



Vapor pressure measurements of ethanol–isooctane and 1-butanol–isooctane systems using a new ebulliometer

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HIGHLIGHTS

- ▶ A new simple ebulliometer has been developed in this work.
- ▶ The ebulliometer is reliable to measure vapor pressure of alcohols–isooctane mixture.
- ▶ The experimental data were well correlated with Wilson, NRTL, and UNIQUAC models.

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ABSTRACT

A new, simpler ebulliometer has been developed to accurately measure the vapor pressure of ethanol–isooctane and 1-butanol–isooctane systems at various mixture compositions. The reliability of the experimental apparatus was validated by comparing the experimental data with published data for pure isooctane and ethanol and ethanol–isooctane system. Our data were in good agreement with the published data, with a deviation of less than 1.9%. The vapor pressure of an ethanol–isooctane mixture was initially elevated by the addition of ethanol until the ethanol concentration reached 0.3 mass fractions, then dropped at concentrations above 0.7 mass fractions. For a 1-butanol–isooctane system, the vapor pressure decreased with increasing 1-butanol concentrations. The experimental data were correlated using the Wilson, Non-Random Two-Liquid (NRTL), and Universal Quasi-Chemical (UNIQUAC) activity coefficient models with an average absolute deviation in the vapor pressures of 3.5%, 3.3%, and 6.1%, respectively.

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1. Introduction

Fossil fuels, which remain the primary energy resources around the world, cause significant increases in the atmospheric CO₂ concentration and have an effect on global warming [1,2]. Fossil fuels combustion contributes 73% of CO₂ production [3]. Thus, finding a solution to greenhouse gas emissions is a major research focus for solutions to the global warming problem.

Recently, the addition of oxygenates, such as simple alcohols, that are primarily produced from renewable resources has received tremendous attention because these compounds have an ability to reduce pollution and enhance the octane number of gasoline. Nicholas Otto [4] first used ethanol in internal combustion engines. Ethanol–gasoline mixtures have been successfully used in various machines and vehicles that typically run on gasoline [5]. However, the mixture can have a higher vapor pressure than the base gasoline which raises concern regarding the evaporative emission from the fuel mixture. Vapor pressure is one of the most important

physical properties of a gasoline mixture because it refers to the volatility of the gasoline and can affect the evaporative emission of the fuel mixture. Therefore, accurate experimental data are important in designing fuel mixtures with specific vapor pressures. The vapor pressures of gasoline mixtures with four alcohols at 37.8 °C (100 °F) of various compositions were measured by Pumphrey et al. [6]. Sun et al. [7] measured the bubble-point vapor pressures for mixtures of an endothermic hydrocarbon fuel with ethanol. The experimental results indicated that the addition of ethanol had a critical effect on the vapor pressure of the fuel. An ethanol–fuel blend with a low ethanol composition can have an increased volatility.

The vapor pressures for both pure substances and mixtures are traditionally measured using an ebulliometer, which was first proposed by Cotrell [8] to determine the boiling points of pure substances. Rogalski and Malanowski [9] developed a new ebulliometer in 1980 using Cotrell's principle for determining vapor–liquid equilibrium data. However, in these ebulliometers analyses of both vapor and liquid-phase compositions are necessary due to a difference between the liquid-phase composition at equilibrium and the feed composition. Therefore, Li et al. [10]

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developed an inclined ebulliometer that no longer required liquid and vapor-phase composition analysis to measure the bubble point for a chloroform–ethanol–benzene system. Li et al. [10] also noted that they adopted the angle of inclination of 30° for their work to ensure the accuracy of the equilibrium temperature measurement.

In this work, a new, simpler ebulliometer is proposed based on the work of Li et al. [10] by removing the inclination and enlarging the volume of the cell. We can ensure that the equilibrium and the feed compositions do not change significantly during the measurements due to the small proportion of vapor formed during equilibrium by enlarging the volume of the cell. Therefore, the present apparatus can be used to accurately measure the vapor pressure of a gasoline mixture. We used isooctane in this work to represent gasoline because it is the main component in gasoline.

2. Experimental

2.1. Materials

All materials used in this work (isooctane, ethanol, and 1-butanol) were supplied by Merck, Darmstadt, Germany. These materials were used without further purification. Table 1 lists the properties of all materials used.

