EXPERIMENTAL MEASUREMENT AND PREDICTION OF THERMOPHYSICAL PROPERTIES OF METHYL TERT-BUTYL ETHER (MTBE) WITH METHANOL, DIISOPROPYL ETHER (DIPE) AND N-METHYLANILINE BINARY MIXTURES

MOHAMMED RASHID ALI

MASTER OF SCIENCE

UNIVERSITI MALAYSIA PAHANG

SUPERVISOR'S DECLARATION

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 Master of Science.

____

(Supervisor's Signature)

Full Name : DR NOOR ASMA FAZLI BIN ABDUL SAMAD

Position : SENIOR LECTURER

Date :

____

(Co-supervisor's Signature)

Full Name : DR MOHD YUSRI BIN MOHD YUNUS

Position : SENIOR LECTURER

Date :

STUDENT'S DECLARATION

I hereby declare that the work in this thesis is based on my original work except for quotations and citations 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.

____

(Student's Signature)

Full Name : MOHAMMED RASHID ALI

ID Number : MKC 16025

Date :

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MOHAMMED RASHID ALI

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ABSTRAK

Sifat termofizikal bagi campuran hidrokarbon memainkan peranan penting dalam prosess industri seperti proses penyulingan, operasi pengekstrakan, imbangan jisim dan tenaga serta menyelesaikan masalah dalam pemindahan haba, pemindahan jisim dan aliran bendalir. Kajian ke atas sifat termofizikal seperti ketumpatan dan kelikatan cecair campuran pada komposisi dan suhu yang berbeza adalah sumber maklumat yang bernilai dalam menilai hubungan diantara struktur dalaman dan sifat fizikal. Dalam kajian ini, ketumpatan dan kelikatan campuran cecair binari bagi *methyl tert-butyl ether* (MTBE) dengan metanol, *diisopropyl ether* (DIPE) dan *N-methylaniline* (NMA) telah diukur dengan menggunakan alat DMA 4500 M meter ketumpatan dan alat Anton Paar´s Stabinger Viscometer™ SVM™ 3000 pada suhu 35 °C, 40 °C dan 45 °C. Ketumpatan dan kelikatan bagi campuran MTBE-metanol dan MTBE-NMA menunjukkan penurunan dengan peningkatan komposisi MTBE pada semua suhu. Manakala campuran MTBE-DIPE menunjukkan peningkatan dalam ketumpatan dan kelikatan. Selain itu, persamaan Jouyban-Acree telah digunakan untuk menganggarkan nilai ketumpatan dan kelikatan yang mana purata ralat mutlak (MAE) yang rendah iaitu 0.3 % dan 2.2 % telah diperolehi. Lebihan isipadu molar dan sisihan kelikatan telah dikira menggunakan data eksperiman bagi ketumpatan dan kelikatan. Ia juga telah dikorelasikan dengan persamaan Redlich-Kister yang mana kesesuaian yang baik telah dicapai. Berdasarkan nilai tersebut, lebihan isipadu molar yang negatif adalah lebih bersesuain untuk mendapatkan kombinasi bahan api yang kuat. Maka campuran MTBE-metanol dan MTBE-NMA mesti dicampur pada komposisi MTBE yang tinggi. Walau bagaimanapun komposisi MTBE yang rendah perlu digunakan bagi campuran MTBE-DIPE. Sisihan kelikatan yang negatif telah dicapai bagi semua campuran dan nilainya semakin berkurang apabila suhu meningkat. Ini menunjukkan kekuatan kimia atau interaksi yang spesifik untuk campuran binari dari segi pengikatan hidrogen diantara ketidaksamaan molekul-molekul. Kajian mengenai interaksi molekul dilaksanakan dengan menggunakan analisis pengecilan jumlah pemantulan berdasarkan inframerah jelmaan Fourier (ATR-FTIR) dan analisis spektroskopi menggunakan salunan proton nuklear secara magnetik (^{1}H-NMR) berdasarkan campuran binari pada nisbah 3:7 dan 7:3. Berdasarkan pemerhatian, interaksi molekular lebih terbukti pada nisbah 7:3 bagi cecair campuran MTBE-metanol dan MTBE-NMA. Walau bagaimanapun interaksi molekular bagi MTBE-DIPE tidak menunjukkan perubahan pada kedua-dua nisbah. Ini menunjukkan kekuatan interaksi inter-molekul bagi MTBE dan DIPE tidak bergantung kepada kuantiti komponen DIPE dalam campuran cecair binari tersebut.

