



Total and partial uptakes of multicomponent vapor-gas mixtures on 13X zeolite at 343K: Experimental and modeling study



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ABSTRACT

In this work, GERG2008 EoS embedded in volumetric-gravimetric system was developed to allocate multicomponent partial/individual components uptakes in the mixture. The refined arrangement may overlay the current theoretical anticipated outcomes and interchange it with experimental and more trustworthy selective adsorption outcomes. 13X zeolite was utilized as a solid adsorbent for binary and ternary CO₂:CH₄:H₂O mixtures adsorption. Premixed and preloaded water vapor was studied at 343 K and up to 10 bar. Artificial neural network (ANN) modeling was engaged to predict binary and ternary mixtures. ANN results disclosed decent promise with experimental data. Besides, simulated formations utilizing ANN model replicated high consistency. The testified outcomes magnificently identified particular components behavior in ternary and higher multicomponent mixtures.

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1. Introduction

Natural gas has emerged as one of the cleanest primary sources of energy in the world. The increasing global energy demand is responsible for the depletion of natural gas reserves. The exploration of untapped gas reserves with high CO₂ content is unavoidable due to the increased worldwide consumption. According to Darman and Harun [1], the composition of natural gas reserves takes another trend in some Asian pacific region, as the CO₂ content ranges in some fields from 30 to 90% at high CO₂ content reserve region. The discovery of natural gas field with the CO₂ content of as high as 90% in Malaysia postures new challenges linked to the separation processes of CO₂. The impact of separation and purification process take place, as the main pretreatment stage need for natural gas before considered as sales gas [2,3].

This leads to the multicomponent mixtures separation and purification to deliberate significant issue in the industrial field. This emphasis more concerns to observe more sufficient and developed selective separation techniques. Adsorption process via solid adsorbents is contributed as one of the most potential alternative technique for multi-components mixtures separation and

purification process. The necessity to identify the equilibrium data that emphasize a wide range of operational conditions is crucial to determine the optimized design for adsorption systems and ultimate solid adsorbents [4–7].

In the case of pure components, the data can easily be measured. However, in multicomponent systems, the measurement is usually complicated and required further calculations. Therefore, many theoretical models were developed to predict the behavior of binary and ternary mixtures, relying on pure component adsorption data which have various drawbacks and limitations in terms of accuracy, ideality and intensity. Some examples of theoretical models e.g. ideal adsorption solution theory (IAST), real adsorption solution theory (RAST), heterogeneous ideal adsorption solution theory (HIAST), grand canonical Monte Carlo (GCMC) simulation were utilized to predict the behavior of binary and higher multicomponent mixtures. However, most of these models have limitations when it is related to the water-gas multicomponent mixtures prediction [8–10]. There are no satisfactory experimental data on ternary multicomponent mixtures [11], and a minor amendment of the adsorbent commonly demands a modification of parameters in the existing adsorption models.

However, most of the modeling techniques reported in the literature did not address the influence of adsorption parameters on partial/individual components uptakes in the H₂O-CO₂-CH₄ ternary mixtures. Also, there are limited studies available on the

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partial components uptakes that emphasize the influence of adsorption parameters such as temperature, pressure, and initial mixtures compositions, especially in the presence of water-gas combinations [12–14]. Likely, all the efforts did not finalize a feasible solution, model or experimental approach that can accurately describe ternary mixtures for their partial/individual components adsorption isotherm during separation process.

Several equations of states (EoSs) were developed and applied to demonstrate binary and ternary mixtures behavior, the choice of suitable EoSs rely on the importance of system investigated conditions and parameters i.e. temperature, pressure, and mixture composition. Hence, non-ideality of mixtures played an important role in EoS utilization and validation [15]. The GERG as a wide-range equation of state for natural gases and other mixtures was developed by Kunz et al. [16]. They were claimed GERG2008 as the most reliable equation of state that is applicable for practically all of the fundamental sectors related to vapor-liquid equilibrium, multi-component phase separations and descriptions. Most recently, reliable computational prediction methods such as artificial neural network (ANN) were concerned with excessive attraction in the prediction of density, surface tension, and viscosity of components and studied mixtures [17–19]. Hajir and Fakhri [20] studied the vapor liquid equilibrium of binary mixtures utilizing back propagation artificial neural network (BP-ANN) model for the simultaneous estimation of vapor liquid equilibrium (VLE) of four binary systems. Fatemeh et al. [11] studied binary mixtures prediction from data reported in the literature utilizing (2-D EoS) i.e. Redlich–Kwong (RK), Soave–Redlich–Kwong (SRK), Peng–Robinson (PR), and Modified Mohsenia–Modarress–Mansoori (M4), compared with the data predicted utilizing ANN predictions. However, they claimed that the binary mixtures predicted utilizing ANN model showed better agreement and precision compared to the EoSs mentioned and studied.

