



High efficient carbon dioxide capture onto as-synthesized activated carbon by chemical activation of Persian Ironwood biomass and the economic pre-feasibility study for scale-up



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ABSTRACT

The adsorption capacity of CO₂ and the economic estimation of synthesized activated carbons (ACs) from Persian Ironwood biomass were evaluated in the present study. In addition, the effect of different impregnation ratios of activation agents, including KOH and H₃PO₄, and various carbonization temperatures on the properties of synthesized ACs was examined by different analyses. The obtained results revealed that BET surface area (S_{BET}) and micropore volume of synthesized ACs were enhanced with an increase in the impregnation ratio of the precursor to KOH. Moreover, the synthesized AC by KOH activation with an impregnation ratio of 1:3 (precursor: activating agent), and a carbonization temperature of 1073 K demonstrated the higher S_{BET} (1935.8 m²/g), micropore volume percentage (about 98%), and CO₂ adsorption (5.05 mmol/g) at 1 bar and 303 K. Considering the thermodynamic parameters, the dominant mechanism for CO₂ uptake was physisorption, spontaneous, and exothermic in nature. Furthermore, based on the annual production of 900,000 kg, the estimated capital costs for AC by KOH activation and AC by H₃PO₄ activation would be \$1.65 and \$1.58 per kg, respectively.

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1. Introduction

The vast majority of emissions of greenhouse gases from anthropogenic sources are CO₂, which makes a significant contribution to global warming and climate change. Unfortunately, it is estimated that the increasing trend of atmospheric CO₂ concentration will remain unchanged in the next several decades since fossil fuels will continue to be the dominant energy source (Rashidi and Yusup, 2016). Therefore, considering the continuous CO₂ emission from fossil fuels, the unforeseen damages of natural ecosystems could be probable. Among technologies suggested for the reduction of CO₂ emissions, adsorption is regarded as a very promising process for CO₂ capture (Cuéllar-Franca and Azapagic, 2015), because of its low energy requirement, cost effectiveness and simplicity of use at a wide variety of temperatures and pressures (Heidari et al., 2014a). Numerous adsorbents have been investigated for CO₂ adsorption including zeolites, carbon

nanotube, activated carbons, silicas, hydrotalcites, metal oxides, MOFs, etc. (Rashidi and Yusup, 2016).

Activated carbon is known as an excellent adsorbent for a variety of environmental applications, i.e. purification and storage of gases and the removal of organic materials and metals from aqueous solution (Kazemi et al., 2016). This is due to some of the unique characteristics of AC, such as the large surface area and micropore volume, the appropriate pore size distribution, the surface chemistry and, the degree of polarity. However, the broad application of this novel adsorbent to control the environmental pollution has been restricted by the final cost of AC production. As a result, a large-scale effort has been conducted to produce high-quality AC from the low-cost precursors with favorable properties to use in various potential applications. Accordingly, close attention has been paid to synthesis of AC from waste material thanks to the unlimited availability of sources for scale-up production and for eco-environmental reasons (Choi et al., 2009; Wang et al., 2011). Considering this point, the different lignocellulosic wastes have been applied to produce low-cost ACs so far (Laksaci et al., 2017). Therefore, in the current study, the final capital cost of AC production is estimated by the application of process flow diagrams for large-scale production. The obtained results can be utilized to

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| Nomenclature | | | |
|------------------------------------|--|-------------------|--|
| AC | Activated Carbon | DTA | Differential thermal analysis |
| CAC | Commercial Activated Carbon | R | Gas constant (8.314 J/K·mol) |
| KH-T-X | Activating agent (KOH)-Activating temperature (T)-Activating ratio (X) | T | Temperature (°C or K) |
| HP-T-X | Activating agent (H ₃ PO ₄)-Activating temperature (T)-Activating ratio (X) | P | Equilibrium pressure (bar) |
| BET | Brunauer–Emmett–Teller | P _s | Equilibrium pressure of the adsorbate at standard pressure (bar) |
| S _{BET} | Specific surface area determined by BET method (m ² /g) | q | Adsorption capacity (mmol/g) |
| BJH method | Barrett–Joyner–Halenda method | q _m | Maximum adsorption capacity (mmol/g) |
| MP method | Micropore analysis method | b | Langmuir constant (1/bar) |
| V _{tot} | Total pore volume (cm ³ /g) | R _L | Dimensionless constant separation factor |
| V _{mic} | Micropore volume (cm ³ /g) | K _F | Freundlich constant [(mmol/g) (1/bar) ^(1/n)] |
| V _{mic} /V _{tot} | Micropore volume percentage (%) | N | Freundlich Exponent (dimensionless) |
| R _{mic} | Average micropore radius (nm) | k _T | Temkin isotherm equilibrium binding constant (1/bar) |
| ATR-FTIR | Attenuated Total Reflection-Fourier Transform Infrared spectroscopy | b _T | Temkin isotherm constant (dimensionless) |
| TEM | Transmission Electron Microscopy | Q | Temkin constant related to heat of sorption (J/mol) |
| TGA | Thermal Gravimetric Analysis | K _b | BET constant |
| | | ΔH _{ads} | Isosteric heat of adsorption (kJ/mol) |
| | | ΔS | Entropy changes (J/K·mol) |
| | | ΔG | Gibbs free energy (kJ/mol) |
| | | R ² | Squared correlation coefficient |