ABSTRACT

Thermophysical properties of hydrocarbon mixtures play a vital role in process industries such as distillation, extraction operations, material and energy balance as well as solving problems in heat transfer, mass transfer, and fluid flow. Studies on thermophysical properties such as density and viscosity of liquid mixtures within a wide range of composition and temperatures are valuable sources of information for examining the relationship between the internal structure of the system and its physical properties. In this work density and viscosity of methyl tert-butyl ether (MTBE) with methanol, diisopropyl ether (DIPE) and N-methylaniline (NMA) binary liquid mixtures have been measured by using DMA 4500 M density meter and Anton Paar´s Stabinger Viscometer™ SVM™ 3000 at temperatures of 35 °C, 40 °C and 45 °C. The densities and viscosities of MTBE-methanol and MTBE-NMA binary mixtures show decreasing trends with the increase of MTBE composition at all temperature. On the contrary, binary mixture of MTBE-DIPE shows an increment in density and viscosity under the same condition. In addition, Jouyban-Acree equation has been used to predict density and viscosity where low mean absolute error (MAE) of 0.3 % and 2.2 % have been obtained. The excess molar volumes and viscosity deviation have been calculated using the density and viscosity experimental data and correlated with Redlich-Kister equation where a good fitted has been obtained. Sigmoid shapes of excess molar volumes have been obtained for all binary mixture. It has been found that in order to obtain a strong combination fuel, negative excess molar volumes are preferable. Thus binary mixtures of MTBE-methanol and MTBE-NMA must be mixed at high mole fraction of MTBE. However low mole fraction of MTBE must be used in the case of MTBE-DIPE binary mixture. The viscosity deviations of all binary mixtures are negative and were observed to decrease with an increase of temperatures. This indicates the strength of chemical or specific interaction for binary mixtures such as hydrogen binding between dissimilar molecules. Further investigation of molecular interaction has been performed using Attenuated Total Reflectance-Fourier Transform Infrared (ATR-FTIR) and Proton Nuclear Magnetic Resonance (^{1}H-NMR) spectroscopy analysis using ratios 3:7 and 7:3 of binary mixture. It has been observed that intermolecular interaction is more evident for ratio 7:3 for MTBE-methanol and MTBE-NMA. However, the variation in percentage composition of MTBE with DIPE of the individual components does not seem to affect significantly the spectrum of the binary mixture. Hence, this suggests that the intermolecular interaction between MTBE and DIPE is less dependent on the amount of DIPE present in the binary mixture.

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LIST OF SYMBOLS

-	Subtract
%	Percentage
+	Add
$<$	Less than
=	Equals
$>$	Greater than
$\leq$	Less than or equal to
$\geq$	More than or equal to
°C	Degree Celsius
s	second
cm	Centimetre
cm^{-1}	Reciprocal centimetre
ml	Millilitre
g	Gram
g/cm^3	Gram per cubic centimetre
g/mol	Gram per mol
m^2/s	Square metre per second
mPa.s	Megapascal second
cm^3/mol	Cubic centimetre per mol
×	Multiple
σ	Standard deviation
ρ	Density
η	Dynamic viscosity
ν	Kinematic viscosity
V	Molar volume
M	Molecular mass
V^E	Excess molar volume
$\Delta\eta$	Viscosity deviation
$\Delta G*$	Gibbs free energy of activation
R^2	Correlation coefficient
x_1	mole fraction of MTBE

LIST OF ABBREVIATIONS

APD	Average Percentage Deviations
API	American Petroleum Institute
ASTM	American Society for Testing and Materials
ATR-FTIR	Attenuated Total Reflectance-Fourier Transform Infrared
CAA	Clean Air Act Amendments
DEE	2-(2- ethoxy-ethoxy) ethanol
DIPE	Diisopropyl ether
EE	Ethoxyethanol
ETBE	Ethyl tert-butyl ether
ICE	Internal Combustion Engine
IPE	Isopropyl ether
MMA	Monomethylaniline
MTBE	Methyl-tert butyl ether
NMA	N-Methylaniline
RFG	Reformulated Gasoline
TAME	Tert-amyl methyl ether
TEE	2-[2-(2-ethoxyethoxy) ethoxy] ethanol
[EMIM][ES]	1-Ethyl-3-methylimidazolium
% MAE	Percentage of mean absolute error
^{1}H-NMR	Proton Nuclear Magnetic Resonance

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