2.2. Experimental apparatus

A new, simpler ebulliometer based on the principle of the quasi-static method is shown in Fig. 1b. The ebulliometer has a 5.5-cm cell diameter, 14-cm height, and a 332.5-cm³ volume and is connected with a vacuum pump. The ebulliometer cell was turned on via moderate heating, which was controlled using a temperature controller to maintain equilibrium at the desired temperature.

The quasi-static method is based on the overall concentration instead of the equilibrium composition of the liquid phase as described by Li et al. [10] and shown in the inclined region in Fig. 1a. The enlargement of the cell volume as shown in Fig. 1b allows for an increase in the amount of substance being tested so that the composition of the fluid hardly changes during the vapor pressure measurement. This can be calculated using the simple mass balance in Eq. (1) as follows:

$$z_i F = x_i L + y_i V \quad (1)$$

In which, z_i is the mole fraction of component i in the initial mixture injected into the ebulliometer, F the total moles of mixture injected into the ebulliometer, x_i the mole fraction of component i in the liquid phase at equilibrium, L the number of moles in the liquid phase at equilibrium, y_i the mole fraction of component i in the vapor phase at equilibrium, V the number of moles in the vapor phase at equilibrium.

We assume, with the same value of V , the same mole fraction for component i in the vapor phase (y_i), and the same feed composition (z_i), that a smaller ebulliometer means that the amount of mixture injected into the ebulliometer (F) must be smaller, and significant change can occur in the mole fraction of component i in liquid phase at equilibrium (x_i). In our work, we developed a new,

simpler ebulliometer by removing the inclined portion from the previous ebulliometer. In addition, a temperature detector was attached at the liquid–vapor interface to ensure the accuracy of equilibrium temperature measurements.

The mixtures with varying isooctane and ethanol, isooctane and 1-butanol compositions were prepared by directly weighing the constituent component on an Ohaus balance with a precision of ± 0.0001 g. The temperature was measured using an RTD Pt 100 sensor connected to a Shimaden SD 15 temperature display with a measurement accuracy of ± 0.1 K. The vapor pressures at various temperatures and compositions were measured using a mercury manometer with an accuracy of ± 0.1 mm Hg.

2.3. Experimental procedure

A sample containing approximately 220 cm³ of a liquid mixture of known composition was charged in the ebulliometer cell. First, the system was created under vacuum conditions to remove air and impurities from the equilibrium cell. Then, the equilibrium cell was heated to reach the desired equilibrium temperature. The temperature was controlled by a PID controller (Shimaden SR 64). After the ebulliometer reached a constant pressure at a desired equilibrium temperature, the pressure was recorded as the vapor pressure at a desired temperature. The experimental procedure was repeated at different temperatures and compositions.

3. Results and discussion

3.1. Reliability test of the apparatus

The proposed ebulliometer was tested for the reliability by comparing the experimental data for the vapor pressures of both pure liquids and mixtures as measured by this apparatus with the published data for pure ethanol, isooctane, and an ethanol–isooctane mixture. The Antoine equation [11] for pure isooctane and the Wagner equation [12] for pure ethanol were used to compare the experimental data. The Antoine and Wagner constants were obtained from Poling et al. [13]. For the ethanol–isooctane mixture, the experimental data were compared with the experimental data from Hull et al. [14].

A comparison of the experimental data obtained in this work with the published data is presented in Tables 2 and 3 and Figs. 2 and 3 along with their deviations. The results indicate that the data were in good agreement with the published data, with a deviation of less than 1.9% calculated as an average absolute deviation (AAD) in the vapor pressures as follows:

$$\text{AAD} = \frac{1}{n} \sum_{i=1}^n \left| \frac{P_{lit} - P_{exp}}{P_{exp}} \times 100\% \right|_i \quad (2)$$

in which P_{exp} is the vapor pressure obtained in the experiment while P_{lit} is the vapor pressure calculated using both the Antoine [11] and Wagner [12] equations and n is the number of data.

Table 1
Properties of materials used in this work.

Property	1-butanol CH ₃ (CH ₂) ₃ OH	Isooctane C ₈ H ₁₈	Ethanol C ₂ H ₅ OH
Molecular weight (g/mol)	74.12	114.23	46.07
Purity	>99.5%	>99.5%	99.9%
Density (d 20 °C/20 °C)	0.809–0.812	0.691–0.696	0.790–0.793
Boiling range (°C)	116–119	98–100	78

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