assess the fixed capital investment required to construct a manufacturing facility.

Persian ironwood (*Parrotia persica*) biomass is an abundant type of lignocellulosic material found in the moist forests. It constitutes 5.4% of the standing volume of Hyrcanian forest in the north of Iran. Moreover, the area covered by this species has remarkably increased over the past two decades and its biomass carbon density is moderately high. Mainly due to the low commercial value of its wood, no industrial harvesting is carried out in this forest area (Sagheb-Talebi et al., 2014). On the other hand, due to its physical features, such as high density, high cellulose content taking into consideration no industrial exploitation, ease of access, and cost-effectiveness, Persian ironwood biomass can be considered as an appropriate source to produce high quality AC for different environmental applications. By the application of this precursor, the feasibility of production of AC from a cheap and abundant waste can be possible. However, there are fundamentally two methods for preparing activated carbon: physical and chemical activation (Ello et al., 2013). Ordinarily, physical activation is accomplished using carbon dioxide, steam, and air, or a mixture of the three (Baklanova et al., 2003). Chemical activation is carried out by the application of various activating agents such as zinc chloride, acids, and alkali. Among the abundant chemical activating agents, H₃PO₄ and KOH are widely used and preferred due to their significant role in the development of the surface area and porosity; but the use of ZnCl₂ has reduced in the past few years because of its harmful environmental impacts (El-Hendawy et al., 2008). To the author's knowledge, there is no publication about the application of Persian Ironwood biomass as a precursor for AC production.

The main objective of the present study was to prepare AC from Persian Ironwood biomass, including fallen foliage, dried trunks etc. However, alongside the potential of the CO₂ capture technologies by the AC-derived biomass, the estimation of capital costs of the final production was evaluated. In order to achieve this, the optimum temperatures for the synthesis of ACs with the application of H₃PO₄ and KOH were determined. Then, the effect of different impregnation ratios of KOH and H₃PO₄, and the influence of different temperatures onto a CO₂ adsorption capacity by the prepared ACs were evaluated. Finally, the capital costs for the large

production of synthesis ACs were estimated from a combination of vendor quotes and printed literature.

2. Material and methods

2.1. Preparation of AC

Synthesis of ACs was carried out in the two-step carbonization process. The pyrolysis experiments were performed in a stationary horizontal stainless steel tube furnace. The detailed pyrolysis experiment is described in supporting information (SI). Afterwards, the prepared ACs were labeled KH-t-x or HP-t-x, where KH or HP denotes the activating agents, t denotes the activating temperature, and x denotes the activating ratio. Finally, the Langmuir, Freundlich and Temkin isotherm models were applied in order to compare the experimental data with theoretical models as well as thermodynamic parameters of the adsorption process. The materials used in the experiments, detailed characterization of the ACs, description of the adsorption isotherms and the adsorption thermodynamics are given in SI.

2.2. CO₂ adsorption measurement

In order to investigate the behavior of AC samples in CO₂ capture, gas adsorption determination was carried out using the static volumetric method in different pressures ranging from 1 to 25 bar and in different temperatures (288–303 K) by a home-made gas adsorption apparatus presented in Fig. 1. Also, 10 successive adsorption/desorption cycles using temperature-vacuum regeneration operation was performed at 523 K for 2 h to assess the regenerability potential of the sorbents. The full experimental procedure of CO₂ adsorption is described in SI. In summary, in this apparatus, two precision pressure transducers with high accuracy were used to determine the pressure alterations in the gas phase and the adsorption cell in each CO₂ adsorption test. Before the CO₂ adsorption test, the sample was outgassed for 2 h at 523 K, and a vacuum pump (Rocker 420, China) was used to evacuate the vessel filled with the sample. Ultimately, the quantity of CO₂ adsorption by samples was evaluated by the Soave-Redlich-Kwong (SRK)

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