2004

A study of the viscometric and volumetric properties of some multi-component liquid regular solutions at different temperatures.

Omar Medhat El-Hadad
University of Windsor

Recommended Citation
El-Hadad, Omar Medhat, "A study of the viscometric and volumetric properties of some multi-component liquid regular solutions at different temperatures." (2004). *Electronic Theses and Dissertations*. 818.
https://scholar.uwindsor.ca/etd/818

A Study of the Viscometric and Volumetric Properties of Some Multi-Component Liquid Regular Solutions at Different Temperatures

by

Omar Medhat El-Hadad

A Thesis Submitted to the Faculty of Graduate Studies and Research
Through the Environmental Engineering Program
in Partial Fulfilment of the Requirements
for the Degree of
Masters of Applied Science
at the University of Windsor

Windsor, Ontario, Canada
2004

National Library
of Canada

Acquisitions and
Bibliographic Services

395 Wellington Street
Ottawa ON K1A 0N4
Canada

Bibliothèque nationale
du Canada

Acquisisitons et
services bibliographiques

395, rue Wellington
Ottawa ON K1A 0N4
Canada

Your file Votre référence
ISBN: 0-612-92467-X
Our file Notre référence
ISBN: 0-612-92467-X

The author has granted a non-exclusive licence allowing the National Library of Canada to reproduce, loan, distribute or sell copies of this thesis in microform, paper or electronic formats.

L'auteur a accordé une licence non exclusive permettant à la Bibliothèque nationale du Canada de reproduire, prêter, distribuer ou vendre des copies de cette thèse sous la forme de microfiche/film, de reproduction sur papier ou sur format électronique.

The author retains ownership of the copyright in this thesis. Neither the thesis nor substantial extracts from it may be printed or otherwise reproduced without the author's permission.

L'auteur conserve la propriété du droit d'auteur qui protège cette thèse. Ni la thèse ni des extraits substantiels de celle-ci ne doivent être imprimés ou aturement reproduits sans son autorisation.

In compliance with the Canadian Privacy Act some supporting forms may have been removed from this dissertation.

Conformément à la loi canadienne sur la protection de la vie privée, quelques formulaires secondaires ont été enlevés de ce manuscrit.

While these forms may be included in the document page count, their removal does not represent any loss of content from the dissertation.

Bien que ces formulaires aient inclus dans la pagination, il n'y aura aucun contenu manquant.

Canada

Reproduced with permission of the copyright owner. Further reproduction prohibited without permission.

ABSTRACT

The densities and kinematic viscosities of the quinary liquid regular system octane(1)-hexane(2)-ethylbenzene(3)-cyclohexane(4)-toluene(5) and two of its quaternary subsystems were measured at 293.15 K and 298.15 K. In addition, the densities and kinematic viscosities of the quinary liquid regular system octane(1)-heptane(2)-ethylbenzene(3)-cyclohexane(4)-toluene(5) and all of its quaternary and ternary subsystems were measured at the same two temperatures.

The data collected in the present study were employed in testing the predictive capabilities of some of the most widely used viscosity models for liquid mixtures.

Comparison of the predictive capabilities of the various literature models showed indicated that the generalized McAllister three-body collision model gave the lowest deviation from the experimental results.

بِسْمِ اللَّهِ الرَّحْمَـنِ الرَّحِيمِ

"وَجَاهِدُوا فِي اللَّهِ حَقَّ جِهَادِهِ"
سورة الحج

In the name of Allah, Most Gracious, Most Merciful

“And strive for Allah with the endeavour which is His right.”

AL-HAJJ (THE PILGRIMAGE): 78

ACKNOWLEDGEMENTS

All praise and thanks is to His Almighty Allah, who gave me the ability to start and complete this study, and who granted me help and support.

I would like also to thank Dr. Abdul-Fattah Asfour, for guiding me throughout the present study. I have to thank Dr. Asfour for being one of the kindest and most helping and co-operating professors I have encountered. May Allah accept this from him.

I have also to thank my parents who encouraged me throughout my study. My thanks are also due to Dr. R. R. Zahran from Alexandria University for his help.

I would also like to thank my kind colleague and partner, Mr. Ruo Cai, for all his help and co-operation during the present study.

My thanks also go to my colleague Mr Walid Al-Gherwi for help with the computer work employed in the calculations reported in this study and in the preparation of the graphs.

I appreciate and thank my colleagues in the Environmental Engineering program at the University of Windsor, who were always present at the time of need.

TABLE OF CONTENTS

LIST OF TABLES

List of Figures

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 General

Viscosity is a physical property that describes the resistance to the flow of simple fluids and is necessary in the calculations of fluid flow. It can be defined as the ratio of shear stress to velocity gradient that is perpendicular to flow as a result of stress. Thus, the viscosity gives an idea about the internal fluid friction that acts against dynamic change. Viscosity of liquid mixtures depends on the composition, and knowledge of this dependence is required in many engineering applications that deal with heat transfer, fluid flow or mass transfer.

Viscosity of gases results from the transfer of momentum emanating from the translational motion of the molecules having few collisions; this is described by the kinetic theory of gases. However, the momentum transfer in liquids is controlled by collisions and interacting force fields between the molecules. Because of the nature of the intermolecular forces involved in liquids, very little progress has been made when compared with the progress made in the case of the viscosity of gases. This is simply because of the negligible interaction among molecules in case of gases. Another source of complication is our very little knowledge about the structure of liquids. Our little knowledge in this area hindered researchers from developing a reliable theory for predicting the viscosities of liquid mixtures.

In order to circumvent those problems, Asfour (1980, 1985 and 1991) resorted to classifying liquid mixtures into three categories; viz., n-alkane systems, regular solutions and associated liquid mixtures. Asfour and co-workers (1991, 1992, 1998, 2000a, 2000b) reported substantial contributions on the prediction of viscosity of both n-alkane systems and regular solutions. The most significant contributions made by Asfour and co-workers involved the successful prediction of viscosities of multi-component liquid systems (Wu and Asfour 1992; Nhaesi and Asfour 2000a and 2000b).

The models employed in the literature to calculate the viscosities of liquid mixtures can be classified into correlative or predictive equations. The correlative models depend on experimental data which are used to calculate the values of the adjustable parameters involved in a mathematical model. In case of the predictive models, the properties of pure components as well as molecular parameters are utilized to describe the dependence of viscosity on composition.

In this study, the experimental data obtained are employed to test the validity of models reported by Asfour and co-workers as well as models reported in the literature and comparing the performance of those models. The literature models include: the Generalized Corresponding State Principle (GCSP), the modified GCSP, the Group Contribution Method, the McAllister Model, and the Allan and Teja method.

The GCSP when used to test systems containing more than two components, an appropriate choice of two reference fluids is required. It was shown by Wu and

Asfour (1992) that for n-alkane systems, selecting different reference fluids could give significantly different results. But actually there is no basis on which the two reference fluids are chosen. To solve this problem, Asfour and Wu (1992) suggested treating a multi-component n-alkane mixture as a pseudo-binary mixture, and the GCSP was modified on that basis. The results reported by Wu (1992) showed that the results of the modified GCSP are better than the results of the original GCSP.

However, Wu and Asfour (1992) focused on n-alkane systems and compared the accuracy of the modified GCSP with the original GCSP method; also this method is limited to certain temperature ranges. Asfour and Nhaesi (1998) extended the modified GCSP to include any regular multi-component liquid mixture at several temperature levels. They also compared the predictive capability of the method with other methods.

The group contribution method is based on the assumption of additivity, which means that each component is treated as a number of groups rather than a single molecule, and the sum of contributions made by each group represents the physical property of the compound concerned. Based on this, one can conclude that the group contribution method is approximate because the contribution of a given group in one defined molecule is not the same as that in another defined molecule.

Based on Eyring's absolute rate theory, a semi-empirical equation was proposed by McAllister (1960). Many researchers consider the McAllister equation as the best correlative method for binary liquid systems (Reid *et al.* 1977). The McAllister model was extended to cover ternary systems by Chandramouli and

Laddha (1963). It was pointed out by Asfour *et al.* (1991) that the need to determine the adjustable parameters in the McAllister model limits its use and does not make it attractive since obtaining experimental data is both time consuming and costly.

Asfour *et al.* (1991) developed a technique to predict the numerical values of the McAllister model parameters for *n*-alkane systems from pure component properties and molecular parameters. In this way, the Asfour *et al.* (1991) technique effectively converts the McAllister model from a correlative equation to a predictive model.

Nhaesi and Asfour (1998) reported a method for predicting the McAllister model interaction parameters from pure component properties; this can be done for binary and ternary liquid regular systems. They have also reported, for the first time in the literature (2000a), a general expression for McAllister three-body model for multi-component systems. The combination of this method for predicting the binary and ternary interaction parameters with the general expression of the McAllister three-body model for multi-component systems can be used for the prediction of the viscosity of the regular multicomponent liquid mixtures.

The experimental data obtained during this study were used to subject to further testing the method reported by Nhaesi and Asfour (1998, 2000a and 2000b). The results were also compared with the results obtained using the other method mentioned earlier.

1.2 Objectives

The present study is dealing with the prediction of viscosities of liquid regular solutions, where published data is rare. The objectives of this work include:

(i) Obtaining viscosity data for the quinary system octane(1)-heptane(2)-ethylbenzene(3)-cyclohexane(4)-toluene(5) and its quaternary and ternary subsystems over the entire composition range at 293.15 and 298.15.

(ii) Obtaining viscosity data for the quinary system octane(1)-hexane(2)-ethylbenzene(3)-cyclohexane(4)-toluene(5) and two of its quaternary subsystems over the entire composition range at 293.15 and 298.15.

(iii) Using the experimental data gathered in this study to test the predictive capability of some literature models.

1.3 Contributions and significance

Values of viscosities of liquid mixtures are needed in many engineering applications as well as for design purposes. Experimental measurement of such viscosities is both time consuming and costly. Therefore, obtaining reliable models for estimating viscosities over the entire composition range represents a valuable contribution.

In this study, Experimental measurements of viscosities of multicomponent regular liquid systems are reported over the entire composition range. These data represent a valuable addition to the literature where data on the viscosities of multicomponent systems are scarce.

In Addition, the experimental data reported herein were employed in testing the viscosity models reported in the literature.

CHAPTER 2

LITERATURE SURVEY

2.1 General

Viscosity as a transport property plays a significant role in many engineering applications. The knowledge of viscosity is required for the design of equipment involving heat transfer and mass transfer operations. There are some excellent reviews in the literature about models for the prediction of the viscosity of liquid mixtures, e.g., Reid *et al.* (1987) and Monnery *et al.* (1995) and Mehrotra *et al.* (1996). According to Mehrotra, there are two types of models to predict the viscosity of liquid mixtures; these are either empirical or semi-theoretical. Empirical models are based on experimental data, whereas semi-theoretical models have a theoretical basis but contain adjustable parameters which are determined from experimental data. Viscosity models can also be classified as predictive or correlative. Predictive models use the properties of pure components as well was molecular parameters to calculate viscosities whereas correlative models use experimental mixture data.

The classification of Monnery *et al.* (1995) is used in this chapter. Each type is presented in a section, and the models of this type are reported in the subsections. At the end of the chapter, there is the terminology of some viscosity related properties.

2.2 Semi-Theoretical Models for Liquid Mixtures Viscosity

2.2.1 The corresponding states principle

Van der Waals proposed the theory of corresponding states on the basis of his equation of state. And on the basis of the principle of continuity of the gaseous and liquid phases, at the critical point van der Waals showed that:

$$\left(\frac{\partial P}{\partial V}\right)_T = \left(\frac{\partial^2 P}{\partial V^2}\right)_T = 0 \tag{2.1}$$

According to equation (2.1), van der Waals generalized the result so that for the variables (PVT) there exists a universal function at the critical point, so that

$$f(T_r, P_r, V_r) = 0 \tag{2.2}$$

where $T_r = T/T_c$, $P_r = P/P_c$, and $V_r = V/V_c$. Another way to express this result is to say that for all substances, the equation of state for the reduced variables will be valid. One can say about any two substances that they are in corresponding states when the reduced variables for both of them are equal.

Helfand and Rice (1960), Preston *et al.* (1967) and Tham and Gubbins (1969, 1970) extended the corresponding states principle to transport properties. According to this principle, one can say that a dimensionless property of one substance is equal to that of another substance at the same reduced conditions. On that basis if X is a reduced property for fluid i, one can express it in terms of reference fluid "o" as follows:

$$X^i(T_r, V_r) = X^o(T_r, V_r) \tag{2.3}$$

or

$$X^i(T_r,P_r) = X^o(T_r,P_r) \tag{2.4}$$

Teja *et al.* (1985) reported that it is not a must for the correspondence calculated in terms of the same reduced temperature and pressure to be equivalent to correspondence calculated in terms of reduced temperature and volume, but when the critical compressibility of the fluid of interest and reference are equal, correspondence calculated in terms of the same reduced temperature and pressure will be equivalent to correspondence calculated in terms of reduced temperature and volume.

By substituting the reduced property X with the reduced viscosity η, equation (2.3) becomes:

$$(\eta\varepsilon)^i(T_r,V_r) = (\eta\varepsilon)^o(T_r,V_r) \tag{2.5}$$

where η is the viscosity at the temperature of interest and ε is the reciprocal of viscosity at T_c (Reid *et al.* 1987). Employing dimensional analysis to calculate ε, the following equation can be obtained:

$$\varepsilon = M^{1/2}V_c^{1/2}T_c^{-1/2} \tag{2.6}$$

or in terms of T_c and P_c as

$$\varepsilon = M^{1/2}P_c^{-2/3}T_c^{1/6} \tag{2.7}$$

According to Hwang *et al.* (1987), in the two-parameter corresponding states principle, the ratios of equivalent to actual temperature and of equivalent to actual densities are equated to the ratios of critical constants as follows:

$$\frac{T^o}{T^i} = \frac{T_c^o}{T_c^i} \tag{2.8}$$

$$\frac{\rho^o}{\rho^i} = \frac{\rho_c^o}{\rho_c^i} \tag{2.9}$$

This method is valid only for simple fluids, which conform to the two-parameter corresponding state and is not valid for poly-atomic fluids or mixtures. Ely and Hanely (1981) and William and Teja (1988) suggested two different approaches for extending the applicability of the two-parameter corresponding states.

2.2.2 The Extended corresponding states principle (ECSP)

(i) Shape factor approach

This method depends on the extended corresponding states model (Leland *et al.* 1968; Rowlinson and Watson 1969). In this method, the two-parameter corresponding states formalism is maintained, the only change is that equations (2.8) and (2.9) now contain a term called the "shape factor"

$$\frac{T^o}{T^i} = \frac{T_c^o}{T_c^i \theta_{i,o}(T_r, V_r, \omega^i)} \tag{2.10}$$

$$\frac{\rho^o}{\rho^i} = \frac{\rho_c^o}{\rho_c^i}\varphi_{i,o}(T_r, V_r, \omega^i) \tag{2.11}$$

where $\theta_{i,o}$ and $\varphi_{i,o}$ are called the shape factors. These are functions of the Pitzer acentric factor, ω, and of the reduced variables. Rowlinson and Watson (1969) reported that the shape factors can be calculated exactly for any pure fluid by solving the following two simultaneous equations:

$$Z^i\left(T^i, \rho^i\right) = Z^o\left(T^o, \rho^o\right) \tag{2.12}$$

$$A^i\left(T^i, \rho^i\right) = A^o\left(T^o, \rho^o\right) \tag{2.13}$$

where Z is the compressibility factor, A is the molar reduced Helmholtz free energy relative to the ideal gas at the same temperature and pressure. Equations similar to equations (2.12) and (2.13) for paraffins with carbon atoms between C_1 and C_{20} and with methane as the reference fluid were solved by Leach (1967). The results gave the following empirical expressions:

$$\theta_{i,o} = 1 + (\omega_i - \omega_o)\left[a_1 + b_1 \ln(T_{r,i}) + \left(c_1 + \frac{d_1}{T_{r,i}}\right)(V_{r,i} - 0.5)\right] \tag{2.14}$$

$$\varphi_{i,o} = \left[1 + (\omega_i - \omega_o)a_2(V_{r,i} + b_2) + c_2(V_{r,i} + d_2)\ln(T_{r,i})\right](Z_{c,o}/Z_{c,i}) \tag{2.15}$$

where the reduced parameters $V_{r,i}$ and $T_{r,i}$ must be between 0.5 and 2.0. If not, $V_{r,i}$ will be equal to 0.5 and $T_{r,i}$ will be equal to 2.0.

The "van der Waals" one fluid nixing rules are used for fluid mixtures to calculate the pseudo-critical properties as follow:

$$V_{cm} = \sum_i \sum_j X_i X_j V_{cij} \tag{2.16}$$

$$T_{cm} V_{cm} = \sum_i \sum_j X_i X_j T_{cij} V_{cij} \tag{2.17}$$

T_{cij} and V_{cij} are the cross parameters which can be calculated by the following equations:

$$V_{cij} = \eta_{ij}\left[\frac{\left(V_{ci}^{1/3} + V_{cj}^{1/3}\right)}{2}\right]^3 \tag{2.18}$$

$$T_{cij} = \xi_{ij}\left[T_{ci} T_{cj}\right]^{0.5} \tag{2.19}$$

where η_{ij} and ξ_{ij} are the binary interaction parameters. These parameters characterize the non-ideality of interaction between the binary pair. In many cases, their values are assumed to be one (Wong *el al.* 1984). These parameters may be obtained by experimental data.

