



Prediction of ternary azeotropic refrigerants with a simple method

Yanxing Zhao ^{a, b}, Maoqiong Gong ^{a,*}, Xueqiang Dong ^{a,**}, Haiyang Zhang ^{a, b}, Hao Guo ^a, Jianfeng Wu ^a

^a Key Laboratory of Cryogenics, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, P. O. Box 2711, Beijing 100190, China

^b University of Chinese Academy of Sciences, Beijing 100039, China



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ABSTRACT

Refrigeration systems with azeotropic mixtures can achieve lower energy consumption, higher refrigeration capacity and coefficient of performance than both individual fluids and zeotropic refrigerants. In this paper, a simple method for predicting homogenous ternary azeotropic refrigerants was presented. The Peng-Robinson equation of state combined the Van der Waals mixing rule was proved successfully to represent the vapor + liquid equilibrium behavior of binary system and was employed to describe the ternary mixture phase equilibrium property. One hundred and seventy-one ternary systems were tested and eight azeotropes with six saddle-point azeotropes and two maximum-point azeotropes were found. The Antoine equation was used to correlate the azeotropic pressures and the temperatures, which showed the similar behavior to pure fluids and revealed the reliability of the calculated value. It can be concluded that to form ternary azeotropes in refrigerant mixtures at least two subsystems are azeotropic and if all subsystems are azeotropic, the ternary system is more likely to form ternary azeotrope.

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1. Introduction

Since the halogenated refrigerants are restrictedly used in the vapor compression refrigeration system for its high ozone depleting potential, it is necessary to look for long-term alternatives to satisfy the objectives of international protocols. However, the single refrigerants including fluorohydrocarbons, hydrocarbons and other natural refrigerants have all kind of drawbacks such as inflammability (HCs), incompatibility with the mineral lubricating oils (HFCs), toxicity (ammonia), etc. These limitations shifted the focus on the mixed refrigerant alternatives, which are obtained as a mixture of two or more components. Both zeotropic and azeotropic mixtures have been investigated. Due to the mass transfer resistance in nucleate boiling and high temperature glide, heat transfer coefficients of zeotropic mixtures are normally lower than the single refrigerants and the azeotropic mixtures [1]. Further, refrigeration systems with azeotropic mixtures can achieve lower energy consumption, higher refrigeration capacity and coefficient of performance than that with both individual and zeotropic

refrigerants [2–7]. Besides, zeotropic refrigerant mixtures have a fractionation problem caused by a leak in the system, while azeotropic mixtures can overcome this difficulty [8].

The phase equilibrium properties of the mixed refrigerants can be decided by experimental or theoretical methods. Accurate phase equilibrium data can be obtained experimentally but it is too time consuming, especially for azeotropes. To solve this problem, many prediction methods have been developed. Wang and Whiting [9] designed an algorithm for azeotropic prediction by treating vapor liquid equilibrium for the azeotropic point of a mixture as similar to that for a pure compound. Their algorithm was useful for rapid detection of azeotropes and did not search in regions where vapor and liquid compositions are not identical, with which the problem of spurious roots was avoided. Harding [10] described a homotopy method which, together with an arc length continuation, gave an efficient and robust scheme for computing azeotropes in multi-component mixtures. Artemenko and Mazur [11] developed an approach for the prediction of azeotrope formation in a mixture that does not require vapor liquid equilibrium calculations. The method employs neural networks and global phase diagram methodologies to correlate azeotropic data for binary mixtures based only on critical properties and acentric factor of the individual components in refrigerant mixtures. Fedali et al. [12] predicted the azeotropic behavior of the mixtures using the mole

* Corresponding author.

** Corresponding author.

E-mail addresses: gongmq@mail.ipc.ac.cn (M. Gong), dxq@mail.ipc.ac.cn (X. Dong).