For separation and purification of multicomponent mixtures molecular sieve zeolites (MSZ) adsorbents were deliberated as one of the most potential solid adsorbents. According to the low Si/Al ratios MSZ which usually showed higher adsorption capacity of CO₂ at a wide range of operational conditions over CH₄. This might refer to the presence of extra-framework alkali cations (e.g. Li⁺, Na⁺, K⁺, Cs⁺), that increase the adsorption capacity of CO₂ via electrostatic (charge-quadrupole) interactions [21]. On the other hand, zeolite performance related to CO₂ adsorption were reported to be drastically declined in the water-gas systems, which needs further challenges outlook studies [22,23]. The effect of SiO₂/AlO₂ Atomic Ratio has high intention in the field of multicomponent mixtures selective separation, which related to the vapor–solid interaction on ionic solid adsorbents. This might be affected by the hydrophobicity of the solid adsorbents and in conclusion negatively affect the adsorption capacity and selectivity. Abhimanyu et al. [24] reported that the adhesion forces are an exponential function of macro scale water contact and surface energy due to strong liquid-solid or vapor-solid interaction elaborated in capillary forces. These interactions are subjected by electrostatic-dipole-dipole interaction (hydrogen bonding) on the surface and/or few atomic water layers above the surface. Kumar and Chowdhury [25] were reported that the relatively high selectivity for nitriles on H-USY samples with a high SiO₂/Al₂O₃ ratio might be related to their hydrophobicity, as the hydrophobic H-USY samples could not retain sufficient H₂O. Ye et al. [26] reported that zeolites as a significant material in the application of catalysis, absorption and ion-exchanging, according to their micro porous structure. The most commonly used zeolite is FAU type adsorbents, the framework of which is composed of AlO₄ and SiO₄ tetrahedra. It can also be viewed as SiO₂ with some SiO₄ tetrahedron substituted by AlO₄, resulting in electronegativity of the framework. It can be balanced by the cations attaching to the

framework in the pores, such as Na⁺, K⁺, Mg²⁺, Ca²⁺, NH₄⁺, etc. These cations are always easy to be exchanged with external ions. And the number and sites of the cations strongly affect the properties of the zeolites.

Therefore, in this study, 13X MSZ is considered as the potential solid adsorbent which has the high adsorption capacity compared to silica gel for pure CO₂ and H₂O, which can be easily benchmarked and linked to other aforementioned studies. This can correlate the outcomes of the proposed approach with earlier reported data, and to differentiate adsorption capacity and selectivity for individual components within the mixture with theoretically predicted and pure adsorption outputs.

In this work, a thermal EoS namely GERG 2008 was embedded in the volumetric-gravimetric adsorption experimental setup customized RUBOTHERM controlling c-RCS software, to measure total and partial/individual components uptakes in the multicomponent binary and ternary i.e. CO₂-CH₄-H₂O mixtures at various combinations. The experimental data were correlated and applied to ANN and two non-ideal models i.e. Extended Langmuir and modified Extended Langmuir to predict the mixtures and simulated further configurations. This can deliberate ease comparison and data correlation with the current potential multicomponent analysis models.

2. Experimental setup

The adsorption measurements were performed in the custom designed volume-gravimetric system. The Gas Adsorption Separation Unit (GSAU) was proposed and requested by RCCO₂C and custom made, commissioned, delivered, and assembled by Rubotherm GmbH., Germany. The setup consists of three main sections: The first section is the dosing system which was utilized to generate and dose gases and vapors either into the balance directly (for pure gases and vapors measurements) or into the mixing chamber (for multi-component measurements). The second section is a closed loop mixing chamber, while the last section consists of a high accuracy magnetic suspension balance (MSB), which measures the change in mass and density of the sample of material and gases, respectively, with a precision of 2×10^{-6} g.

The system was designed to perform multicomponent mixtures analysis at a wide range of operational conditions. Experimental operational temperatures can be conducted from -20 °C up to 150 °C and pressures from vacuum range up to 150 bar. Pretreatment and regeneration of samples can be performed in vacuum range up to 400 °C. Pretreatment can also perform at purged He/N₂ environment conditions for adsorbents according to the adsorbents pretreatment characteristics.

Fig. 1 demonstrates the main parts and sections of the experimental setup. The dosing system is responsible for gases and vapor dosing to the mixing chamber, while the mixing chamber/unit (MC) is responsible for mixing the adsorbates and created a homogeneous mixture before expanded to the reaction chamber/cell (RC). The RC consisted of magnetic suspension balance, which was connected to the custom made Danni-Master Gas Chromatograph (GC). The role of the GC is to analyze the inlet and outlet mixtures before and after expansion of adsorbates combinations to the reaction chamber. The utilized GC has capability to analyze the composition of wide range of gases, hydrocarbons and vapors mixtures as detailed in the previous work [27].

The measurements of the partial uptakes of the components within the mixture, need to be determined. The mass adsorbed of each gas required to be identified and measured at the early dosing stage before and after expansion to the reaction chamber. The pressures, temperatures, initial and final compositions, and the amount adsorbed for the total mixture of the specific segment were

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