Many investigators, such as Lee and Kesler (1975), Joffe (1971), and Ploecker *et al.* (1978) worked to determine the mixture acentric factor by using the following simple linear mixing rule:

$$\omega_m = \sum_i X_i \omega_i \tag{2.20}$$

In order to use this method for the prediction of viscosity, Ely and Hanley (1981) developed a program for predicting the transport properties (TRAPP: **Tra**nsport **P**roperties **P**rediction). The above authors tested their method by using the experimental data of CO_2, 35 pure hydrocarbons, and 26 binary hydrocarbon mixtures. The predicted results had an absolute percent deviation of 8.5% and 7% for pure hydrocarbons and hydrocarbon mixtures, respectively. If the adjustable parameters in equations (2.18) and (2.19), η_{ij} and ξ_{ij}, are set equal to unity; the method can be used as a predictive method.

Ely and Hanley (1981) pointed out that there is no restriction imposed by their method in choosing the reference fluid, and also the reference fluid may be a component that is not present in the mixture.

Although the results of the method depend mainly on the choice of an appropriate reference fluid, the sensitivity of the method to the selection of the reference fluid was not tested. It is generally advisable to fix the reference the fluid.

(ii) the two reference fluid approach

Pitzer *et al.* (1995 a, b) made the analogy on which the method of corresponding states principle was based; they also showed that the compressibility factor can be defined as a linear function of the acentric factor:

$$Z = Z^{o} + \omega Z^{1} \tag{2.21}$$

where Z^{o} is the compressibility factor of a simple spherical fluid with zero acentric factor. Another name for this method is the three-parameters corresponding states. A model of this type for saturated liquids was developed by Letsou and Stiel (1973):

$$Z^{o} = \frac{\partial Z}{\partial \omega} \tag{2.22}$$

$$\ell n(\eta\varepsilon) = \ell n(\eta\varepsilon)^{o} + \ell n(\eta\varepsilon)^{1} \tag{2.23}$$

According to Lee and Kesler (1975), equation (2.21) can be expressed as:

$$Z = Z^{o} + \frac{\omega}{\omega^{r}}\left(Z^{r} - Z^{o}\right) \tag{2.24}$$

where ω^{r} and Z^{r} are the acentric factor and compressibility factor of a non-spherical reference fluid, respectively. The partial derivative with a finite difference between a simple fluid and non-spherical reference fluid was approximated basically by Lee and Kesler (1975).

Teja *et al.* (1981) reported that by eliminating the restrictive assumptions in the Lee-Kesler expression of choosing a spherical fluid having an acentric factor of zero as one of the references, this gave better predictive accuracy. Consequently, the Lee-Kesler form can be re-written as:

$$Z = Z^{r1} + \frac{\omega - \omega^{r1}}{\omega^{r2} - \omega^{r1}}\left(Z^{r2} - Z^{r1}\right) \tag{2.25}$$

where the superscripts r1 and r2 refer to two non-spherical reference fluids which are similar to the component studied, or for mixtures, to the key components of interest. The extension to the corresponding states principle gives this form which is known as the Generalized Corresponding States Principle (GCSP). Equation (2.25) takes the form of Lee-Kesler, equation (2.24), if one of the reference fluids is a simple fluid.

In the same way, equation 2.25 was applied to viscosity calculations by Teja and Rice (1981):

$$\ell n(\eta\varepsilon) = \ell n(\eta\varepsilon)^{r1} + \frac{\omega - \omega^{r1}}{\omega^{r2} - \omega^{r1}}\left[\ell n(\eta\varepsilon)^{r2} - \ell n(\eta\varepsilon)^{r1}\right] \tag{2.26}$$

Equation (2.26) was extended to mixtures by Teja and Rice (1981) by using the van der Waals one fluid model mixing rules in order to calculate the properties of the pseudo-critical mixture T_{cm} and V_{cm}. The Teja-Rice viscosity method was expressed by Teja and Thurner (1986) in terms of reduced temperature and pressure instead of critical volume. Therefore, they had to use the Wong *et al.* (1984) mixing rules. These mixing rules are given by:

$$\frac{T_{cm}^2}{P_{cm}} = \sum_i \sum_j X_i X_j \left(\frac{T_{cij}^2}{P_{cij}}\right) \tag{2.27}$$

$$\frac{T_{cm}}{P_{cm}} = \sum_i \sum_j X_i X_j \left(\frac{T_{cij}}{P_{cij}} \right) \tag{2.28}$$

$$\omega \left(\frac{T_{cm}^2}{P_{cm}} \right)^{2/3} = \sum_i \sum_j X_i X_j \left(\frac{T_{cij}^2}{P_{cij}} \right)^{2/3} \omega_{ij} \tag{2.29}$$

$$T_{cij} = \xi_{ij} \left(T_{cii} T_{cjj} \right)^{1/2} \tag{2.30}$$

$$\left(\frac{T_{cij}}{P_{cij}} \right)^{1/3} = \frac{1}{2} \left[\left(\frac{T_{cii}}{P_{cii}} \right)^{1/3} + \left(\frac{T_{cjj}}{P_{cjj}} \right)^{1/3} \right] \tag{2.31}$$

$$\omega_{ij} = \frac{1}{2} \left(\omega_{ii} + \omega_{jj} \right)^{1/2} \tag{2.32}$$

$$M_m = \sum_i X_i M_i \tag{2.33}$$

Equation (2.33) represents a simple mixing rule for molecular weights. If Z_c does not change much from one substance to another, one can consider that V_c is equal to T_c/P_c. In this case V_c is proportional to Z_cT_c/P_c. Therefore, equations (2.27) and (2.28) are equivalent to the van der Waals mixing rules. If one followed that procedure, it is clear that the mixing rules in this case are more accurate to use as they do not contain the critical volume, which has a higher experimental error than the critical temperature and pressure.

If the binary interaction parameter in equation (2.30), ξ_{ij}, was not assumed a given value, i.e. its value has to be calculated, the method is correlative, but if it is assumed to have a value of one, the GCSP method can be used as a predictive technique.

Teja and Thurner (1986) employed the technique in a comparison with the TRAPP method for the five binary systems methane(1)-butane(2), ethane(1)-ethylene(2), benzene(1)-hexane(2), benzene(1)-decane(2), and methane(1)-decane(2). In either method, the binary interaction parameters were not used. The results show that the GCSP method is slightly better than those obtained from TRAPP, but this depends on the choice of reference fluids. The accuracy remarkably decreases (the average deviations are higher) if the viscosity data for the reference fluids were extrapolated.

As with the corresponding states principle method, the choice of the reference fluids has an effect on the accuracy of the results. The problem here again is that there is no basis on which the reference fluids can be selected. Wu and Asfour (1992) have discussed the effect of the choice of the reference fluids on the accuracy of prediction. To alleviate that reference fluid selection problem, they suggested the use of a pseudo-binary model, a modification to the GCSP method. Discussion of the pseudo-binary model is in order.

(iii) the modified GCSP[MGCSP] (pseudo-binary model approach)

The model reported by Wu and Asfour (1992) modifies the GCSP method in such a way that the unreliability of the choice of a reference fluid is eliminated. This increases the reliability of the GCSP in predicting the viscosities of multi-component liquid systems as was shown by Wu and Asfour (1992). The main idea of the method is to treat the muliticomponent mixture as a binary one, component (1) is the first pure component, whereas component (2') is the pseudo-single component representing components (2), (3) ... etc. Other parameters, e.g., the acentric factor, the viscosity of component (2,), the critical temperature and pressure, can be calculated by using the following polynomials:

$$\omega^{r2} = \sum_i A_i \overline{N}^i \qquad (2.34)$$

$$T_{c,r2} = \sum_i B_i \overline{N}^i \qquad (2.35)$$

$$P_{c,r2} = \sum_i C_i \overline{N}^i \qquad (2.36)$$

$$\ln(\xi\varepsilon)^{r2} = \sum_i D_i \overline{N}^i \qquad (2.37)$$

where the values of A_i, B_i, C_i, are listed in Table 2.1. As for both the viscosity and D_i, they are temperature dependent. The values of both of them at different temperatures are given in Table 2.2, and $\overline{N}$ is the average chain length of the pseudo-component, and is given by:

$$N = \sum_i X_i N_i \qquad (2.38)$$

Wong *et al.* (1984) mixing rules and equation (2.7) were used to calculate ω and ε for the mixture.

The modified GCSP (MGCSP) can either be correlative or predictive; this depends on how to deal with the interaction

Table 2.1: Least squares constants for equations (2.34), (2.35) and (2.36) (Wu and Asfour 1992)

	i=0	i=1	i=2	i=3
A_i	0.0399	0.04423		
B_i	226.487	60.8027	-2.6379	0.0477
C_i	58.5846	-6.8989	0.4039	-0.0092

Table 2.2: Least squares constants for equations (2.37) (Wu and Asfour 1992)

T (K)	D_0	D_1	D_2
293.15	-6.5212	0.3446	-0.0044
298.15	-6.5229	0.3368	-0.0043
303.15	-6.5239	0.3295	-0.0043
308.15	-6.5437	0.3256	-0.0044
313.15	-6.5324	0.3167	-0.0043

parameter, ξ_{12}. The MGCSP represents a significant improvement over the original GCSP since it results in reducing the number of several interaction parameters to one. The presence of many interaction parameters makes it correlative in nature and may result in large deviations if their value was set to unity. But at the same time, one can find the value of the interaction parameter, ξ_{12}, and the carbon number of the components of ternary n-alkane systems by using the correlation proposed by Wu (1992):

$$\xi_{12} = 0.8367 + 0.0328(N_2 - N_1) + 0.0426(N_3 - N_2) \qquad (2.39)$$

Following that, as the GCSP can be either correlative or predictive; the modified version which was tested by Wu and Asfour (1992) indicated that MGCSP is superior to the original GCSP.

As a final comment, the method suffers from two limitations when used, the first one is that it can only be used for n-alkane systems. The second limitation arises when trying to use equation (2.37) at other temperatures, as the constants were measured only at a certain temperature, i.e. the method is only applicable at the temperatures used for its development.

2.2.3 Reaction rate theory

This theory was developed by Eyring (1936) and his co-workers (Ewell and Eyring, 1937, Kincaid *et al.* 1941, Glasstone *et al.* 1941). This theory is one of the most popular theories because of its mathematical amenability. Ewell and Eyring (1937) considered viscous flow as a rate process to which the reaction rate theory was applied to. According to that theory, a single molecule is assumed to pass from one location to another over a potential energy barrier.

The theory depends on the idea of "holes". Eyring assumed that there is a shear between two layers of a liquid at a distance λ_1 between them, and then for the individual molecules to pass from one equilibrium position to another, a hole or an empty site must be available, as shown in Figure 2.1a. The molecule occupies, on the average, an area of $\lambda_2\ \lambda_3$. If there is no force affecting the liquid, there is molecular jumping to neighboring sites due to thermal activation. As there is no net flow, the rate of "jumping" in a certain direction must be equal to the rate of "jumping" in the opposite direction. For example, the rate of jumping to the left is equal to the rate of jumping to the right. For the molecule to go to another equilibrium position, it must cross a potential energy barrier ΔG_o, which is shown in Figure 2.1b. The frequency for both forward and backward molecule jumps, when there is no force acting on the liquid is given by:

$$r_o = \frac{KT}{h}\exp\left(-\frac{\Delta G_o}{KT}\right) \tag{2.40}$$

where K is Boltzmann's constant and h is Planck's constant.

Assume a shear stress, f, is applied on the liquid, so that the shear force on the molecule is equal to f $\lambda_2\lambda_3$. The only mechanical work resulting from this force is the work used to carry the molecule to the top of the potential energy barrier, and to reach equilibrium at the end, one assumes that the molecule gives all its energy as heat on the other side of the energy barrier. Consequently, the work done is:

$$\text{Work} = f\lambda_2\lambda_3\frac{\lambda}{2} \tag{2.41}$$

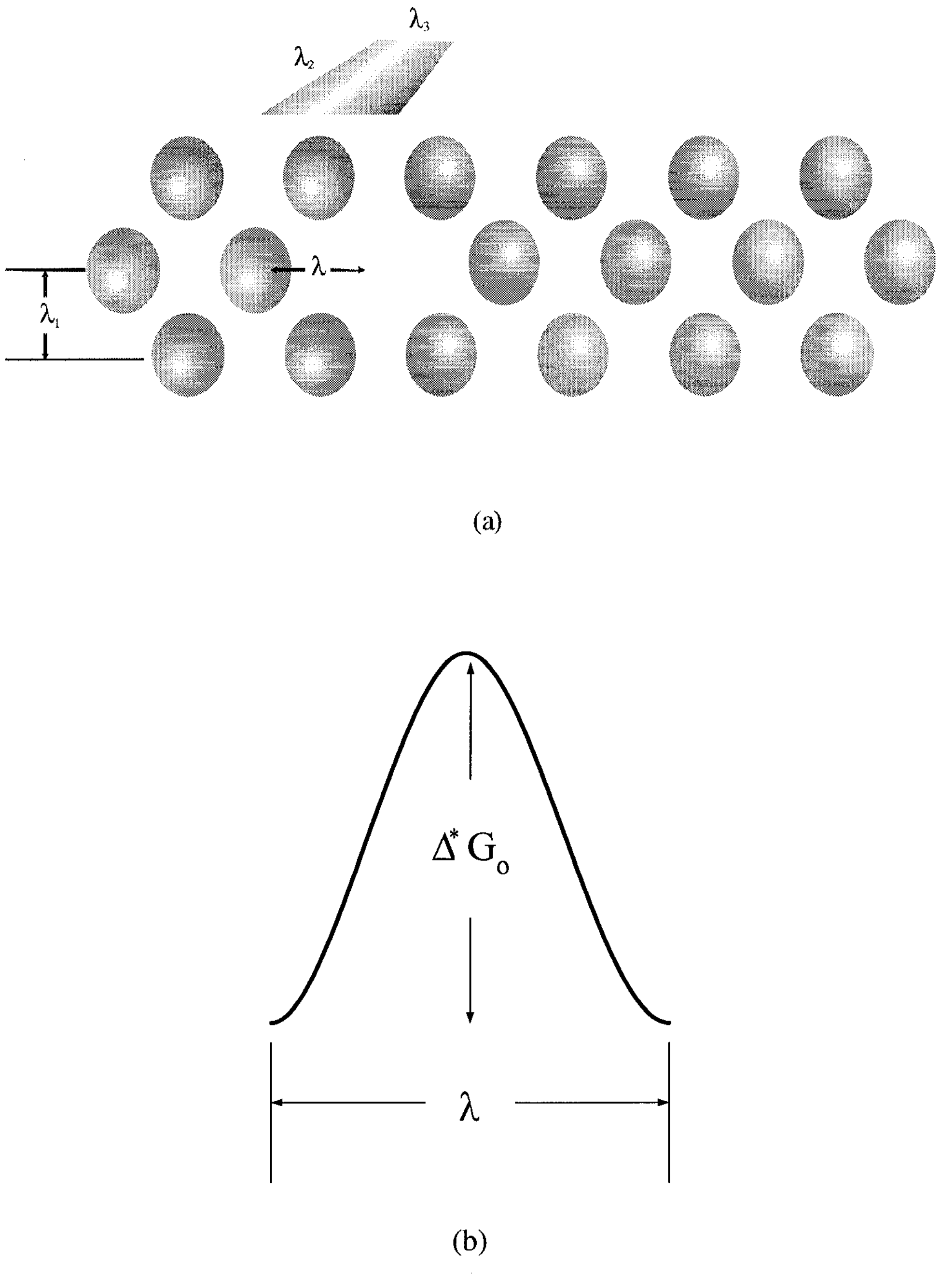

Figure 2.1: The Eyring Molecular Model of Liquid Viscosity

$$r_{\text{forward}} = \left(\frac{KT}{h}\right)\left[\exp\left(-\frac{\Delta G_o - f\lambda_2\lambda_3}{KT}\right)\right] \tag{2.42}$$

and the backward rate of jumping is

$$r_{\text{backward}} = \left(\frac{KT}{h}\right)\left[\exp\left(-\frac{\Delta G_o + f\lambda_2\lambda_3}{KT}\right)\right] \tag{2.43}$$

therefore, the net rate of jumping is

$$r_{\text{net}} = \left(\frac{KT}{h}\right)\exp\left(-\frac{\Delta G_o}{KT}\right)\left[\exp\left(\frac{f\lambda_2\lambda_3\lambda}{2KT}\right) - \exp\left(-\frac{f\lambda_2\lambda_3\lambda}{2KT}\right)\right] \tag{2.44}$$

the above equation can be simplified to give

$$r_{\text{net}} = \left(\frac{KT}{h}\right)\exp\left(-\frac{\Delta G_o}{KT}\right)\left[\exp\left(2\frac{f\lambda_2\lambda_3\lambda}{2KT}\right)\right] = \left(\frac{f\lambda_2\lambda_3\lambda}{h}\right)\exp\left(-\frac{\Delta G_o}{KT}\right) \tag{2.45}$$

the velocity gradient across the two molecular layers separated by a distance λ_1 is

$$\text{Velocity Gradient} = \frac{\text{Velocity Difference}}{\lambda_1}$$

$$\text{Velocity Gradient} = \frac{\text{Distance per jump x number of jumps per second}}{\lambda_1}$$

$$\text{Velocity Gradient} = \frac{\lambda r_{\text{net}}}{\lambda_1} \tag{2.46}$$

Since the viscosity η is defined as

$$\eta = \frac{\text{shear stress}}{\text{velocity gradient}} \tag{2.47}$$

Substituting equation (2.46) into equation (2.47) gives

$$\eta = \frac{f\lambda_1}{\lambda r_{\text{net}}} \tag{2.48}$$

Substituting equation (2.45) into equation (2.48) gives

$$\eta = \frac{h\lambda_1}{\lambda_2\lambda_3\lambda^2}\exp\left(\frac{\Delta G_o}{KT}\right) \tag{2.49}$$

Assuming $\lambda_1 = \lambda$ and using $\lambda_1 \lambda_2 \lambda_3$ as the effective volume of a molecule V_o, equation (2.49) becomes

$$\eta = \frac{h}{V_o} \exp\left(\frac{\Delta^* G_o}{RT}\right) \tag{2.50}$$

Equation (2.50) can be re-written as:

$$\eta = \frac{hN}{V_m} \exp\left(\frac{\Delta^* G_o}{RT}\right) \tag{2.51}$$

where N is Avogadro's number, V_m is the molar volume of the liquid and Δ^*G is the molar activation energy of viscous flow. The barrier energy Δ^*G_0 may also be assumed as the energy needed to create a hole for the molecule to drop in.

