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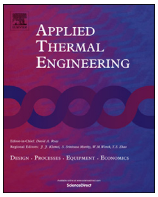

Adsorption kinetics of propane on energetically heterogeneous activated carbon

Azhar Bin Ismail [a], Kyaw Thu [b], Kandadai Srinivasan [b], Kim Choon Ng [a, b, *]

[a] *Water Desalination and Reuse Center, 4700 King Abdullah University of Science and Technology, Thuwal 23955-6900, Saudi Arabia*
[b] *Department of Mechanical Engineering, National University of Singapore, 9 Engineering Drive 1, 117576 Singapore, Singapore*

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ABSTRACT

The modeling of the adsorption isotherms and kinetics of the adsorbent + adsorbate pair is essential in simulating the performance of a pressurized adsorption chiller. In this work, the adsorption kinetics is analyzed from data measured using a magnetic suspension balance. The Statistical Rate Theory describes the Dubinin–Astakhov (DA) equation and extended to obtain an expression for transient analysis. Hence both the experimental excess equilibria data and the adsorption kinetics data may then be fitted to obtain the necessary parameters to fit the curves. The results fit the data very well within 6% of the error of regression.

1. Introduction

Interest in adsorption (AD) refrigeration has grown due to its advantages related to its direct utilization of thermal energy sources such as low grade waste heat from various industrial sources, solar hot water as well as geothermal sources. As a result, there has been a diversified approach in the study of adsorption with a goal to explore various applications of the thermal heat pump system [1,2]. In this work, the authors report the kinetics of adsorption of propane at various temperatures and pressures. They are presented as an ongoing study of utilizing alternative refrigerants as adsorbate in an AD system.

Ismail et al. (2014) [3] has previously elaborated how the highly porous activated carbon Maxsorb III + propane gas pair is favorable amongst tested pressurized adsorption pairs when low temperature-cooling is required or operated in localities where the ambient temperature is relatively high (above 40 °C). This is because, as illustrated using the experimental isotherms [4], the steady state simulation suggests that the cooling capacity (SCE) of an adsorption chiller declines with an increasing ambient temperature and decreasing evaporator temperatures until it is deemed inoperable. Under these adverse conditions in arid localities, a pressurized adsorption chiller which utilizes propane as a refrigerant with activated carbon as the adsorbent offers a best selection amongst the tested refrigerant pairs for the utilization of low grade waste heat in continuous batch-operated cooling [3]. The properties of this adsorption pair had been acquired experimentally and its theoretical thermodynamic framework developed and presented in previous works [3,4]. The kinetics of Maxsorb III + propane pair is first measured using a thermal-gravimetric approach by utilizing a magnetic suspension balance. The Statistical Rate Theory model of Rudzinski et al. which has previously been extended to describe the kinetics process of heterogeneous systems [5], is used to mathematically describe the adsorption rate of propane on Maxsorb III.

2. Materials and methods

A magnetic suspension balance (Rubotherm) is used to measure the instantaneous uptake of the adsorbent as shown in Fig. 1. This balance measures the weight of the sample with a reproducibility of ±0.03 mg [6]. The advantages of utilizing such a suspension balance are its high accuracy as well as stability. This is because of the placement of the suspension balance outside the measuring chamber and it thus does not have any interactions with the refrigerant. This system also allows for continuous logging of data. Maxsorb III activated carbon (by Kansai Coke Company, Japan) is utilized with pure propane as the adsorbate at a purity of 99.5%. The values of derived quantities of Nitrogen is taken from Wagner (2000) [7], while that of helium from Ortiz-Vega et al. (2010) [8]

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* Corresponding author. Department of Mechanical Engineering, National University of Singapore, 9 Engineering Drive 1, Singapore 117576, Singapore. Tel.: +65 65162214.

E-mail addresses: azhar.binismail@kaust.edu.sa (A.B. Ismail), mpekyaw@nus.edu.sg (K. Thu), mecks@gmail.com (K. Srinivasan), mpengkc@nus.edu.sg (K.C. Ng).

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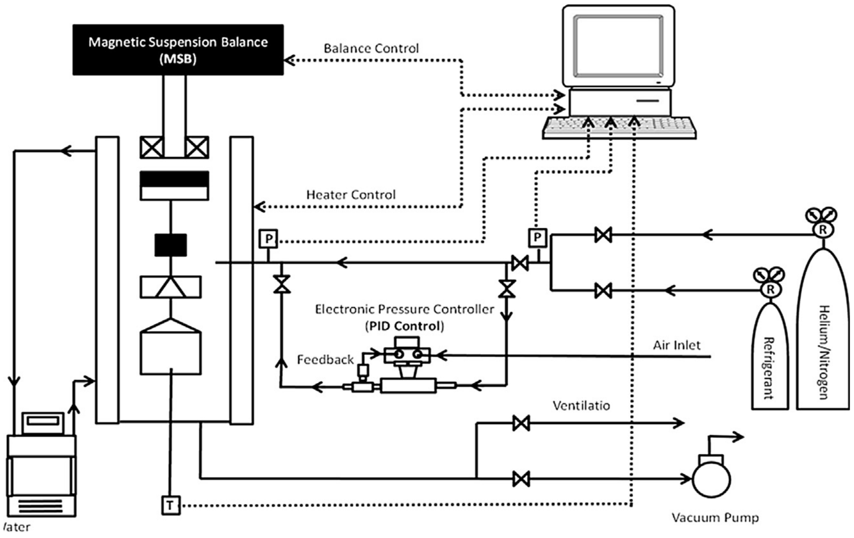