Nomenclature

Abbreviations

AAD	The average absolute deviation
AARD	The average absolute relative deviation
AZ	azeotropic
c ₁ , c ₂	PR EoS parameter
EoS	equation of state
HC	hydrocarbon
HFC	fluorohydrocarbon
MIX	mixture
BAS	binary azeotropic subsystems
PR	Peng-Robinson
R170	ethane
R290	propane
R600	n-butane
R600a	isobutane
R23	trifluoromethane
R32	Difluoromethane
R116	hexafluoroethane
R125	pentafluoroethane
R134	1, 1, 2, 2-tetrafluoroethane
R134a	1, 1, 1, 2-tetrafluoroethane
R143a	1, 1, 1-trifluoroethane
R152a	1, 1-difluoroethane
R161	fluoroethane
RE170	methoxyethane
R227ea	1, 1, 1, 2, 3, 3, 3-heptafluoropropane
R236ea	1, 1, 1, 2, 3, 3-hexafluoropropane
R236fa	1, 1, 1, 3, 3, 3-hexafluoropropane
R245fa	1, 1, 1, 3, 3-pentafluoropropane
R1234ze(E)	trans-1, 3, 3, 3-tetrafluoropropene
R1234yf	2, 3, 3, 3-tetrafluoroprop-1-ene
R1270	propylene
RC270	cyclopropane
R1311	trifluoriodomethane
R1216	1, 1, 2, 3, 3, 3-hexafluoro-1-propene
RK	Redlich-Kwong

TOTA	type of ternary azeotropy
VDW	Van der Waals
VLE	vapor liquid equilibrium
Z	zeotropic

Symbols

A	Antoine equation parameter
a	attractive parameter in the EoS
a _m	attractive parameter of the mixture
a _{ij}	cross parameter of an EoS
a' _m	the partial derivative of a _m with respect to x _i
B	Antoine equation parameter
b	co-volume in the EoS
b _m	co-volume of the mixture
b' _m	the partial derivative of b _m with respect to x _i
C	Antoine equation parameter
K _{ij}	binary interaction parameter between components i and j
L	liquid phase
N	number of components
p	pressure, MPa
R	universal gas constant, J·mol ⁻¹ ·K ⁻¹
s	entropy, J·mol ⁻¹ ·K ⁻¹
T	temperature, K
x	liquid phase composition
y	vapor phase composition
Z	compressibility factor

Greek letters

μ	chemical potential
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Subscripts

i, j	component index
m	mixture

Superscripts

V	vapor phase
L	liquid phase

fractions instead of pressure. Hu and Chen et al. [13] estimated the vapor–liquid equilibria properties of several HFC binary refrigerant mixtures with a corresponding equation. The equation only needed the vapor pressures, critical constants and dipole moments of pure components, without any adjustable parameters or interaction coefficients. Hou and Duan et al. [14] developed the group contribution model to describe the vapor–liquid equilibria of the refrigerant mixtures and a ternary system was accurately predicted. Barley et al. [15] studied the ternary mixture (R32 + R125 + R143a) with Wilson activity coefficient model combined RK equation of state and predicted a saddle point azeotrope based on the binary experimental data. Aslam and Sunol [16] proposed a method establishing the pressure dependency of azeotropic composition allowing prediction of bifurcation pressure where refrigerant azeotropes may appear or disappear, with which the ternary azeotropic system proposed by Barley et al. was successfully predicted.

Most of those approaches are difficult to operate because of complicated mathematical computation, although they were proved successful to predict azeotropic behavior. In this paper, a simple method originally proposed by Dong et al. [17,18] was

extended to predict ternary systems. Based on the MATLAB procedure, one hundred and seventy-one interested ternary systems were investigated.

2. Method description

2.1. Azeotropy

In order to develop a method for finding all azeotropes of a mixture, it is essential to first determine the thermodynamic conditions for azeotropy. Homogeneous azeotropes occur in a boiling mixture of one liquid phase when the composition of the vapor phase is the same as the composition of the liquid phase. The thermodynamic condition was mentioned by Malesiński [19].

The Gibbs-Duhem relation gives

$$sdT - vdp + \sum_i n_i d\mu_i = 0. \quad (1)$$

The symbol s refers to the molar entropy, and v refers to the molar volume. The symbol μ refers to the chemical potential with the subscript *i* refers to the component *i*. For a ternary system, the

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