(i) McAllister's models

(a) Binary mixtures

The following form of Eyring equation was utilized by McAllister in the development of his binary-mixture model

$$\upsilon = \frac{hN}{M} \exp\left(\frac{\Delta^* G}{RT}\right) \tag{2.52}$$

For a binary mixture composed of components 1 and 2, as a molecule from 1 moves over the energy barrier, it may meet another molecule of 1, a molecule of 2, or a mixture of 1 and 2 depending on the local concentration. Also, the interaction can be assumed as a binary type, meaning a three-body type, four-body type, ... etc. The simplest of these forms of interaction is the three-body type assumed by McAllister (1960) as shown in Figure 2.2. According to Figure 2.2, there are six forms of interactions: 1-1-1, 2-2-2, 1-2-1, 2-1-2, 1-1-2, and 1-2-2.

Two assumptions must be stated here: (i) the free energy of activation of flow is additive, and (ii) the probability of interaction is proportional to the mole fractions of the species involved. Therefore, the free activation energy of the mixture can be expressed in the following general form:

$$\Delta * G = \sum_{i=1}^{2} \sum_{j=1}^{2} \sum_{k=1}^{2} X_i X_j X_k \Delta * G_{ijk} \tag{2.53}$$

where X is the mole fraction. Because it is difficult to differentiate between Δ^*G_{121} and Δ^*G_{112} and similar terms, the following equations were assumed, by McAllister, for that case:

$$\Delta * G_{121} = \Delta * G_{112} = \Delta * G_{12} \tag{2.54}$$

$$\Delta * G_{212} = \Delta * G_{122} = \Delta * G_{12} \tag{2.55}$$

By expanding equation (2.53) and substituting into it equations (2.54) and (2.55) yields

$$\Delta * G = X_1^3 \Delta * G_1 + 3X_1^2 X_2 \Delta * G_{12} + 3X_1 X_2^2 \Delta * G_{21} + X_2^3 \Delta * G_2 \tag{2.56}$$

Equation (2.56) indicates that each type of activation energy can be related to its corresponding kinematic viscosity in accordance with equation (2.52) as follows:

$$\nu = \left(\frac{hN}{M} \right) e^{\Delta * G/RT} \tag{2.57}$$

where

$$M = \sum_i X_i M_i \tag{2.58}$$

and, for pure component i

$$\nu_i = \left(\frac{hN}{M_i} \right) e^{\Delta * G_i/RT} \tag{2.59}$$

and for interactions

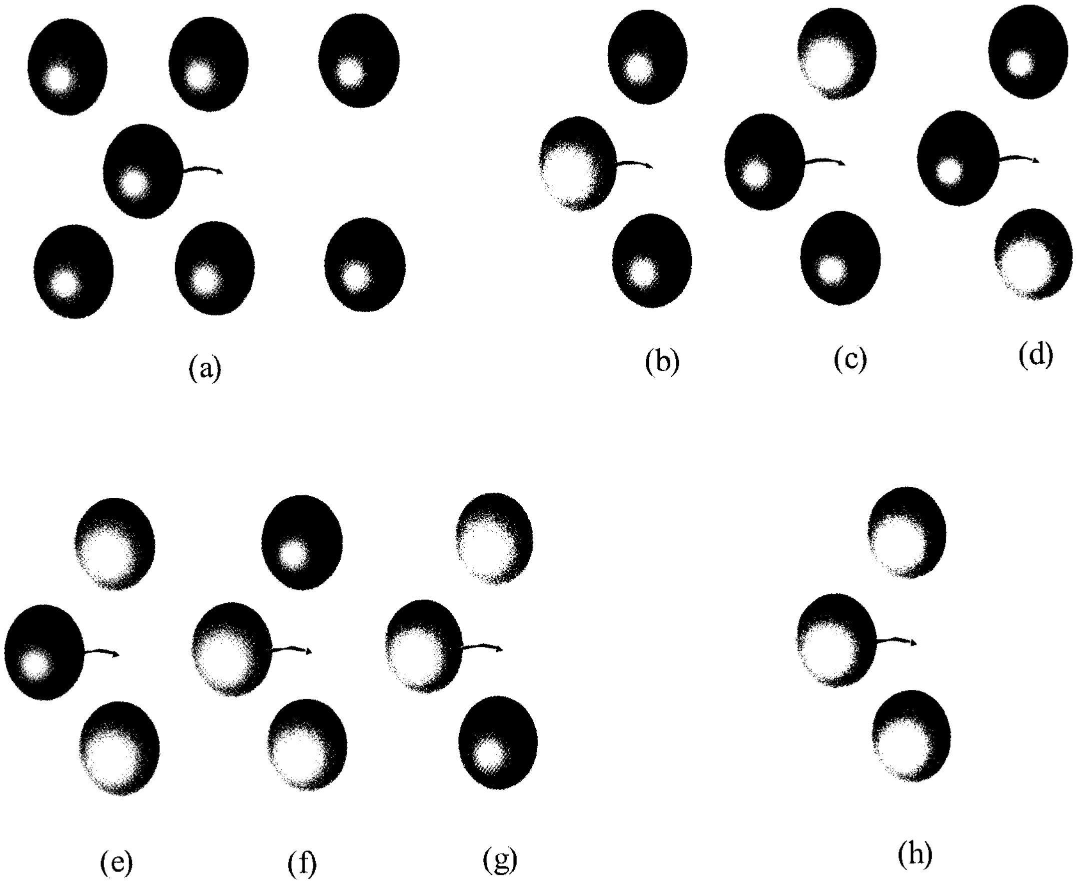

Figure 2.2: Types of Viscosity Interactions in a Binary Mixture; Three-body Collision Model

$$\nu_{ij} = \left(\frac{hN}{M_{ij}}\right) e^{\Delta^* G_{ij}/RT} \tag{2.60}$$

where

$$M_{ij} = \frac{2M_i + M_j}{3} \tag{2.61}$$

Substituting equation (2.56) into (2.57) yields

$$\nu = \left(\frac{hN}{M}\right) e^{\left(X_1^3 \Delta^* G_1 + 3X_1^2 X_2 \Delta^* G_{12} + 3X_1 X_2^2 \Delta^* G_{21} + X_2^3 \Delta^* G_2\right)/RT} \tag{2.62}$$

By substituting equations (2.59) and (2.60) into (2.62) and rearranging, one obtains the following three-body equation:

$$\ell n\nu = X_1^3 \ell n\nu_1 + 3X_1^2 X_2 \ell n\nu_{12} + 3X_1 X_2^2 \ell n\nu_{21} + X_2^3 \ell n\nu_2 - \ell n\left(X_1 + X_2 \frac{M_2}{M_1}\right)$$

$$+ 3X_1^2 X_2 \ell n\left(\frac{2 + \frac{M_2}{M_1}}{3}\right) + 3X_1 X_2^2 \ell n\left(\frac{1 + \frac{2M_2}{M_1}}{3}\right) + X_2^3 \ell n\left(\frac{M_2}{M_1}\right) \tag{2.63}$$

Equation (2.63) contains two adjustable parameters, ν_{12} and ν_{21}. The values of the parameters have to be determined from experimental data. Moreover, equation (2.63) is a cubic equation, which means it can have a maximum, a minimum, neither or both, and all these can be found on the viscosity-composition profile. It was reported by McAllister that the three body-models can give good results if the ratio of the molecular sizes of the components is less than 1.5.

Following similar procedures, McAllister developed a four-body model, to be used if the diameter of one molecule is much larger than the diameter of the other component molecule in the mixture. The McAllister four-body model is given by:

$$\ell n\nu = X_1^4 \ell n\nu_1 + 4X_1^3 X_2 \ell n\nu_{1112} + 6X_1^2 X_2^2 \ell n\nu_{1122} + 4X_1 X_2^3 \ell n\nu_{2221} + X_2^4 \ell n\nu_2$$

$$-\ell n\left(X_1 + X_2\frac{M_2}{M_1}\right) + 4X_1^3X_2\ell n\left(\frac{3+\frac{M_2}{M_1}}{4}\right) + 6X_1^2X_2^2\ell n\left(\frac{1+\frac{M_2}{M_1}}{2}\right)$$

$$4X_1X_2^3\ell n\left(\frac{3+\frac{M_2}{M_1}}{4}\right) + X_2^4\ell n\left(\frac{M_2}{M_1}\right) \tag{2.64}$$

The above equation has three parameters which have also to be determined from experimental data. This, of course reduces the usefulness of the McAllister model since it would need obtaining costly and time consuming experimental data equation.

(b) The Asfour *et al.* (1991) technique

Realizing that the McAllister equation is one of the best correlative methods for viscosity calculation of liquid mixtures, Asfour *et al.* (1991) developed a technique by which one can obtain the values of the parameters in McAllister model from the properties of the pure components and molecular parameters. As a first step, Asfour *et al.* (1991) successfully applied their technique to n-alkane mixtures.

Asfour *et al.* (1991) used viscosity data available from the literature on some n-alkane liquid mixtures at different temperature levels to calculate the values of the parameters ν_{12} and ν_{21} of the McAllister model. They Plotted the lumped parameter $\nu_{12}/(\nu_1^2\nu_2)^{1/3}$ versus the inverse of absolute temperature (1/T). The plot gave horizontal straight lines as shown in Figure 2.3. This provided clear evidence to them that the lumped parameter was independent of temperature.

Moreover, Asfour *et al.* (1991) plotted the lumped parameter $\nu_{12}/(\nu_1^2\ \nu_2)^{1/3}$ versus $[(N_2 - N_1)^2/(N_1^2 N_2)^{1/3}]$, where N_1 and N_2 are the number of carbon atoms per molecule of components 1 and 2, respectively. This resulted in the straight line as that shown in Figure 2.4. Asfour *et al.* (1991) reported the following equation for the straight line they had obtained:

$$\frac{\nu_{12}}{\left(\nu_1^2 \nu_2\right)^{1/3}} = 1 + 0.044 \frac{(N_2 - N_1)^2}{\left(N_1^2 N_2\right)^{1/3}} \qquad (2.65)$$

Using equation (2.65) it is possible to calculate the McAllister parameter ν_{12}. Asfour *et al.* (1991) also showed that the second McAllister parameter, ν_{21}, can be calculated from the following relationship:

$$\nu_{21} = \nu_{12}\left(\frac{\nu_2}{\nu_1}\right)^{1/3} \qquad (2.66)$$

where ν_{12} is the McAllister interaction parameter calculated by equation (2.65) and the viscosities of pure components 1 and 2, ν_1 and ν_2, respectively.

Asfour *et al.* (1991) recommended that when the difference between the numbers of carbon atoms of the two components in an n-alkane binary mixture exceeds 3, one should use the McAllister four-body collision model. This is consistent with the McAllister's (1960) recommendation that when the ratio of molecular size of the two components in a binary system exceeds 1.5; one should use the four-body collision model instead of the three-body collision model for better results.

Consequently, Asfour *et al.* (1991) employed the same rationale they used in case of the three-body collision model to develop similar equations for the prediction of the McAllister four-body collision model parameters. Therefore, when $|N_2 - N_1| \geq 4$, they reported the following equations:.

$$\frac{v_{1122}}{\left(v_1^2 v_2^2\right)^{1/4}} = 1 + 0.03\frac{\left(N_2 - N_1\right)^2}{\left(N_1^2 N_2^2\right)^{1/4}} \tag{2.67}$$

$$v_{1112} = v_{1122}\left(\frac{v_1}{v_2}\right)^{1/4} \tag{2.68}$$

$$v_{2221} = v_{1122}\left(\frac{v_2}{v_1}\right)^{1/4} \tag{2.69}$$

In light of the discussion given above, it is clear now that the technique reported by Asfour *et al.* (1991) converts the correlative McAllister model into a predictive model. This transformation results in significant savings in terms of time and cost when reliable liquid mixture viscosity values are needed for design purposes.

Asfour and Nhaesi (1998, 2000) extended the Asfour *et al.* (1991) technique to binary and multi-component liquid systems of regular solutions. Review of their work is in order.

(c) **Ternary mixtures**

Chandramouli and Laddha (1963) extended McAllister's three-body collision model to ternary liquid systems. However, their extended model was still correlative in nature, i.e., it needed experimental data for the determination of the values of the

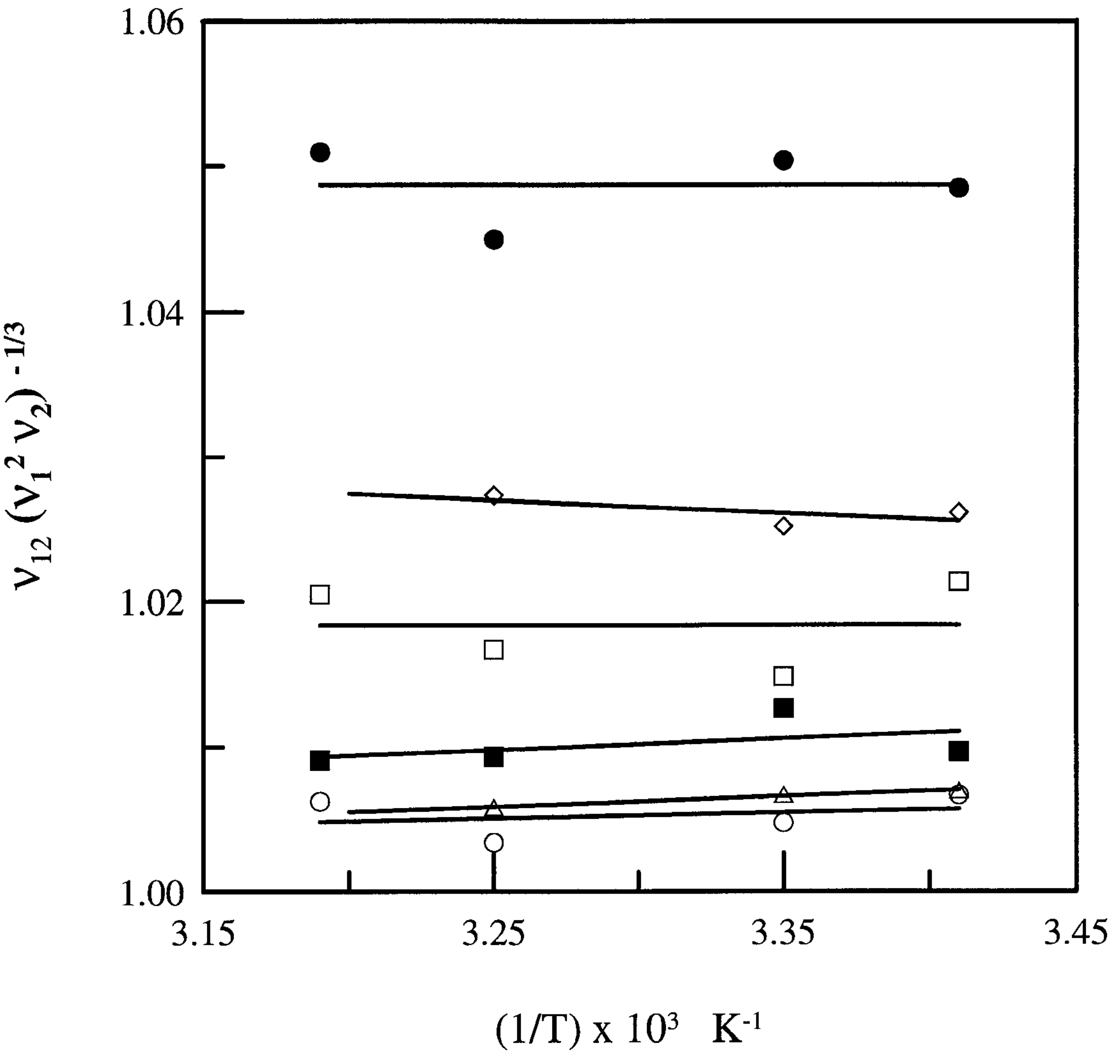

Figure 2.3: Variation of the Lumped Parameter $v_{12}/(v_1^2 v_2)^{1/3}$ with 1/T for n-Alkane systems for which $|N_2 - N_1| \le 3$ (data taken from Cooper, 1998).