Fig. 1. Schematics diagram for the magnetic suspension balance unit (Rubotherm).

and the thermodynamic properties of pure propane from Lemmon et al. (2009) [9].

In the uptake measurements, the buoyancy effects resulted from gas density variances needs to be eliminated. The volumetric displacements of the propane gas by the container, activated carbon, and the adsorbed phase propane are taken into account. The correction related to the adsorbent container is calculated by performing blank experiments at different densities (ρ (P,T)) with the empty container. The buoyancy caused by the solid matrix of the activated carbon, which caused a mass reduction from the measurements, is estimated using the Archimedes' principle which is the product of the skeletal volume of the adsorbent and the gas density. Finally, the buoyancy effect exerted on the adsorbed phase is corrected to obtain the absolute adsorption uptake q (P,T).

The weight, m, displayed by the balance is an addition of various contributions which summing up to the net force exerted on the sample:

$$m = m_h\left(1 - \rho_g/\rho_h\right) + m_s\left[1 - \rho_g/\rho_s + q\left(1 - \rho_g/\rho_a\right)\right] \tag{1}$$

Here, m_h and ρ_h are the mass and density of the container m_s and ρ_s are the mass and density of the activated carbon sample while ρ_g and ρ_a are the densities of the refrigerant at the gas and adsorbed phases at equilibrium.

The blank experiments with an empty holder give the mass and density of the holder from the intercept and the slope. The graph of apparent weight against gas density is expected to be a linear decrease as shown in the following equation:

$$m = m_h - \frac{m_g}{\rho_h}\rho_g \tag{2}$$

The density (ρ_g) of nitrogen gas is obtained using the equations of Span et al. [7]. Adsorption experiments using helium which is assumed to be a non-adsorbing gas since it is carried out at a high temperature of 120 °C, provide the mass (m_s) and density (ρ_s) of the solid matrix of the activated carbon sample by utilizing Eq. (3):

$$m - m_h\left(1 - \rho_g/\rho_h\right) = m_s - m_s/\rho_s \times \rho_g \tag{3}$$

In Eq. (3) it is assumed that helium is inert, taking up the volumes of all available pores of the carbon i.e., without being adsorbed. Finally, the experiments of propane gas with the activated carbon provide the uptake q from Eq. (4):

$$m_s q\left(1 - \rho_g/\rho_a\right) = m - m_h\left(1 - \rho_g/\rho_h\right) - m_s\left(1 - \rho_g/\rho_s\right) \tag{4}$$

The value of ρ_a is estimated using the approximation by Ozawa (1976) [10], given by Eq. (5) as follows

$$\rho_a = m\frac{\rho_a^*}{\exp[\alpha_e(T - T_b)]} \tag{5}$$

In Eq. (5), ρ_a^* is the density of the refrigerant in the liquid state at its normal boiling point T_b. α_e on the other hand is its thermal expansion. The pressure dependencies of ρ_a, ρ_a^* and α_e are regarded to be small in the pressure ranges of the present work and are thus not considered. The normal boiling point of propane and its liquid density at the normal boiling point are taken from Lemmon et al. (2000) [9]. Further, the thermal expansion was assumed to be independent of the adsorbate species, and the mean value of gases ($\alpha_e = 2.5 \times 10^{-3}$ K^{-1}) was used in the numerical calculation. The utilization of this equation accounts for the adsorbed phase volume correction has successfully been used in previous works resulting in an improved fit [11,12] with relative ease.

A pressure controller ER3000 is used to set the chamber pressure, and water from the water bath enters the jacket to maintain the desired temperature. When equilibrium is reached, the valve is opened to allow the propane gas to enter the chamber, and the pressure, temperature and weight changes are logged in the data logger.

3. Supporting theory

In this paper, the kinetics of propane on activated carbon Maxsorb III is described using the statistical theory. The Statistical Rate Theory assumes a Gaussian distribution for the distribution of adsorption sites and it successfully derives the well-known Dubinin–Astakhov (DA) equation [13] which is as follows

$$\frac{q}{q_o} = \exp\left[-\left(\frac{kT}{E}\ln\frac{p^o}{P}\right)^r\right] \tag{6}$$