EXPERIMENTAL MEASUREMENTS AND PREDICTION OF THERMOPHYSICAL PROPERTIES OF HYDROCARBON MIXTURES USING NONLINEAR MODELS

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We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Doctor of Philosophy in Chemical Engineering.

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I hereby declare that the work in this thesis is based on my original work except for quotations and citation which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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DEDICATION

This thesis is dedicated to my parents and all family members whose love, care and prayers enabled me to complete this task.

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LIST OF SYMBOLS

η_1 , η_2	Absolute viscosity for pure components 1 and 2
η_{mix}	Absolute viscosity for binary mixtures
$\Delta\ \eta$	Viscosity deviation
A_j	Polynomial coefficient
^oC	Degree celsius
d	Grunberg-nissan interaction parameter
ρ_{mix}	Density of the binary liquid mixture
ρ_1, ρ_2	Density of the component liquids 1 and 2
G^E	Excess gibbs energy
ΔG^E	Excess gibbs free energy of activation of viscous flow
ΔG^*	Free energy of activation
H^E	Excess molar enthalpy
K	Kelvins
N	Number of data points
P	Number of coefficients
R	Gas constant
r_1, r_2	Radius of two liquid components 1 and 2
S^E	Excess molar entropy
σ	Standard deviation
T	Temperature
t	Flow time
ν_1, ν_2	Kinematic viscosity for pure components 1 and 2

ν_{mix}	Kinematic viscosity for binary mixtures
V_1, V_2	Molar volume of components 1 and 2
V^E	Excess molar volume
$\nu_{12,}$ ν_{21}	Binary interaction parameters of liquids
W	Mass fraction of solute
$x_{1,}$ x_2	Mole fraction of components 1 and 2
Y	Parameter of equation (3.10)

LIST OF ABBREVIATIONS

CFT	Collison factor theory
CFV	Cannon fenske viscometers
GLC	Gas liquid chromotography
G-N	Grunberg-Nissan model
GSCP	Generalized corresponding states principle
IMR	Ideal mixing relation
J-A	Jouyban-Acree model
K-L	Krishnan-Laddha model
LDF	London dispersion force
M	Molecular weight
M-A	Mc-Allister model
N	Avogadro’s number
NR	Nomotos relations
PFPT	Progogine Flory Patterson theory
R-K	Redlich-Kister model
SA	Self associated
U	Ultrasonic velocity
UNIQUAC	Universal quasi chemical
V	Molar volume

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ABSTRAK

Pada masa kini, sifat-sifat termofizikal campuran hidrokarbon memainkan peranan yang penting dalam industri proses. Aset termofizikal campuran pelarut menyediakan struktur maklumat untuk penyulingan, operasi pengekstrakan serta keseimbangan unsur dan tenaga dalam industri proses. Ia juga memainkan peranan penting dalam menyelesaikan masalah pemindahan haba, pemindahan jisim dan aliran bendalir. Baru-baru ini terdapat kemajuan besar dalam kajian mengenai interaksi antara molekul dan struktur dalaman campuran cecair. Kajian mengenai sifat-sifat termo-fizikal campuran cecair di dalam pelbagai komposisi dan suhu memberi sumber maklumat berharga yang boleh digunakan untuk meneliti hubungan di antara struktur dalaman sistem dan sifat fizikalnya. Walaupun banyak kajian telah dilakukan dalam cecair tulen, data tentang sifat-sifat termofizikal campuran cecair tidak terdapat di dalam kebanyakan sistem. Oleh itu, sifat-sifat termofizikal campuran cecair telah dikaji bagi empat campuran cecair yang berasaskan sistem perduaan. Dalam kajian ini, sifat-sifat termofizikal campuran dwi cecair hidrokarbon telah ditentukan berdasarkan pada suhu 303.15, 308.15 dan 313.15K dengan menggunakan *Oswald Viscometer* and *Pycnometer*.