△ Hexane (A) - Heptane (B)
◇ Hexane (A) - Octane (B)
○ Heptane (A) - Octane (B)
● Heptane (A) - Decane (B)
■ Tetradecane (A) - Hexadecane (B)
□ Octane (A) - Decane (B)

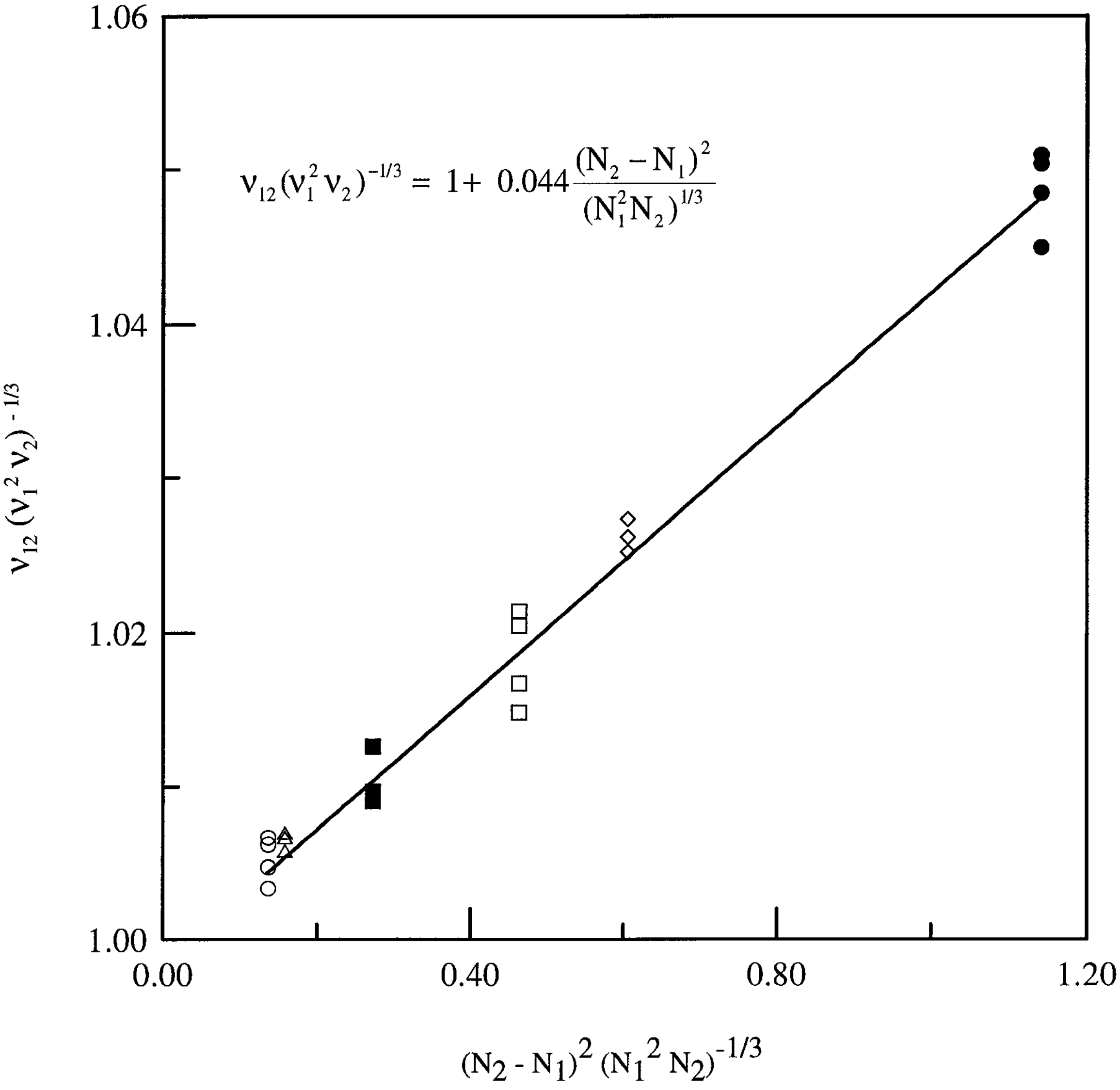

Figure 2.4: Variation of Lumped Parameter $\nu_{12}/(\nu_1^2\,\nu_2)^{1/3}$ with $[(N_2-N_1)^2/(N_1^2N_2)^{1/3}]$

(Legends as those in Figure 2.3)

adjustable parameters contained in the model. Kalidas and Laddha (1964) used experimental data to test the validity of the extended McAllister model which was developed by the Chandramouli and Laddha model. Figure 2.5 shows the types of viscosity interactions that may occur in case of a ternary system consisting of molecules of types1, 2 and 3. The Chandramouli and Laddha extended form of the McAllister model is given by equation (2.70).

$$
\begin{aligned}
\ell \mathrm{nv} = {} & X_1^3 \ell \mathrm{nv}_1 + X_2^3 \ell \mathrm{nv}_2 + X_3^2 \ell \mathrm{nv}_3 + 3X_1^2 X_2 \ell \mathrm{nv}_{12} + 3X_1^2 X_3 \ell \mathrm{nv}_{13} + 3X_2^2 X_1 \ell \mathrm{nv}_{21} \\
& + 3X_2^2 X_3 \ell \mathrm{nv}_{23} + 3X_3^2 X_1 \ell \mathrm{nv}_{31} + 3X_3^2 X_2 \ell \mathrm{nv}_{32} + 6X_1 X_2 X_3 \ell \mathrm{nv}_{123} \\
& - \ell n(X_1 M_1 + X_2 M_2 + X_3 M_3) + X_1^3 \ell \mathrm{n} M_1 + X_2^3 \ell \mathrm{n} M_2 + X_3^3 \ell \mathrm{n} M_3 \\
& + 3X_1^2 X_2 \ell \mathrm{n}\left(\frac{2M_1 + M_2}{3}\right) + 3X_1^2 X_3 \ell \mathrm{n}\left(\frac{2M_1 + M_3}{3}\right) \\
& + 3X_2^2 X_1 \ell \mathrm{n}\left(\frac{2M_2 + M_1}{3}\right) + 3X_2^2 X_3 \ell \mathrm{n}\left(\frac{2M_2 + M_3}{3}\right) \\
& + 3X_3^2 X_1 \ell \mathrm{n}\left(\frac{2M_3 + M_1}{3}\right) + 3X_3^2 X_2 \ell \mathrm{n}\left(\frac{2M_3 + M_2}{3}\right) \\
& + 6X_1 X_2 X_3 \ell \mathrm{n}\left(\frac{M_1 + M_1 + M_3}{3}\right)
\end{aligned}
\tag{2.70}
$$

where v_{12}, v_{21}, v_{13}, v_{31}, v_{23}, v_{32} are six binary interaction parameters and v_{123} is the ternary interaction parameter. Using the technique developed by Asfour *et al.* (1991), the six binary parameters can be determined as explained earlier in case of the binary interaction parameters. Asfour and Nhaesi (1998) reported the following equation for the calculation of the ternary parameter v_{123}.

$$
\frac{v_{123}}{(v_1 v_2 v_3)^{1/3}} = 0.9637 + 0.0313 \frac{(N_3 - N_1)^2}{N_2}
\tag{2.71}
$$

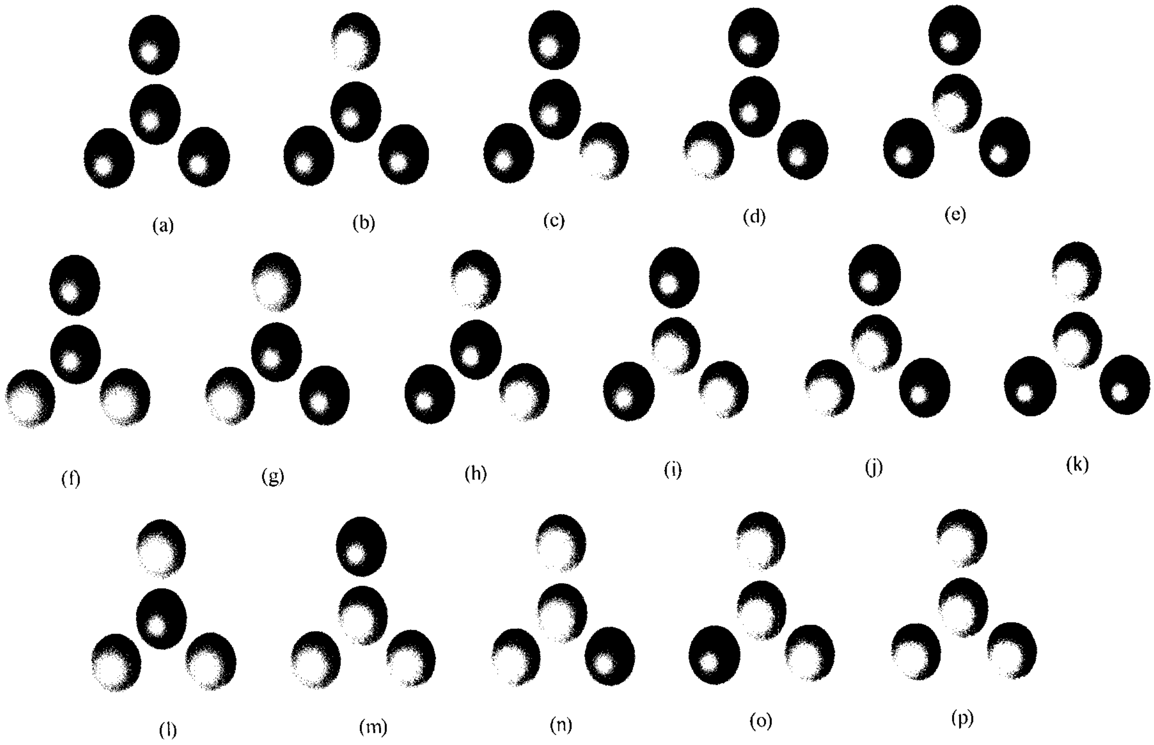

Figure 2.5: Types of Viscosity Interactions in a Binary Mixture; Four-body Collision Model

(d) The Generalized McAllister three-body model

Assuming that the interactions are three-body, and that the free energy of activations for viscous flow are additive on the basis of a mole fraction, Nhaesi and Asfour (2000a) reported the following equation:

$$\Delta G_m = \sum_{i=1}^{n} X_i^3 \Delta G_i + 3\sum_{i=1}^{n}\sum_{i=1}^{n} X_i^2 X_j \Delta G_{ij} + 6\sum_{i=1}^{n}\sum_{i=1}^{n}\sum_{k=1}^{n} X_i X_j X_k \Delta G_{ijk} \qquad (2.72)$$

They also assumed that :

$$\Delta G_{iji} = \Delta G_{iij} = \Delta G_{ij} \qquad (2.73)$$

$$\Delta G_{jij} = \Delta G_{ijj} = \Delta G_{ji} \qquad (2.74)$$

For each type of energy of activation involved in equation (2.72) one can assign a corresponding kinematic viscosity which can be expressed in the form of Arrhenius- type equation. For the mixture

$$\upsilon_m = \frac{hN}{M_{avg}} \exp\left(\frac{\Delta * G_m}{RT}\right) \qquad (2.75)$$

where

$$M_{avg} = \sum_{i=1}^{n} X_i M_i \qquad (2.76)$$

And, for pure component i

$$\upsilon_i = \frac{hN}{M_{avg}} \exp\left(\frac{\Delta * G_i}{RT}\right) \qquad (2.77)$$

Following the same procedure, one can show that for binary interactions:

$$\upsilon_{ij} = \frac{hN}{M_{ij}} \exp\left(\frac{\Delta * G_{ij}}{RT}\right) \qquad (2.78)$$

where

$$M_{ij} = \frac{(2M_i + M_j)}{3} \tag{2.79}$$

for ternary interactions:

$$\upsilon_{ijk} = \frac{hN}{M_{ijk}} \exp\left(\frac{\Delta * G_{ijk}}{RT}\right) \tag{2.80}$$

where

$$M_{ijk} = \frac{(M_i + M_j + M_k)}{3} \tag{2.81}$$

Taking the natural logarithms of equations (2.75), (2.77), (2.78), and (2.80), and substituting the resulting forms of those equations into equation (2.72) in order to eliminate the free energies of activation and rearranging, the Asfour-Nhaesi extended form of the McAllister three-body collision model for n-component liquid systems takes the following form:

$$\begin{aligned} \ell n v_m &= \sum_{i=1}^{n} X_i^3 \ell n(v_i M_i) + 3\sum_{i=1}^{n}\sum_{i=1}^{n} X_i^2 X_j \ell n(v_{ij} M_{ij}) \\ &+ 6\sum_{i=1}^{n}\sum_{i=1}^{n}\sum_{k=1}^{n} X_i X_j X_k \ell n(v_{ijk} M_{ijk}) - \ell n(M_{avg}) \end{aligned} \tag{2.82}$$

Using equation (2.82), one can predict the kinematic viscosity of an n-components regular solution by substituting the values of the binary and ternary adjustable parameters. The values of those parameters are calculated from the values of the kinematic viscosities of the pure components and their molecular parameters as outlined shown earlier. The number of binary parameters N_2, in a multi-component system, can be obtained from the following equation:

$$N_2 = \frac{n!}{(n-2)!} \tag{2.83}$$

where n is the number of components in the mixture. The number of ternary parameters N_3 can be obtained from the following equation:

$$N_3 = \frac{n!}{3!(n-3)!} \tag{2.84}$$

Nhaesi and Asfour (2000a) were first in reporting and using the extended form of the McAllister model for n-component system. They used an extensive database, from the literature as well as measured by them to test the validity of the extended form of the McAllister model, given by equation (2.82), which they reported. The results they reported showed excellent agreement with experimental data and indicated that the results of predicting they obtained by employing their technique were far superior to the results obtained by other models.

(e) The effective carbon number (ECN)

Nhaesi and Asfour (1998) reported a technique for calculating the effective carbon number. This enabled them to apply their technique which was described earlier in subsection (b) to regular solutions.

Nhaesi and Asfour (1998) prepared a semi-log plot for the kinematic viscosities of liquid pure n-alkane hydrocarbons at 308.15 versus the number of carbon atoms in each molecule. A straight line relationship was obtained, which is shown in Figure 2.6, using the least-squares technique. Nhaesi and Asfour (1998) indicated that the line shown in Fgure 2.6 can be represented by the following equation

$$\ell n(v) = A + B(N) \tag{2.85}$$

where A = - 1.943 and B = 0.193, and v in cSt.

According to Nhaesi and Asfour (1998), for any regular compound, if its kinematic viscosity at 308.15 is substituted in equation (2.85), one can obtain the ECN for that compound.

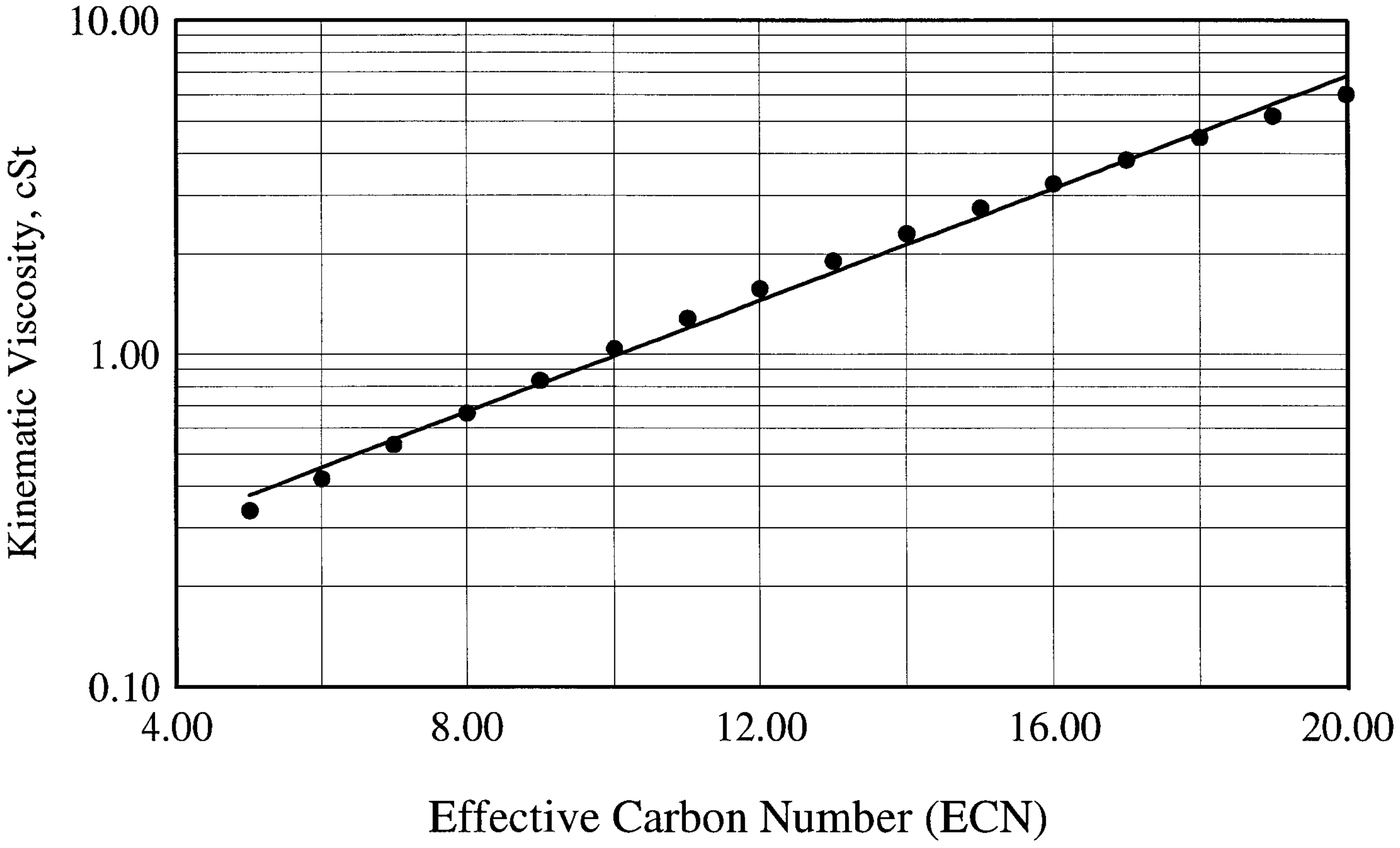

Figure 2.6: Experimental kinematic Viscosities for n-Alkanes at 308.15 K versus the Effective Carbon Numbers (Reported by Nhaesi, 1998).

