasselt, Addoun, & Dubois, 2005; Merzougui & Addoun, 2008). As it is well known, two methods are commonly used for the preparation of activated carbon: physical and chemical activations. Compared with the physical process, the chemical activation presents some advantages like low activation temperature, short activation time, high surface area, well developed microporosity of activated carbon, simple operation and low energy consumption (Deng, Yang, Tao, & Dai, 2009; Pereira et al., 2014). Therefore, the date stones can be activated with chemical agents such as KOH, $ZnCl_2$, H_3PO_4, K_2CO_3 and NaOH, to obtain activated carbons with well-developed textural characteristics. To the best of our knowledge, the use of activated carbons for the L-phenylalanine recovery from aqueous solutions by adsorbents based on date stones are not available in the open literature. Thus, the principal objective of this work was to prepare porous activated carbons with high surface areas from date stones by chemical activation with KOH and $ZnCl_2$. The activated carbons proved to be good candidates for the adsorption of L-phenylalanine in an aqueous medium.

2. Materials and methods

2.1. Materials

The date stones used in this study were from Algerian origin. The following reagents were used: L-phenylalanine standard (>98%, Fluka, France), potassium hydroxide (>98%, Sigma Aldrich, USA), zinc chloride (>98%, Sigma Aldrich, USA), KH_2PO_4 (>99%, Fluka, France), K_2HPO_4 (>99%, Fluka, France), $NaHCO_3$ (>99%, Sigma Aldrich, USA), Na_2CO_3 (>99%, Sigma Aldrich, USA), HCl (37%, Sigma Aldrich, USA), KCl (>98%, Fluka, France), NaCl (>99%, Sigma Aldrich, USA), NaOH (>99%, Sigma Aldrich, USA). Ultrapure water was obtained from milli-Q system (Millipore, France).

2.2. Preparation of the activated carbons

The activated carbons were prepared from date stones. At first, the stones were thoroughly washed with distilled water and dried in an air oven at 120 °C; such protocol was effective to facilitate crushing and grinding. A fraction particle size of between 0.5 and 1 mm was used for the preparation of activated carbons by impregnation with $ZnCl_2$ and KOH. The precursor was impregnated with a chemical activating agent in a solid form. The impregnated precursor was carbonized in a horizontal tubular furnace under nitrogen flow with a heating rate of 5 °C min^{-1}, to allow free evolution of volatiles, up to the hold temperature for 1 h. The resulting activated carbon was immersed in HCl solution (0.1 mol L^{-1}) under reflux ebullition (3 h) in order to extract the compound formed and reagent excess. Then, the solution was filtered and the black solid was washed with hot distilled water until the test with $AgNO_3$ became negative. The adsorbent was dried at 120 °C, and kept in tightly closed bottles until use. The activated carbons were named ACZ (1 g $ZnCl_2$: 1 g date stones, activated at 600 °C), ACK (9 mmol KOH: 1 g date stones, activated at 800 °C).

2.3. Characterization

The specific surface area and pore structure of the activated carbons were characterized by nitrogen adsorption-desorption isotherms at −196 °C using the ASAP 2010 Micromeritics equipment. All the activated carbons were outgassed at 150 °C overnight. The specific surface area was calculated by the Brunauer–Emmett–Teller (BET) equation (Brunauer, Emmett, & Teller, 1938). The external surface area, micropore area and micropore volume were calculated by the *t*-plot method. The total pore volume was evaluated from the liquid volume of N_2 at a high relative pressure near unity 0.99 (Guo & Lua, 2000). The mesopore volume was calculated by subtracting the micropore volume from the total volume. The pore size distribution (PSD) was determined using the density functional theory (DFT) model. The morphology of activated carbons was visualized by scanning electron microscopy (SEM) using a Philips XL 30 equipped with an energy dispersive spectrometer (EDS). The Fourier transform infrared (FT-IR) spectroscopy was used to determine the functional groups of the activated carbons; the spectra were recorded over the range (400–4000 cm^{-1}) on a Perkin-Elmer spectrum two spectrometer using KBr pellets.

2.4. Determination of zero point charge pH_{PZC}

The determination of the point of zero charge (pH_{PZC}) was conducted to investigate how the surface charge of ACK and ACZ adsorbents depends on pH. pH_{PZC} of the activated carbons was determined using the procedure described elsewhere

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