1, 4 Dioxane + Bromobenzena

1, 4 Dioxane + Etilbenzena

Acetophenone + P-Xylena

Acetophenone + O-Xylena

Sifat-sifat termofizikal seperti ketumpatan dan kelikatan campuran perduaan cecair telah ditentukan secara ujikaji di dalam pelbagai komposisi pada suhu 303.15K, 308.15K dan 313.15K. Sifat-sifat termo-fizikal campuran dwi cecair yang ditentukan melalui kaedah eksperimen digunakan untuk pengiraan lebihan isipadu molar V^E dan sisihan kelikatan $\Delta\eta$. Sifat-sifat termo-fizikal lebihan campuran cecair memberi maklumat tambahan mengenai interaksi molekul. Dengan itu, interaksi intermolekular campuran dibincangkan dengan bantuan data sifat-sifat termofizikal lebihan.

Penelitian terperinci kesusasteraan mendedahkan bahawa ramalan sifat-sifat termo-fizikal campuran cecair adalah terhad. Dengan satu matlamat, sifat-sifat termo-fizikal ketumpatan, kelikatan, lebihan isipadu molar V^E dan penyimpangan kelikatan $\Delta\eta$ campuran cecair diramal dengan menggunakan pelbagai model tidak linear. Nilai-nilai lebihan sifat-sifat termofizikal campuran cecair dihubungkan dengan menggunakan persamaan polinomial *Redlich-Kister* untuk mendapatkan pekali dan sisihan piawai. Model tidak linear *Grundberg-Nissan, Krishnan-Laddha, McAllister dan Jouyban-Acree* telah digunakan untuk meramal kelikatan campuran cecair biner dalam kajian ini. Parameter model *McAllister* ditentukan dengan menggunakan perisian 'pengaturcaraan - *Software'* manakala model parameter lain-lain ditentukan menggunakan kaedah persamaan *polinomial*. Semua nilai eksperimen dan model ramalan dibandingkan untuk mendapatkan sisihan piawai bagi setiap model.

Daripada pemerhatian, data lebihan isipadu molar V^E dan penyimpangan kelikatan $\Delta\eta$ untuk campuran cecair biner yang dikaji, menyokong faktor utama iaitu terdapat gangguan secara beransur-ansur molekul-molekul hidrokarbon aromatik dan mengesahkan bahawa hidrogen ikatan dan interaksi dwikutub dalam hidrokarbon aromatik menyebabkan nilai positif untuk lebihan isipadu molar V^E dan nilai negatif untuk penyimpangan kelikatan. Interaksi intermolekular fizikal yang lemah antara molekul hidrokarbon aromatik menguasai kesan struktur pecah campuran hidrokarbon pada penambahan hidrokarbon aromatik. Interaksi antara cecair dalam campuran cecair hidrokarbon adalah jenis interaksi sisihan piawai yang lemah. Persamaan *polinomial Redlich-Kister* mewakili lebihan isipadu molar dan deviasi kelikatan yang ditandakan dengan tepat oleh peratus sisihan kurang daripada 1.25. Data kelikatan adalah selari dengan *Grundberg-Nissan, Krishnan-Laddha, Jouyban-Acree* dan model *McAllister* untuk memperolehi pekali binari. Sisihan piawai telah dianggap antara hasil yang betul dan data yang dikira adalah sangat berguna untuk tingkah laku bercampur daripada campuran binari. Secara kesimpulannya dalam semua kes, data yang terdapat dengan nilai adalah sama apabila dibandingkan dengan model-model lain dan model *Jouyban-Acree* juga telah diperluaskan kepada kepadatan yang sangat baik untuk campuran ini. Interaksi molekul yang wujud diantara komponen-komponen dan perbandingan campuran cecair juga telah dibincangkan. Data yang diperolehi dari eksperimen dan korelasi sangat mengurangkan kesukaran dalam kerja kerja ujikaji dan menyediakan cara yang lebih cepat untuk meramalkan sifat-sifat termodinamik. Penggunaan data korelasi membolehkan untuk ramalan data pada masa akan datang. Apabila korelasi dikenali dengan hubungan yang kuat antara pembolehubah kajian, maka ramalan itu akan menjadi lebih tepat. Selain itu, penggunaan model korelasi ini memberi pengetahuan tentang tingkah-laku sebenar campuran cecair yang menggunakan hanya keputusan eksperimen dan sifat-sifat komponen tulen.