This enabled Nhaesi and Asfour (1998) to calculate and report the ECN values for a variety of compounds, they also utilized these values of the ECNs to develop equations similar to equations 2.65 for regular solutions. They reported the following equation for regular solutions:

$$\frac{v_{12}}{\left(v_1^2 v_2\right)^{1/3}} = 0.8735 + 0.0715\frac{\left(ECN_2 - ECN_1\right)^2}{\left(ECN_1^2\ ECN_2\right)^{1/3}} \tag{2.86}$$

The values of v_{21} can still be calculated from equation (2.66).

For ternary parameters, they reported the following equation (Nhaesi and Asfour 2001a):

$$\frac{v_{123}}{\left(v_1 v_2 v_3\right)^{1/3}} = 0.9637 + 0.0313\frac{\left(ECN_3 - ECN_1\right)^2}{ECN_2} \tag{2.87}$$

The values of the Mcallister model parameters calculated with the help of equations (2.86) and (2.87) enables one to calculate the kinematic viscosity of a mixture by using equation (2.82).

(ii) The group contribution approach

The group contribution approach is based on the "additivity" of the contribution of each chemical group contained in a compound to its physical property. In other words, the physical property of a compound is the sum of the contributions made by the different chemical groups constituting that compound. In addition, it is assumed that the contributions made by a group have no relation to contributions made by another group. This method aims at utilizing existing data for predicting data for systems for which have no experimental data are available.

Many researchers as Redlich *et al.* (1959), Pierotti (1959), and Derr and Papandopoulos (1959) used the idea of group contribution for the estimation of heats of mixing. Langmuir (1925) originally suggested employing group contribution for the estimation of some thermodynamic properties of liquid mixtures. The group contribution method was also utilized by Wilson and Deal (1962) to correlate activity coefficients. This method was extended by Derr and Deal (1969) with their Analytical Solution of Groups (ASOG) method, and also by Ronc and Ratcliff (1971).

Abrams and Prausnitz (1975) proposed a model for activity coefficients based on the extension of the quasi-chemical theory of liquid mixtures (UNIQUAC). This model was combined with the concept of functional groups by Fredenslund *at al.* (1975) to predict the activity coefficients of non-ideal liquid mixtures. The model that resulted from that combination is known as the UNIFAC (UNIQUAC Functional-group Activity Coefficients). By combining the concepts of group contributions with the correlative UNIQUAC model, the predictive UNIFAC model was the result; this model has concentrations of the functional groups as the important independent variables rather than the concentrations of the molecules themselves.

In fact, viscosity and activity coefficients are two properties which are entirely different, since viscosity is a transport property and activity coefficient is a thermodynamic property. However, different models to calculate activity coefficients of liquid mixtures were utilized to predict the viscosity of liquid solutions.

One of the methods which depend on utilizing the direct proportionality between the activation energy in Eyring's theory and the Gibbs free energy was suggested by Wu (1986). The group contribution approach of UNIFAC was employed by Wu (1986) to convert his model from a correlative relationship into a predictive model.

Cao *et al.* (1992) used the Eyring reaction rate theory to develop a model by using statistical thermodynamics and local composition. That model can be used to correlate the viscosities of pure liquids and binary systems. On the basis of the results they obtained, those authors predicted the viscosities of multi-component liquid systems. They reported the following equation for predicting the kinematic viscosity:

$$\ell n(\nu M) = \sum_{i=1}^{n} X_i \ell n(\nu_i M_i) - \sum_{i=1}^{n} q_i n_i X_i \sum_{j=1}^{n} \theta_{ji} \ell n(\tau_{ji}) \tag{2.88}$$

In equation (2.88), q_i is the area parameter of molecule i, θ_{ij} is the local composition, τ_{ij} is the interaction parameter and n_i is a pure component parameter which is calculated as follows:

$$\ell n(n_i) = \sum_{j=0} A_j T^j \tag{2.89}$$

where the A_j's were obtained by correlating the experimental viscosities of 314 pure fluids.

For binary systems, there are two binary interaction parameters, namely, τ_{21} and τ_{12}, which are to be determined by fitting equation (2.88) to the experimental viscosity values of the binary systems under consideration. According to those authors, their method is correlative, as the interaction parameters have to be determined experimentally. But, if the parameters in equation (2.88) were obtained from properties of pure liquids and binary systems, the method can be considered predictive. When testing their method, those authors employed experimental data on 15 different multi-component systems. The resulting overall absolute deviation (%AAD) reported by those authors was about 2.9%. The model developed by Cao *et al.* (1992) was compared by those authors to the method reported by Wu (1986). The latter gave an overall %AAD of 5.7%, for the same systems. Furthermore, a new feature in the model reported by Cao *et al.* (1992) was the presence of the molecular size. Three equations were derived from the model, one for the viscosities of pure components, a second for the viscosities of liquid mixtures, and the last for the activity coefficients. The three equations were expressed by using the same parameters. The model was called "viscosity-thermodynamics" model (UNIMOD). The following equation was suggested for predicting the kinematic viscosity:

$$\ell n(\nu M) = \sum_{i=1}^{n} \varphi_i \ell n(\nu_i M_i) + 2\sum_{i=1}^{n} \varphi_i \ell n\left(\frac{X_i}{\varphi_i}\right) - \sum_{i=1}^{n} \frac{q_i n_i \varphi_i}{r_i} \sum_{j=1}^{n} \theta_{ji} \ell n(\tau_{ji}) \qquad (2.90)$$

In a follow-up paper by Cao *et al.* (1993b), the group-contribution concept was introduced into the viscosity equation of UNIMOD. That viscosity equation was divided into two parts: a combinatorial part and a residual part. The equation was expressed as follows:

$$\ell n(\nu) = \sum_{i=1}^{n} \left[\xi_i^c + \xi_i^R\right] \qquad (2.91)$$

$$\xi_i^c = \varphi_i \ell n\left[\nu_i \frac{M_i}{M}\right] + 2\varphi_i \ell n\left[\frac{X_i}{\varphi_i}\right] \qquad (2.92)$$

$$\xi_i^R = \sum_k \nu_k^i [\Theta_{ki} - \Theta_{ki}^i] \qquad (2.93)$$

In equation (2.91), ξ_i^c, which expresses only the properties of pure liquids, represents component i as a pure component, thus giving its viscosity contribution plus the excess viscosity contribution of component i in terms of size difference. ξ_i^R, which represents the interaction parameters between component i and the other components, gives the excess viscosity contribution of component i in terms of interaction differences. It should be noted here that the combinatorial part of the GC-UNIMOD, given by equation (2.91) has an exact correspondence with the combinatorial part of the UNIMOD for viscosity, given by equation (2.90). As for the residual part of the viscosity model, it used the same UNIFAC VLE model's group binary interaction parameters. The viscosity equation of the GC-UNIMOD was reported as:

$$\ell n(\nu) = \sum_{i=1}^{n} \left[\varphi_i \ell n\left(\nu_i \frac{M_i}{M} \right) + 2\varphi_i \ell n\left(\frac{X_i}{\varphi_i} \right) + \sum_{\text{all groups k}} \nu_k^{(i)} \left(\Theta_{ki} - \Theta_{ki}^{(i)} \right) \right] \qquad (2.94)$$

$$\Theta_{mi} = -\frac{Q_m}{R_m} N_{mi}^{vis} \varphi_i \sum_{\text{all groups k}} \theta_{km} \ell n(\Psi_{km}) \qquad (2.95)$$

where N_{ki}^{vis} is a group viscosity parameter for group k in component i:

$$N_{ki}^{vis} = Q_k \left(\frac{q_i - r_i}{2} - \frac{1 - r_i}{10} \right) \qquad (2.96)$$

from the pure group parameters Q_k and R_k, one can calculate the pure-molecule parameters, the area parameters q_i and the number of segments r_i as follows

$$q_i = \sum_{\text{all groups k}} \nu_k^{(i)} Q_k \qquad\qquad r_i = \sum_{\text{all groups k}} \nu_k^{(i)} R_k \qquad (2.97)$$

In the above equations, θ_k is the area fraction of group k in the mixture, whereas ν_k is the number of group k in molecule i. The local composition of the groups θ_{mn} and the group interaction parameter between groups m and n can be calculated from:

$$\theta_{mn} = \frac{\theta_m \Psi_{mn}}{\sum\limits_{\text{all groups } k} \theta_k \Psi_{kn}} \tag{2.98}$$

where

$$\theta_m = \frac{X_m Q_m}{\sum\limits_{\text{all groups } k} X_k Q_k} \qquad \Psi_i = \exp\left(-\frac{a_{mn}}{T}\right) \tag{2.99}$$

$$x_m = \frac{\sum\limits_j \nu_m^{(j)} X_j}{\sum\limits_j \sum\limits_n \nu_m^{(j)} X_j} \qquad \varphi_i = \frac{X_i r_i}{\sum\limits_j X_j r_j} \tag{2.100}$$

where in equation (2.99), a_{mn} is a group interaction energy parameter which is determined from experimental viscosity or VLE data.

Since data for the liquid mixtures are not needed, the viscosity calculations by the GC-UNIMOD are predictive in nature. As for testing the model, a large database of binary and ternary systems was used to test the GC-UNIMOD. The results obtained are considered good for a generalized predictive model.

As a general conclusion, group-contribution methods are generally approximate because the contribution of a given group in one molecule is not necessarily the same as its contribution in another molecule.

2.3 The Allan and Teja Correlation

The following Antoine-type equation was proposed by Allan and Teja (1991) for calculating the absolute viscosities of liquid systems.

$$\ell n(\eta) = A\left[-\frac{1}{B} + \frac{1}{T+C}\right] \tag{2.101}$$

where T is the temperature in degree Kelvin, η is the absolute viscosity in centipoise. A,B and C are correlated according to the number of carbon atoms in n-alkane by simultaneously fitting liquid viscosities of n-alkanes, ethane through eicosane. Alan and Teja (1991) reported the following equations for the calculation of the values of the parameters A, B, and C:

$$A = 145.73 + 99.01n + 0.83n^2 - 0.125n^3 \tag{2.102}$$

$$B = 30.48 + 34.04n - 1.23n^2 + 0.017n^3 \tag{2.103}$$

$$C = -3.07 - 1.99n \tag{2.104}$$

In order to determine the ECN (effective carbon number) for any hydrocarbon, a viscosity data point is needed. This method employs a simple mole averaging mixing rule for calculating the effective carbon number for hydrocarbon liquid mixtures, and hence the viscosities of of hydrocarbon liquid mixtures.

Allan and Teja (1991) compared the viscosity prediction results obtained from their correlation to those obtained by the TRAPP method. According to those authors the AAD of 5.6% for 10 binary systems was obtained, whereas for the TRAPP method an AAD of 9.5%, was obtained for the same systems. For components of ECN higher than 22, the method gives incorrect qualitative behavior and therefore should not be employed under such conditions (Gregory 1992).

CHAPTER 3

EXPERIMENTAL EQUIPMENT AND PROCEDURES

3.1 Materials

The pure compounds used in this study were purchased from Aldrich Chemical Company and from Fluka Chemika. The stated purities of all chemicals are 99+ mol%. The purities were verified by Gas Chromatography on an HP 5890 Gas Chromatograph equipped with an FID and an HP1 [cross linked methyl silicone gum] 30m (long) x 0.53 mm (diameter) and 2.65 μm (film thickness) column. The column is covered by US Patent #4,293,415. Table 3.1 reports all the pure chemicals used in this study, their supplier, and their stated purity and the purities as confirmed by the chromatographic analysis. As is clear from that table, the purities of all the chemicals used in this study exceeded their stated purities.

The pure chemicals used in this study were: octane, heptane, hexane, ethylbenzene, cyclohexane, and toluene. The densities and viscosities of the pure components were measured at the temperature levels employed in this study. Values of the densities and viscosities of the pure components and their comparison with the literature values are given in Chapter 4, Tables 4.5 and 4.6.

Double-distilled water was used, in addition to other pure chemicals, for the calibration of the density meter. The Double-distilled water was prepared in

Table 3.1: Specifications of the Chemicals used in this study.

Supplier	Compound	Specification	GC analysis, mass%
Aldrich Chemical Company	Pentane	99+%	99.8
	Hexane	99+%	99.7
	Heptane	99+%	99.9
	p-Xylene	99+%	99.7
	Ethylbenzene	99.80%	99.9
	Toluene	99.80%	99.9
	Cyclohexane	99+%	99.9
	Tri-decane	99+%	99.8
	Octane	99+%	99.5
Fluka Chima	Hexanol	>99% (GC)	99.1
	Heptanol	>99% (GC)	99.7

the laboratory starting with de-ionized water. The double-distilled water was prepared according to the procedure outlined earlier by Asfour (1979) and was kept in sealed glass vials until it was used.

3.2 Solution Preparation:

The solutions used in this study were prepared by weighing the different pure components constituting a solution in 30 ml glass vials purchased from Chromatographic Specialties Ltd. The vials were fitted with Tuf-bond Teflon/silicone discs and aluminum seals. Again, the Tuf-Bond discs and the aluminum seals were purchased from Chromatographic Specialties Ltd. Use of the vials, the Tuf-Bond dics and the aluminum seals was recommended by Asfour (1979) in order to minimize the evaporation of more volatile components of those solutions and hence result in change of composition.

The vials, before their use, were washed with detergent solution, de-ionized water and acetone then placed in an oven at 130° C to dry. The vials were then transferred to a desiccator where they were kept until their use.

The individual pure components were weighed on a Mettler HK 160 balance with a precision of $\pm 1 \times 10^{-7}$ kg. The component with the lower vapour pressure was injected into the vial first and weighed. After each component is injected into the vial, an equivalent volume of air is removed. The individual components were injected into the vials by means of 0.01 L hypodermic syringes fitted with size G23 and G24 needles. Samples from the composed solutions were taken by means of

0.002 L hypodermic syringes fitted with size G24 needles and injected into the density meter for density determination.

3.3 Density Measurement

An Anton Paar density meter was employed for measuring the densities of the components involved in this study as well as all the solutions prepared. A pictorial view of the density meter and the temperature controlled chamber is shown in Figure 3.1.

In the initial phase of this study, an Anton-Paar density meter consisting of a processing unit, DMA 60, and remote measuring cell, DMA 602, was employed for measuring the densities in this investigation. The DMA 60 contains the processor which calculates the change of the natural frequency of a hollow tube (contained in the DMA 602 unit) when filled with the different fluids. The change in the natural frequency changes as the mass of the fluid injected into the measuring cell is changed at a particular temperature. The period of vibration of the tube is displayed to 6 decimal places. The frequency of vibration is related to the density by the following three-parameter equation, which was recommended by the manufacturer:

$$\rho = \frac{A\tau^2}{1 - B\tau^2} - C \tag{3.1}$$

The parameters A, B, and C are determined through calibration by determining the frequency of vibration of compounds with known densities. Equation (3.1) is then fitted, by using least-squares, to the density values of those compounds, which are obtained from the literature. Calibration data of the density meter are given in Chapter 4, Tables 4.2 and 4.3.

Figure 3.1 : Pictorial View of the Anton-Paar Density Meter and the Constant Density Chamber

Since density of fluids is a strong function of temperature, the density meter is kept in a temperature controlled chamber that was designed and described in detail by Asfour (1979). In addition, the density meter's temperature is controlled by an N4B Haake circulator which is fitted with an IPTS-68 platinum temperature sensor. The temperature inside the water bath is displayed digitally. An Omega electronic thermometer fitted with a calibrated ITS-90 platinum temperature sensor was employed to confirm the temperature within the density meter. It was confirmed that the temperature fluctuations around the measuring cell of the density meter were better than ± 0.005 °C.