ABSTRACT

Nowadays, thermophysical properties of hydrocarbon mixtures play a vital role in process industries. A thermophysical asset of the solvent mixture properties provides information for distillation, extraction operations, material balance and energy balance in process industries. They also play an important role in solving problems in heat transfer, mass transfer and fluid flow. Studies on different thermophysical properties of liquid mixtures within a wide ranges of composition and temperatures are valuable sources of information that can be used to examine the relationship between the internal structure of the system and its physical properties. Though a lot of work has been done in the case of pure liquids, data on thermophysical properties of liquid mixtures are not available for many systems. Hence the thermophysical properties of liquid mixtures have been studied for four binary liquid mixtures in the current investigation. Thermophysical properties of the following hydrocarbon binary liquid mixtures have been determined by using Oswald Viscometer and Pycnometer at 303.15, 308.15 and 313.15 K.

1, 4 Dioxane + Bromobenzene

1, 4 Dioxane + Ethylbenzene

Acetophenone + P-Xylene

Acetophenone + O-Xylene

The thermophysical properties such as density, viscosity of binary liquid mixtures were determined experimentally over the entire composition range at 303.15K, 308.15K and 313.15K. The experimentally determined thermophysical properties of the binary liquid mixtures were used to calculate the excess molar volume, V^E and viscosity deviations $\Delta\eta$. The excess thermophysical properties of liquid mixtures provide additional information regarding molecular interactions. Hence the intermolecular interactions of the mixtures are discussed with the help of excess properties.

A perusal of the literature revealed that the predictions of thermophysical properties of liquid mixtures was scarce. With an aim, the thermophysical properties of density, viscosity, excess molar volume V^E, and viscosity deviations $\Delta\eta$ of liquid mixtures were predicted using various nonlinear models. The excess values of thermophysical properties of binary hydrocarbon liquid mixtures were correlated using *Redlich-Kister* polynomial equation to obtain their coefficients and standard deviations. *Grundberg-Nissan, Krishnan-Laddha, McAllister and Jouyban-Acree* viscosity models were used for predicting the viscosity of hydrocarbon liquid mixtures. The parameters of *McAllister* model were determined using a programming software and other models parameters were determined using polynomial equations. The experimental values and model predictions were compared to get the standard deviation for each model.

The observed values of excess molar volume V^E, viscosity deviations $\Delta\eta$ data for the hydrocarbon mixtures support the main factor of gradual disruption of the self-associated aromatic hydrocarbon molecules and confirm that the hydrogen bonds and

dipolar interaction in aromatic hydrocarbons makes V^E and $\Delta\eta$ positive and negative deviations. The weak physical intermolecular interactions between the aromatic hydrocarbon molecules dominate over the structure-breaking effect of hydrocarbon mixtures on the addition of aromatic hydrocarbons. Interactions between the liquids in hydrocarbon mixtures are strong and weak dispersive type interactions. *Redlich-Kister* polynomial equation represented the excess molar volume and viscosity deviations which is indicated accurately well by percent standard deviation of less than 1.25. The viscosity data tailored to *Grundberg-Nissan, Krishnan-Laddha, Jouyban–Acree and McAllister* models to derive the binary coefficients. Standard deviations have been considered between the fitted outcomes and the calculated data is helpful in deliberate mixing behavior of the binary mixtures. It can be concluded that in all cases, the data values found correlated with the corresponding models very well for this mixtures. The molecular interactions existing between the components and comparison of liquid mixtures were also discussed. The obtained experimental and correlation data is greatly reducing the difficulties in a tedious experimental work and provides a faster way of predicting the property values. The use of data correlation enables for prediction of data in future. Once a correlation is known with strong relationship between the variables of the study, then the prediction will be more accurate. Other than that, the use of these correlation models provides knowledge on the real behaviour of the liquid mixtures using only the experimental results and properties of the pure components.

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