During the study, the measuring cell of the DMA 602 broke down. While it was being repaired, another measuring unit, DMA 602T, was calibrated and used to complete this study. The calibration results of the two unite, DMA 602 and DMA 602T, are reported in Chapter 4, Tables 4.2 and 4.3.

The density meter's measuring cell was cleaned by using ethanol and blowing dry air into the cell until a constant reading is obtained. The sample was then injected very slowly into the measuring cell – in order to ensure that no air bubbles were contained in the injected fluid - and was left, after adjusting the temperature to the required level, for at least 30 minutes to reach thermal equilibrium. Density measurements were done in triplicates and the average value of the frequency was reported. Appendix A reports the density meter readings. The density meter readings obtained were repeatable. The density meter precision is ± 1.5×10^{-5} g/cm^3.

3.2 Viscosity Measurement

The viscosities of pure components and samples were measured by using four Cannon-Ubbelohde viscometers. All four viscometers were of the size 25, which is used in the range of kinematic viscosities between 0.5 and 2 cSt. The viscometers used were having the following numbers: 25A483, 25A298, 25B349, 25B250. Such viscometers give high accuracy represented by a standard error of ±0.1%. The Cannon-Ubbelohde viscometer is shown Figure 3.2.

A CT-1000 constant temperature bath purchased from Cannon Instruments Company, which is specifically designed for viscosity measurements, was employed in this study. Figure 3.3 shows a pictorial view of the CT-1000 constant temperature bath. The CT-1000 bath is equipped with a temperature controller that is capable of keeping the temperature fluctuations to within ± 0.005 °C. This was confirmed by using an Omega digital thermometer fitted with a calibrated ITS-90 platinum temperature sensor.

The Cannon-Ubbelohde viscometers were cleaned by a special detergent solution purchased from Fisher Scientific Company, followed by acetone and placing in an oven for 10 minutes to dry. The viscometers were then calibrated by using calibration fluids supplied by Cannon Instruments Company. Calibration fluids N.4, N.8, and N1.0 were employed in the calibration of the viscometers used in this study. Results of the calibration are given in Chapter 4, Table 4.4.

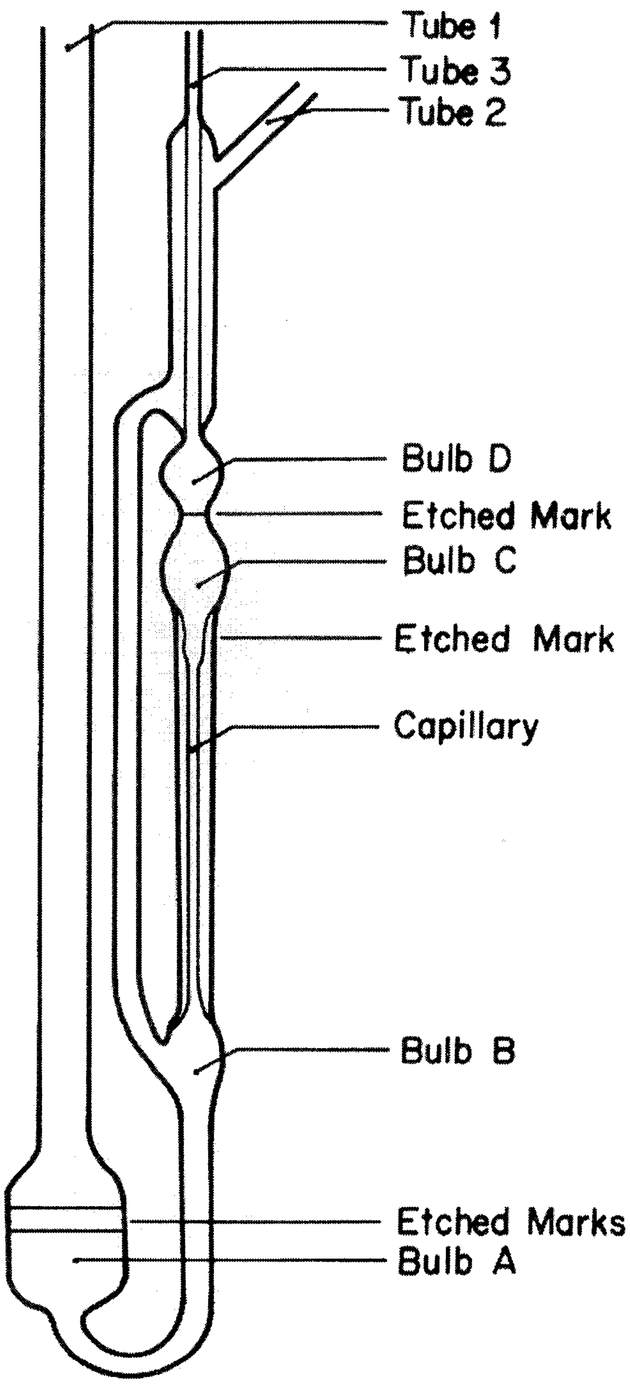

Figure 3.2: The Cannon-Ubbelohde Viscometer

Figure 3.3: Pictorial View of the CT-1000 Cannon Constant Temperature Bath and the Digital Thermometer

The following equation is utilized in the case of the Cannon-Ubbelohde viscometers:

$$\nu = Ct - \frac{E}{t^n} \qquad (3.2)$$

The above equation relates the kinematic viscosity, ν, to the efflux time, t. C and E are constants which can be determined from calibration. For such viscometers with trumpet shaped ends, the value of n is taken as 2.

Viscosity is measured by introducing the liquid of interest into bulb "A", Figure 3.2, where the liquid surface is kept between the etched marks. The viscometer is placed in the CT-1000 constant temperature bath, as shown in the pictorial view in Figure 3.3. The viscometer is kept in a vertical position by a plastic holder supplied by Cannon Instruments Company. The viscometer is left in the constant temperature bath for about 30 minutes to reach thermal equilibrium,

Tube 2 is closed with a finger and suction is applied to tube 3, by using a pipette suction bulb. This draws liquid into bulbs "C" and "D" without entrapping air bubbles into the liquid solution. Once the liquid level reaches the top of bulb "D", suction is removed and tubes 2 is opened. The liquid starts flowing down; when the liquid level reaches the first etched mark an electronic stop watch is clicked. When the liquid level reaches the second etched mark, at the bottom of bulb "C", the stop watch is clicked, again. This determines the time required by the liquid surface to travel between the two etched marks. This time is known as the efflux time. Measurements are carried out in triplicates and the average time is recorded and reported. The efflux times obtained in this study were repeatable to within ±0.1%.

CHAPTER 4

EXPERIMENTAL RESULTS AND DISCUSSION

4.1 General

Two quinary systems and some of their quaternary and ternary subsystems were investigated in this study at different temperature levels; viz., octane(1) - hexane(2) -ethylbenzene(3) - cyclohexane(4) - toluene(5) and octane(1) - heptane(2) - ethylbenzene(3) - cyclohexane(4) – toluene(5). The temperatures employed in the present study are 293.15 and 298.15 K. All the systems investigated in the present study are presented in Table 4.1.

The densities and kinematic viscosities of the systems indicated in Table 4.1 were measured at 293.15 and 298.15 K. The density and kinematic viscosity data were used to calculate the corresponding absolute viscosity values for the pure components as well as for the mixtures investigated. To the best of the present author's knowledge quinary and quaternary system data are very rarely found in the literature. Therefore, data reported in this study should be considered as a valuable addition to the literature of multi-component systems. In addition, the data were utilized in subjecting viscosity models, available from the literature to critical testing.

Table 4.1 : Systems investigated in the present study

1. Systems containing Hexane

(i) *Quinary System*

Octane (1) – Hexane (2) – Ethylbenzene (3) – Cyclohexane (4) –Toluene (5)

(ii) *Quaternary Subsystems*

Octane (1) – Hexane (2) – Ethylbenzene (3) – Cyclohexane (4)

Octane (1) – Hexane (2) – Ethylbenzene (3) -Toluene (4)

2. Systems containing Heptane

(i) *Quinary System*

Octane (1) – Heptane (2) – Ethylbenzene (3) – Cyclohexane (4) –Toluene (5

(ii) *Quaternary Subsystems*

Octane (1) - Heptane (2) – Ethylbenzene (3) – Cyclohexane (4)
Octane (1) - Heptane (2) - Ethylbenzene (3) –Toluene (4)
Octane (1) - Heptane (2) - Cyclohexane (3) –Toluene (4)
Heptane (1) – Ethylbenzene (2) – Cyclohexane (3) –Toluene (4)
Octane (1) - Ethylbenzene (2) – Cyclohexane (3) – Toluene (4)

(iii) *Ternary Subsystems*

Octane (1) – Heptane (2) – Ethylbenzene (3)
Octane (1) – Heptane (2) – Toluene (3)
Octane (1) – Cyclohexane (2)-Toluene (3)
Octane (1) – Ethylbenzene (2) –Toluene (3)
Octane (1) – Heptane (2) – Cyclohexane (3)
Heptane (1) – Ethylbenzene (2) - Cyclohexane(3)
Octane (1) – Ethylbenzene (2) – Cyclohexane (3)
Heptane (1) – Ethylbenzene (2) –Toluene (3)
Heptane (1) – Cyclohexane (2) – Toluene (3)
Ethylbenzene (1) – Cyclohexane (2) –Toluene (3)

4.2 Calibration Data of the Density Meters

Density measurements of the hexane-containing systems were carried out, as indicated earlier in Chapter 3, by using an Anton Paar density meter measuring unit DMA602, referred to herein as the "*first density meter*". However, during the course of the study, the measuring cell broke down and therefore, the measuring unit was replaced by another unit that was available in the laboratory, DMA602T, referred to herein as "*the second density meter*". The calibration data of the density meters employed in this study are given in Tables 4.2 and 4.3.

The first density meter was calibrated at 293.15 K and 298.15 K by using pentane, heptane, tridecane, p-xylene, hexanol, heptanol and double distilled water. All the liquids were used at both temperatures.

The materials used for the calibration of the second density meter were: double distilled water, tridecane, p-xylene, hexanol, heptanol, N0.4, N0.8, N1.0. The last three substances were viscometer calibration fluids supplied by Cannon Instruments Company. The values of the densities of these calibration fluids were supplied by the vendor.

Table 4.2: Calibration Data for the First Density Meter

Substance	Temperature = 293.15 K		Source
	Density, kg/L	Density Meter Reading, seconds	
Pentane	0.6261	0.688061	TRC
Heptane	0.68375	0.699237	TRC
Tridencane	0.7561	0.712946	TRC
P-Xylene	0.861	0.732339	Timmermans 1950
DDWater	0.998234	0.757032	Perry, 1997
Hexanol	0.8198	0.724613	TRC
Heptanol	0.8223	0.725178	TRC

<u>Parameters of the Density Meter</u> (equation 3.1)

A = 3.899617

B =- 0.04134048

C = 1.184867

Standard Deviation of Fit (kg/L) = 3.15×10^{-4}

Substance	Temperature = 298.15 K		Source
	Density, kg/L	Density Meter Reading, seconds	
Pentane	0.6212	0.686877	TRC
Heptane	0.67946	0.698204	TRC
Tridencane	0.75271	0.712034	TRC
P-Xylene	0.85666	0.731270	Timmermans 1950
DDWater	0.99707	0.756572	Perry, 1997
Hexanol	0.8162	0.723730	TRC
Heptanol	0.8186	0.724288	TRC

<u>Parameters of the Density Meter</u> (equation 3.1)

A =3.892602

B =- 0.03890530

C =1.182467

Standard Deviation of Fit (kg/L)= 2.6×10^{-4}

Table 4.3: Calibration Data for the Second Density Meter

Substance	Temperature = 293.15 K		Source
	Density, kg/L	Density Meter Reading, seconds	
Tridencane	0.7561	0.703272	TRC
P-Xylene	0.861	0.722557	Timmermans 1950
DDWater	0.9982343	0.747092	Perry, 1997
Hexanol	0.8198	0.714833	TRC
Heptanol	0.8223	0.715480	TRC
N0.4	0.6681	0.686588	Cannon Instr. Co.
N0.8	0.8664	0.723537	Cannon Instr. Co.
N1.0	0.7302	0.698377	Cannon Instr. Co.

Parameters of the Density Meter (equation 3.1)

A = 3.951236

B =- 0.03650930

C = 0.163113

Standard Deviation of Fit (kg/L)= 3.5 x10^{-4}

Substance	Temperature = 298.15 K		Source
	Density, kg/L	Density Meter Reading, seconds	
Tridencane	0.7527	0.702371	TRC
P-Xylene	0.8567	0.721528	Timmermans 1950
DDWater	0.99707	0.746639	Perry, 1997
Hexanol	0.8162	0.713940	TRC
Heptanol	0.8186	0.714599	TRC
N0.4	0.6635	0.685488	Cannon Instr. Co.
N0.8	0.8621	0.722523	Cannon Instr. Co.
N1.0	0.7264	0.697438	Cannon Instr. Co.

Parameters of the Density Meter (equation 3.1)

A = 3.939776

B =- 0.03300510

C =1.159579

Standard Deviation of Fit (kg/L)=3.46 x 10^{-4}

4.3 Calibration Data of the Viscometers

The four Cannon-Ubbelohde viscometers which were employed in the present study were calibrated by using the viscometer standards that were purchased from Cannon Instruments Company. The three viscosity standards cover the viscosity ranges of all solutions employed in this study. For all the viscosity measurements that were carried out, each experiment was repeated at least three times. Efflux times measured were repeatable to within ± 0.1%.

The two parameters, included in equation (3.3), were determined by the least-squares technique by fitting the calibration data to equation (3.3). Table 4.4 reports the values of the calculated parameters, standard deviations and the corresponding %AAD.

4.4 Accuracy and Precision of the Density and Viscosity Measurements

The density measurements reported in the present study were based on the density meter readings obtained when the solutions investigated were injected into the density meter's measuring cell. The density meter readings represent the period of oscillation of a hollow U-tube that is filled with the sample solution under investigation. The density meter is capable of measuring densities with a reproducibility of $\pm\ 1.5 \times 10^{-5}$ kg/L as indicated by the manufacturer.

The accuracy of the density measurement is subject to the accuracy of the densities of the fluids used in its calibration. The first density meter was calibrated

Table 4.4:Calibration Data for the Viscometers at 293.15 K

Viscometer	Viscosity Standard	Efflux Time (seconds)	Standard Viscosity x 10^6 m^2/s	Parameters		Standard Deviation x10^9 m^2/s	% AAD
				C x 10^9 (m/s^2)	E x 10^6 ($m^2.s$)		
25A483	N 0.4	266.57	0.483	1.8081	-74.0539	0.0347	0.00
	N 0.8	422.95	0.7652				
	N 1.0	699.57	1.265				
25A498	N 0.4	261.09	0.483	1.8487	-4.8832	0.5311	0.06
	N 0.8	414.33	0.7652				
	N 1.0	684.06	1.265				
25B349	N 0.4	275.29	0.483	1.7622	346.674	3.3879	0.38
	N 0.8	436.23	0.7652				
	N 1.0	715.36	1.265				
25B350	N 0.4	278.04	0.483	1.7430	175.577	1.3397	0.16
	N 0.8	440.69	0.7652				
	N 1.0	725.41	1.265				

Table 4.4 (Cont'd.): Calibration Data for the Viscometers at 298.15 K

Viscometer	Viscosity Standard	Efflux Time (s)	Standard Viscosity x 10^6 m^2/s	Parameters		Standard Deviation x10^9 m^2/s	% AAD
				C x 10^9 (m/s^2)	E x 10^6 ($m^2.s$)		
25A483	N 0.4	255.84	0.4618	1.8068	51.3873	0.7007	0.9
	N 0.8	400.26	0.7218				
	N 1.0	651.23	1.177				
25A498	N 0.4	250.33	0.4618	1.8476	80.8614	1.1669	0.15
	N 0.8	391.90	0.7218				
	N 1.0	636.69	1.177				
25B349	N 0.4	263.48	0.4618	1.7617	222.4454	1.6644	0.21
	N 0.8	411.87	0.7218				
	N 1.0	667.70	1.177				
25B350	N 0.4	266.33	0.4618	1.7372	89.5974	0.7883	0.10
	N 0.8	416.47	0.7218				
	N 1.0	677.31	1.177				

by using seven fluids with accurately known densities, whereas the second density meter was calibrated with eight fluids with accurately known densities. The densities of the pure components constituting the systems reported in the present study were compared to the densities of these components as reported in the literature. The comparisons are given in Tables 4.5 and 4.6.

The viscosities of the pure components employed in the present study were also compared to those reported in the literature. The comparisons are given in Tables 4.5 and 4.6. The viscosity values obtained in the present study compare very favourably with those reported in the literature.

It is also clear from Tables 4.5 and Table 4.6 that the densities of the pure components measured in the present study are in excellent agreement with the corresponding values reported in the literature.

The insignificant differences between the density and viscosity values reported in this study and those reported in the literature, may be attributed to the differences in the purities of the pure compounds employed in this study and those reported in the literature. Whereas the purities of pure components employed in this study were generally higher than 99%, purities of the corresponding compounds reported in the literature are not readily available.

In addition, densities and viscosities are functions of temperature. In the present study, temperature fluctuations were kept strictly to within ± 0.01 K. An

Table 4.5: Physical Properties of Pure Components at 293.15 K

Compound	Density, kg/L			Kinematic Viscosity x 10^6 m^2/s			Absolute Viscosity x 10^3 Pa.s		
	Experimental Value	Literature Value		Experimental Value	Literature Value		Experimental Value	Literature Value	
		Minimum	Maximum		Minimum	Maximum		Minimum	Maximum
Temperature = 293.15 K									
Hexane	0.6954	0.65925 [1]	0.6595 [4]	0.4704	-	0.4727 [1]	0.3271	0.3117 [1]	0.318 [4]
Heptane	0.6840	0.6836 [4]	0.68375 [1]	0.6023	-	0.6096 [1]	0.4120	0.409 [4]	0.4169 [1]
Octane	0.7025	0.7022 [4]	0.70267 [1]	0.7734	-	0.7758 [1]	0.5433	0.5450 [1]	0.5479 [8]
Toluene	0.8667	0.8666 [3]	0.8760 [2]	0.6797	0.6747 [1]	0.6786 [16]	0.5891	-	0.5848 [1]
Ethylbenzene	0.8671	0.8670 [1]	0.8672 [4]	0.7767	-	0.7800 [1]	0.6735	-	0.6763 [1]
Cyclohexane	0.7787	0.7783[4]	0.77856 [5]	1.2560	-	-	0.9780	-	-

Table 4.6: Physical Properties of Pure Components at 298.15 K

Compound	Density, kg/L			Kinematic Viscosity x 10^6 m^2/s			Absolute Viscosity x 10^3 Pa.s		
	Experimental Value	Literature Value		Experimental Value	Literature Value		Experimental Value	Literature Value	
		Minimum	Maximum		Minimum	Maximum		Minimum	Maximum
Temperature = 298.15 K									
Hexane	0.6549	0.65471 [1]	0.65502 [4]	0.4489	-	0.4545 [1]	0.2940	0.2937 [4]	0.3150 [14]
Heptane	0.6797	0.6793 [6]	0.67963 [4]	0.5731	-	0.5821 [1]	0.3895	0.3855 [4]	0.4040 [14]
Octane	0.6985	0.67849 [6]	0.69862 [1]	0.7313	-	0.7352 [1]	0.5108	0.5136 [1]	0.5030 [14]
Toluene	0.8620	0.8610 [7]	0.86232 [8]	0.6413	0.6378 [1]	0.6421 [12]	0.5528	0.5500 [1]	0.5560 [15]
Ethylbenzene	0.8627	0.8621 [9]	0.8630 [10]	0.7323	-	0.7367 [1]	0.6318	-	0.6345 [1]
Cyclohexane	0.7739	0.7711 [11]	0.77399 [8]	1.1540	-	-	0.8931	0.8770 [13]	0.900 [8]

1. TRC 1986
2. Katayama *et al.* 1980
3. American Chemical Society 1959
4. Timmermans 1950
5. Lien *et al.* 2003
6. Nayak *et al.* 2003
7. Sanni *et al.* 1971
8. Timmermans 1965
9. Hafez *et al.* 1976
10. Vitali and Lobbia 1983
11. Gomez-Diaz *et al.* 2001
12. Ghai 1973
13. Aminbhavi 1996
14. Nayak *et al.* 2003
15. Ritzoulis *et al.* 1986
16. McAllister (1960)

ITS-90, RTD temperature measuring probe connected to a digital thermometer was employed in the present study for temperature measurement. Literature sources do not generally include such fine technical details and this makes the present author believe that the densities and viscosities reported in this study are superior to those reported in the literature in terms of their accuracy.

4.5 Density-Composition Data

Density measurements were carried out in, at least, triplicates with the density meter readings only differing by a few units in the last decimal place between successive readings. The average value of the density meter readings were determined and are reported at each composition in Appendix A. The density meter readings were used in equation (3.1) in order to determine the density of the solution at the composition investigated. The density composition data obtained are reported in Tables 4.7 through 4.10.

4.6 Kinematic Viscosity-Composition Data

Kinematic viscosity-composition data, over the entire composition range, were determined and are reported in Tables 4.7 through 4.10. The absolute viscosity values were obtained from the corresponding kinematic viscosity and density data. Those absolute viscosity values are also reported in Tables 4.7 through 4.11.

Table 4.7: Densities and Viscosities of the Quinary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	X_5	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.0000	0.6594	0.4704	0.3102
0.0000	0.0000	1.0000	0.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	0.0000	1.0000	0.0000	0.7787	1.256	0.9780
0.0000	0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.0986	0.2921	0.4888	0.0493	0.0712	0.7802	0.6316	0.4928
0.1967	0.1433	0.0518	0.1036	0.5046	0.7824	0.64	0.5007
0.1511	0.4904	0.0492	0.1006	0.2087	0.7233	0.5717	0.4135
0.4932	0.1042	0.0563	0.1990	0.1473	0.7355	0.7195	0.5292
0.1514	0.2555	0.1960	0.0531	0.3440	0.7741	0.6157	0.4766
0.1986	0.1022	0.0476	0.4990	0.1526	0.7588	0.7789	0.5910

Table 4.7 (Cont'd): Densities and Viscosities of the Quinary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	X_5	Density (kg/L)	Kinematic Viscosity $x10^6$ (m^2/s)	Absolute Viscosity $x10^3$ (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.0000	0.6549	0.4489	0.2940
0.0000	0.0000	1.0000	0.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	0.0000	1.0000	0.0000	0.7739	1.154	0.8931
0.0000	0.0000	0.0000	0.0000	1.0000	0.862	0.6413	0.5528
0.0986	0.2921	0.4888	0.0493	0.0712	0.7758	0.5988	0.4645
0.1967	0.1433	0.0518	0.1036	0.5046	0.7779	0.6083	0.4732
0.1511	0.4904	0.0492	0.1006	0.2087	0.7189	0.5447	0.3916
0.4932	0.1042	0.0563	0.1990	0.1473	0.7313	0.6804	0.4976
0.1514	0.2555	0.1960	0.0531	0.3440	0.7697	0.5853	0.4505
0.1986	0.1022	0.0476	0.4990	0.1526	0.7543	0.7349	0.5543

Table 4.8: Densities and Viscosities of the Quaternary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)
Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.6594	0.4704	0.3102
0.0000	0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	0.0000	1.0000	0.7787	1.256	0.9780
0.1019	0.4972	0.1537	0.2472	0.7202	0.6094	0.4389
0.1562	0.1977	0.5956	0.0505	0.7893	0.6697	0.5286
0.0747	0.2779	0.4496	0.1978	0.7748	0.6605	0.5118
0.5816	0.0474	0.1887	0.1823	0.7362	0.7537	0.5549
0.1029	0.0458	0.2476	0.6037	0.7814	0.857	0.6697
0.0530	0.6952	0.0955	0.1563	0.6969	0.5462	0.3806
0.4429	0.1540	0.1950	0.2081	0.7357	0.7215	0.5308
0.0508	0.0987	0.2852	0.5653	0.7837	0.8146	0.6384

Table 4.8 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.6549	0.4489	0.2940
0.0000	0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	0.0000	1.0000	0.7739	1.154	0.8931
0.1019	0.4972	0.1537	0.2472	0.7158	0.5786	0.4142
0.1562	0.1977	0.5956	0.0505	0.7849	0.6352	0.4986
0.0747	0.2779	0.4496	0.1978	0.7704	0.6256	0.4820
0.5816	0.0474	0.1887	0.1823	0.732	0.711	0.5205
0.1029	0.0458	0.2476	0.6037	0.7769	0.8044	0.6249
0.0530	0.6952	0.0955	0.1563	0.6924	0.5197	0.3598
0.4429	0.1540	0.1950	0.2081	0.7315	0.684	0.5003
0.0508	0.0987	0.2852	0.5653	0.7792	0.7668	0.5975

Table 4.8 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.6594	0.4704	0.3102
0.0000	0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.1013	0.4867	0.1688	0.2432	0.7416	0.5556	0.4120
0.1477	0.1973	0.6085	0.0465	0.7952	0.6638	0.5279
0.0508	0.2970	0.4658	0.1864	0.7916	0.6177	0.4890
0.4652	0.0581	0.2162	0.2605	0.764	0.6838	0.5224
0.1054	0.0597	0.2008	0.6341	0.8287	0.6675	0.5532
0.0575	0.5928	0.1029	0.2468	0.7272	0.5253	0.3820
0.4513	0.1506	0.2029	0.1952	0.7499	0.663	0.4972
0.0523	0.1054	0.2870	0.5553	0.8304	0.6566	0.5452

Table 4.8 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity $\times 10^6$ (m^2/s)	Absolute Viscosity $\times 10^3$ (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.6549	0.4489	0.2940
0.0000	0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	0.0000	1.0000	0.862	0.6413	0.5528
0.1013	0.4867	0.1688	0.2432	0.7371	0.5281	0.3893
0.1477	0.1973	0.6085	0.0465	0.7908	0.6283	0.4969
0.0508	0.2970	0.4658	0.1864	0.7872	0.5863	0.4615
0.4652	0.0581	0.2162	0.2605	0.7597	0.6488	0.4929
0.1054	0.0597	0.2008	0.6341	0.8242	0.6317	0.5206
0.0575	0.5928	0.1029	0.2468	0.7227	0.5197	0.3756
0.4513	0.1506	0.2029	0.1952	0.7457	0.6297	0.4696
0.0523	0.1054	0.2870	0.5553	0.8259	0.6291	0.5196

Table 4.9: Densities and Viscosities of the Quinary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	X_5	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.0000	0.0000	0.8671	0.7761	0.6730
0.0000	0.0000	0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.1109	0.2602	0.5324	0.0604	0.0360	0.7867	0.6834	0.5376
0.2002	0.1371	0.0580	0.1022	0.5024	0.7851	0.6651	0.5222
0.1446	0.4515	0.1479	0.3373	0.2145	0.7339	0.6428	0.4718
0.4967	0.0917	0.0548	0.1982	0.1586	0.7379	0.7391	0.5454
0.1450	0.2181	0.1763	0.0657	0.3949	0.7612	0.5414	0.4121
0.2087	0.0904	0.0541	0.4948	0.1519	0.7378	0.6435	0.4748

Table 4.9 (Cont'd): Densities and Viscosities of the Quinary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	X_5	Density (kg/L)	Kinematic Viscosity $x10^6$ (m^2/s)	Absolute Viscosity $x10^3$ (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.1109	0.2602	0.5324	0.0604	0.0360	0.7823	0.6467	0.5059
0.2002	0.1371	0.0580	0.1022	0.5024	0.7805	0.6304	0.4920
0.1446	-0.4515	0.1479	0.3373	0.2145	0.7296	0.6101	0.4451
0.4967	0.0917	0.0548	0.1982	0.1586	0.7337	0.6981	0.5122
0.1450	0.2181	0.1763	0.0657	0.3949	0.7680	0.5657	0.4345
0.2087	0.0904	0.0541	0.4948	0.1519	0.7447	0.6781	0.5050

Table 4.10: Densities and Viscosities of the Quaternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)
Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	0.0000	1.0000	0.7787	1.2560	0.9780
0.1110	0.4737	0.1459	0.2694	0.7299	0.6979	0.5094
0.1611	0.1868	0.5878	0.0644	0.7912	0.7014	0.5549
0.0600	0.2772	0.4558	0.2070	0.7791	0.7027	0.5475
0.6018	0.0486	0.1930	0.1566	0.7358	0.7572	0.5571
0.1086	0.0476	0.2520	0.5918	0.7815	0.8587	0.6711
0.0582	0.6711	0.1134	0.1573	0.7144	0.6495	0.4640
0.4618	0.1427	0.2064	0.1890	0.7386	0.7432	0.5489
0.0546	0.0923	0.3040	0.5491	0.7869	0.8277	0.6513

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	0.0000	1.0000	0.7739	1.1540	0.8931
0.1110	0.4737	0.1459	0.2694	0.7256	0.6603	0.4791
0.1611	0.1868	0.5878	0.0644	0.7868	0.6644	0.5227
0.0600	0.2772	0.4558	0.2070	0.7747	0.6650	0.5152
0.6018	0.0486	0.1930	0.1566	0.7316	0.7157	0.5236
0.1086	0.0476	0.2520	0.5918	0.7770	0.8095	0.6290
0.0582	0.6711	0.1134	0.1573	0.7101	0.6166	0.4378
0.4618	0.1427	0.2064	0.1890	0.7344	0.7025	0.5159
0.0546	0.0923	0.3040	0.5491	0.7824	0.7795	0.6099

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.0000	0.8671	0.7761	0.6730
0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.0691	0.4838	0.1711	0.2760	0.7520	0.6225	0.4681
0.1515	0.1760	0.6127	0.0597	0.8000	0.6927	0.5542
0.0548	0.2728	0.5925	0.0798	0.7993	0.6623	0.5294
0.4983	0.0474	0.1958	0.2584	0.7611	0.6952	0.5291
0.1000	0.0452	0.1972	0.6576	0.8331	0.6768	0.5638
0.0599	0.5567	0.1134	0.2700	0.7430	0.6079	0.4517
0.4517	0.1317	0.2104	0.2062	0.7549	0.6852	0.5173
0.0537	0.0923	0.3007	0.5533	0.8335	0.6757	0.5632

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.0691	0.4838	0.1711	0.2760	0.7476	0.5912	0.4420
0.1515	0.1760	0.6127	0.0597	0.7957	0.6557	0.5217
0.0548	0.2728	0.5925	0.0798	0.7949	0.6291	0.5001
0.4983	0.0474	0.1958	0.2584	0.7569	0.6586	0.4985
0.1000	0.0452	0.1972	0.6576	0.8286	0.6383	0.5289
0.0599	0.5567	0.1134	0.2700	0.7386	0.5780	0.4269
0.4517	0.1317	0.2104	0.2062	0.7506	0.6502	0.4880
0.0537	0.0923	0.3007	0.5533	0.8290	0.6399	0.5305

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.1124	0.4611	0.1661	0.2603	0.7363	0.6502	0.4787
0.1523	0.0919	0.6046	0.1512	0.7608	0.8438	0.6420
0.0539	0.2613	0.4757	0.2091	0.7563	0.7476	0.5654
0.4017	0.0457	0.2010	0.3515	0.7582	0.7164	0.5432
0.0962	0.1336	0.0603	0.7099	0.8081	0.6465	0.5224
0.0532	0.7312	0.1053	0.1104	0.7111	0.6546	0.4655
0.0515	0.0900	0.2988	0.5596	0.8051	0.7010	0.5644
0.4458	0.2237	0.2596	0.0709	0.7206	0.7392	0.5327

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.1124	0.4611	0.1661	0.2603	0.7319	0.6163	0.4511
0.1523	0.0919	0.6046	0.1512	0.7563	0.7932	0.5999
0.0539	0.2613	0.4757	0.2091	0.7518	0.7040	0.5293
0.4017	0.0457	0.2010	0.3515	0.7539	0.6777	0.5109
0.0962	0.1336	0.0603	0.7099	0.8036	0.6115	0.4914
0.0532	0.7312	0.1053	0.1104	0.7068	0.6242	0.4412
0.0515	0.0900	0.2988	0.5596	0.8005	0.6632	0.5309
0.4458	0.2237	0.2596	0.0709	0.7163	0.6987	0.5005

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Heptane (1)- Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	1.0000	0.0000	0.0000	0.8671	0.7761	0.6730
0.0000	0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.0879	0.5126	0.1524	0.2471	0.8336	0.7260	0.6052
0.1329	0.1135	0.4007	0.3528	0.7991	0.7355	0.5877
0.0462	0.3050	0.4480	0.2007	0.8154	0.7831	0.6385
0.5125	0.0543	0.3522	0.0809	0.7319	0.6909	0.5057
0.1724	0.1802	0.0551	0.5924	0.8222	0.6556	0.5390
0.1768	0.5958	0.1049	0.1225	0.8197	0.7051	0.5780
0.1348	0.1134	0.6994	0.0524	0.7747	0.8831	0.6841
0.0481	0.1077	0.3027	0.5415	0.8261	0.7216	0.5961

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Heptane (1)- Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	1.0000	0.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.0879	0.5126	0.1524	0.2471	0.8291	0.6874	0.5699
0.1329	0.1135	0.4007	0.3528	0.7946	0.6959	0.5530
0.0462	0.3050	0.4480	0.2007	0.8108	0.7445	0.6036
0.5125	0.0543	0.3522	0.0809	0.7274	0.6577	0.4784
0.1724	0.1802	0.0551	0.5924	0.8176	0.6221	0.5086
0.1768	0.5958	0.1049	0.1225	0.8153	0.6688	0.5453
0.1348	0.1134	0.6994	0.0524	0.7701	0.8274	0.6372
0.0481	0.1077	0.3027	0.5415	0.8215	0.6815	0.5599

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane (1)- Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)

Temperature = 293.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.0000	0.8671	0.7761	0.6730
0.0000	0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.1030	0.5168	0.1707	0.2094	0.8436	0.7309	0.6166
0.1528	0.1096	0.3942	0.3434	0.7964	0.7667	0.6106
0.0554	0.1031	0.6586	0.1829	0.7939	0.8867	0.7040
0.4840	0.0562	0.2141	0.2457	0.7677	0.7033	0.5399
0.1998	0.1475	0.0596	0.5930	0.8158	0.6857	0.5594
0.0993	0.5996	0.1062	0.1949	0.8353	0.7432	0.6208
0.0525	0.0986	0.6948	0.1541	0.7918	0.9033	0.7152
0.0593	0.3498	0.2973	0.2936	0.8263	0.7567	0.6253

Table 4.10 (Cont'd): Densities and Viscosities of the Quaternary System: Octane (1)- Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)

Temperature = 298.15 K

X_1	X_2	X_3	X_4	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.1030	0.5168	0.1707	0.2094	0.8392	0.6917	0.5805
0.1528	0.1096	0.3942	0.3434	0.7919	0.7234	0.5729
0.0554	0.1031	0.6586	0.1829	0.7893	0.8323	0.6569
0.4840	0.0562	0.2141	0.2457	0.7634	0.6659	0.5083
0.1998	0.1475	0.0596	0.5930	0.8113	0.6492	0.5267
0.0993	0.5996	0.1062	0.1949	0.8309	0.7029	0.5840
0.0525	0.0986	0.6948	0.1541	0.7872	0.8477	0.6673
0.0593	0.3498	0.2973	0.2936	0.8218	0.7137	0.5865

Table 4.11: Densities and Viscosities of the Ternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.8671	0.7767	0.6735
0.5078	0.1080	0.3841	0.7532	0.7117	0.5361
0.2283	0.1728	0.5989	0.7976	0.7055	0.5627
0.3525	0.3404	0.3071	0.7390	0.6758	0.4994
0.6567	0.2228	0.1204	0.7142	0.7165	0.5117
0.7961	0.1232	0.0807	0.7106	0.7358	0.5229
0.1030	0.6259	0.2712	0.7298	0.6302	0.4599
0.4259	0.3229	0.2512	0.7311	0.6835	0.4997
0.5053	0.3460	0.1487	0.7162	0.6915	0.4953
0.2131	0.4347	0.3522	0.7449	0.6567	0.4892
0.4167	0.5215	0.0617	0.7017	0.6740	0.4729

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.8627	0.7323	0.6318
0.5078	0.1080	0.3841	0.7490	0.6748	0.5054
0.2283	0.1728	0.5989	0.7933	0.6684	0.5302
0.3525	0.3404	0.3071	0.7347	0.6429	0.4723
0.6567	0.2228	0.1204	0.7100	0.6781	0.4815
0.7961	0.1232	0.0807	0.7064	0.6970	0.4924
0.1030	0.6259	0.2712	0.7255	0.5996	0.4350
0.4259	0.3229	0.2512	0.7268	0.6497	0.4722
0.5053	0.3460	0.1487	0.7120	0.6559	0.4670
0.2131	0.4347	0.3522	0.7406	0.6225	0.4610
0.4167	0.5215	0.0617	0.6976	0.6400	0.4465

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Heptane(2)-Toluene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.5409	0.3113	0.1478	0.7132	0.6849	0.4885
0.2286	0.5408	0.2306	0.7196	0.6296	0.4531
0.3271	0.2149	0.4580	0.7561	0.6470	0.4892
0.6400	0.0999	0.2600	0.7297	0.7006	0.5112
0.7775	0.0624	0.1601	0.7185	0.7254	0.5212
0.0911	0.1415	0.7674	0.8130	0.6368	0.5177
0.2092	0.3478	0.4431	0.7525	0.6269	0.4717
0.3323	0.4415	0.2262	0.7206	0.6472	0.4664
0.2311	0.6804	0.0885	0.7001	0.6345	0.4442
0.3418	0.0386	0.6196	0.7846	0.6618	0.5192

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Heptane(2)-Toluene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.5409	0.3113	0.1478	0.7090	0.6500	0.4609
0.2286	0.5408	0.2306	0.7153	0.5966	0.4267
0.3271	0.2149	0.4580	0.7518	0.6148	0.4622
0.6400	0.0999	0.2600	0.7255	0.6661	0.4833
0.7775	0.0624	0.1601	0.7144	0.6886	0.4919
0.0911	0.1415	0.7674	0.8084	0.6033	0.4877
0.2092	0.3478	0.4431	0.7481	0.5954	0.4454
0.3323	0.4415	0.2262	0.7164	0.6148	0.4404
0.2311	0.6804	0.0885	0.6958	0.6043	0.4205
0.3418	0.0386	0.6196	0.7802	0.6273	0.4894

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Cyclohexane(2)-Toluene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.5017	0.1098	0.3885	0.7566	0.7072	0.5351
0.1987	0.1600	0.6413	0.8057	0.6904	0.5563
0.3443	0.3503	0.3054	0.7635	0.7606	0.5807
0.6440	0.2343	0.1217	0.7283	0.7762	0.5653
0.0666	0.8754	0.0580	0.7738	0.1069	0.0827
0.1040	0.6699	0.2261	0.7818	0.8859	0.6926
0.2056	0.2168	0.5777	0.7994	0.7021	0.5613
0.4979	0.2989	0.2032	0.7432	0.7718	0.5736
0.1953	0.0568	0.7478	0.8155	0.6705	0.5468
0.3079	0.5445	0.1476	0.7547	0.8495	0.6411

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Cyclohexane(2)-Toluene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.5017	0.1098	0.3885	0.7523	0.6695	0.5037
0.1987	0.1600	0.6413	0.8012	0.6532	0.5233
0.3443	0.3503	0.3054	0.7591	0.7201	0.5466
0.6440	0.2343	0.1217	0.7241	0.7334	0.5311
0.0666	0.8754	0.0580	0.7692	0.9941	0.7647
0.1040	0.6699	0.2261	0.7772	0.8318	0.6465
0.2056	0.2168	0.5777	0.7949	0.6659	0.5293
0.4979	0.2989	0.2032	0.7390	0.7282	0.5381
0.1953	0.0568	0.7478	0.8110	0.6348	0.5148
0.3079	0.5445	0.1476	0.7504	0.7984	0.5991

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Ethylbenzene(2)-Toluene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.5013	0.1047	0.3940	0.7661	0.6927	0.5307
0.1992	0.1814	0.6194	0.8214	0.6811	0.5595
0.3460	0.3467	0.3073	0.7952	0.6996	0.5563
0.6385	0.2291	0.1323	0.7482	0.7207	0.5392
0.0796	0.8133	0.1071	0.8497	0.7517	0.6387
0.1099	0.6535	0.2365	0.8429	0.7309	0.6161
0.2050	0.2121	0.5829	0.8205	0.6847	0.5618
0.2757	0.2803	0.4440	0.8072	0.6968	0.5625
0.1962	0.0525	0.7513	0.8212	0.6671	0.5478
0.3145	0.4845	0.2011	0.8023	0.7181	0.5761

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Ethylbenzene(2)-Toluene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.5013	0.1047	0.3940	0.7619	0.6595	0.5025
0.1992	0.1814	0.6194	0.8170	0.6442	0.5263
0.3460	0.3467	0.3073	0.7909	0.6632	0.5245
0.6385	0.2291	0.1323	0.7440	0.6843	0.5091
0.0796	0.8133	0.1071	0.8453	0.7085	0.5989
0.1099	0.6535	0.2365	0.8385	0.6914	0.5797
0.2050	0.2121	0.5829	0.8160	0.6480	0.5288
0.2757	0.2803	0.4440	0.8029	0.6600	0.5299
0.1962	0.0525	0.7513	0.8167	0.6328	0.5168
0.3145	0.4845	0.2011	0.7980	0.6772	0.5404

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Heptane(2)-Cyclohexane(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.6840	0.6023	0.4120
0.0000	0.0000	1.0000	0.7787	1.2560	0.9780
0.5030	0.1011	0.3960	0.7222	0.8237	0.5949
0.1965	0.1606	0.6428	0.7392	0.8864	0.6552
0.3557	0.3279	0.3164	0.7134	0.7571	0.5401
0.6235	0.2089	0.1676	0.7072	0.7570	0.5354
0.1119	0.8070	0.0810	0.6917	0.6383	0.4415
0.1079	0.6529	0.2392	0.7030	0.6792	0.4775
0.2042	0.2090	0.5868	0.7339	0.8534	0.6263
0.5054	0.2819	0.2128	0.7083	0.7505	0.5316
0.1534	0.0946	0.7520	0.7506	0.9774	0.7336
0.3067	0.5225	0.1708	0.7017	0.6979	0.4897

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Heptane(2)-Cyclohexane(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.6797	0.5731	0.3895
0.0000	0.0000	1.0000	0.7739	1.1540	0.8931
0.5030	0.1011	0.3960	0.7179	0.7744	0.5559
0.1965	0.1606	0.6428	0.7348	0.8311	0.6107
0.3557	0.3279	0.3164	0.7092	0.7164	0.5081
0.6235	0.2089	0.1676	0.7030	0.7150	0.5026
0.1119	0.8070	0.0810	0.6875	0.6051	0.4160
0.1079	0.6529	0.2392	0.6987	0.6441	0.4500
0.2042	0.2090	0.5868	0.7295	0.8034	0.5861
0.5054	0.2819	0.2128	0.7041	0.7084	0.4988
0.1534	0.0946	0.7520	0.7462	0.9129	0.6812
0.3067	0.5225	0.1708	0.6975	0.6609	0.4610

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	1.0000	0.7787	1.2560	0.9780
0.4935	0.1008	0.4057	0.7321	0.7214	0.5281
0.1995	0.1518	0.6487	0.7665	0.8565	0.6565
0.3365	0.3542	0.3093	0.7700	0.7106	0.5472
0.6629	0.2501	0.0869	0.7307	0.6317	0.4616
0.1545	0.6886	0.1569	0.8200	0.7285	0.5974
0.1038	0.6610	0.2352	0.8237	0.7539	0.6210
0.1895	0.2488	0.5617	0.7764	0.8130	0.6312
0.4709	0.2904	0.2388	0.7512	0.6772	0.5087
0.1871	0.0550	0.7578	0.7594	0.9289	0.7054
0.2951	0.5494	0.1555	0.7920	0.6937	0.5494

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	1.0000	0.7739	1.1540	0.8931
0.4935	0.1008	0.4057	0.7277	0.6834	0.4973
0.1995	0.1518	0.6487	0.7619	0.8035	0.6122
0.3365	0.3542	0.3093	0.7655	0.6740	0.5159
0.6629	0.2501	0.0869	0.7264	0.6005	0.4362
0.1545	0.6886	0.1569	0.8156	0.6921	0.5645
0.1038	0.6610	0.2352	0.8192	0.7115	0.5829
0.1895	0.2488	0.5617	0.7719	0.7654	0.5908
0.4709	0.2904	0.2388	0.7468	0.6427	0.4800
0.1871	0.0550	0.7578	0.7549	0.8703	0.6570
0.2951	0.5494	0.1555	0.7920	0.6937	0.5494

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Ethylbenzene(2)-Cyclohexane(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.7025	0.7734	0.5433
0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	1.0000	0.7787	1.2560	0.9780
0.4946	0.1011	0.4043	0.7395	0.8277	0.6121
0.1974	0.1536	0.6491	0.7692	0.9055	0.6965
0.3338	0.3512	0.3149	0.7735	0.7866	0.6084
0.6095	0.2235	0.1670	0.7416	0.7672	0.5690
0.1528	0.6993	0.1478	0.8218	0.7589	0.6237
0.1021	0.6666	0.2313	0.8246	0.7748	0.6389
0.1968	0.2249	0.5783	0.7754	0.8742	0.6779
0.4966	0.2962	0.2072	0.7556	0.7689	0.5810
0.1906	0.0570	0.7524	0.7614	0.9752	0.7425
0.2981	0.5517	0.1502	0.7940	0.7489	0.5946

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Octane(1)-Ethylbenzene(2)-Cyclohexane(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6985	0.7313	0.5108
0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	1.0000	0.7739	1.1540	0.8931
0.4946	0.1011	0.4043	0.7352	0.7802	0.5736
0.1974	0.1536	0.6491	0.7647	0.8521	0.6516
0.3338	0.3512	0.3149	0.7692	0.7445	0.5727
0.6095	0.2235	0.1670	0.7374	0.7246	0.5343
0.1528	0.6993	0.1478	0.8174	0.7165	0.5857
0.1021	0.6666	0.2313	0.8202	0.7324	0.6007
0.1968	0.2249	0.5783	0.7710	0.8239	0.6352
0.4966	0.2962	0.2072	0.7514	0.7260	0.5455
0.1906	0.0570	0.7524	0.7569	0.9114	0.6898
0.2981	0.5517	0.1502	0.7896	0.7098	0.5605

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Heptane(1)-Ethylbenzene(2)-Toluene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	1.0000	0.0000	0.8671	0.7767	0.6735
0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.4975	0.1023	0.4002	0.7622	0.5998	0.4572
0.2032	0.1572	0.6396	0.8201	0.6407	0.5254
0.3418	0.3435	0.3146	0.7939	0.6363	0.5052
0.6473	0.2337	0.1190	0.7393	0.6094	0.4505
0.1296	0.7199	0.1505	0.8390	0.7105	0.5961
0.0949	0.6681	0.2370	0.8461	0.7147	0.6047
0.2052	0.2205	0.5742	0.8203	0.6459	0.5298
0.5008	0.2995	0.1997	0.7646	0.6217	0.4754
0.1935	0.0565	0.7500	0.8216	0.6322	0.5194
0.3012	0.5445	0.1543	0.8037	0.6625	0.5325

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Heptane(1)-Ethylbenzene(2)-Toluene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	1.0000	0.0000	0.8627	0.7323	0.6318
0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.4975	0.1023	0.4002	0.7578	0.5736	0.4347
0.2032	0.1572	0.6396	0.8156	0.6070	0.4951
0.3418	0.3435	0.3146	0.7894	0.6050	0.4776
0.6473	0.2337	0.1190	0.7350	0.5790	0.4256
0.1296	0.7199	0.1505	0.8346	0.6743	0.5628
0.0949	0.6681	0.2370	0.8416	0.6762	0.5691
0.2052	0.2205	0.5742	0.8158	0.6135	0.5005
0.5008	0.2995	0.1997	0.7602	0.5900	0.4485
0.1935	0.0565	0.7500	0.8170	0.6010	0.4910
0.3012	0.5445	0.1543	0.7993	0.6289	0.5027

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Heptane(1)-Cyclohexane(2)-Toluene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6840	0.6023	0.4120
0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.4920	0.1114	0.3966	0.7526	0.6140	0.4621
0.1927	0.1501	0.6572	0.8073	0.6482	0.5233
0.3297	0.3651	0.3052	0.7616	0.6927	0.5276
0.6326	0.2396	0.1279	0.7198	0.6520	0.4693
0.1464	0.7008	0.1529	0.7704	0.8755	0.6745
0.1019	0.6700	0.2282	0.7817	0.8578	0.6705
0.1946	0.2259	0.5794	0.8001	0.6643	0.5315
0.4936	0.3023	0.2041	0.7376	0.6702	0.4943
0.1894	0.0538	0.7568	0.8169	0.6338	0.5178
0.3008	0.5466	0.1525	0.7528	0.7729	0.5818

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Heptane(1)-Cyclohexane(2)-Toluene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.6797	0.5731	0.3895
0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.4920	0.1114	0.3966	0.7481	0.5843	0.4371
0.1927	0.1501	0.6572	0.8027	0.6146	0.4933
0.3297	0.3651	0.3052	0.7571	0.6570	0.4974
0.6326	0.2396	0.1279	0.7154	0.6182	0.4423
0.1464	0.7008	0.1529	0.7658	0.8210	0.6287
0.1019	0.6700	0.2282	0.7771	0.8044	0.6251
0.1946	0.2259	0.5794	0.7955	0.6302	0.5013
0.4936	0.3023	0.2041	0.7331	0.6349	0.4654
0.1894	0.0538	0.7568	0.8124	0.6004	0.4878
0.3008	0.5466	0.1525	0.7483	0.7292	0.5457

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)
Temperature = 293.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity $x10^6$ (m^2/s)	Absolute Viscosity $x10^3$ (Pa.s)
1.0000	0.0000	0.0000	0.8671	0.7323	0.6350
0.0000	1.0000	0.0000	0.7787	1.2560	0.9780
0.0000	0.0000	1.0000	0.8667	0.6797	0.5891
0.5033	0.1013	0.3954	0.8574	0.7396	0.6341
0.1977	0.1497	0.6526	0.8523	0.7151	0.6095
0.3354	0.3572	0.3074	0.8336	0.7715	0.6431
0.6430	0.2330	0.1240	0.8461	0.7739	0.6548
0.1520	0.6940	0.1540	0.8033	0.9104	0.7313
0.1055	0.6570	0.2374	0.8059	0.8789	0.7083
0.2000	0.2211	0.5789	0.8455	0.7258	0.6137
0.4964	0.3000	0.2036	0.8395	0.7769	0.6522
0.1950	0.0521	0.7529	0.8619	0.7038	0.6066
0.2965	0.5498	0.1537	0.8166	0.8412	0.6869

Table 4.11 (Cont'd): Densities and Viscosities of the Ternary System: Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)
Temperature = 298.15 K

X_1	X_2	X_3	Density (kg/L)	Kinematic Viscosity x10^6 (m^2/s)	Absolute Viscosity x10^3 (Pa.s)
1.0000	0.0000	0.0000	0.8627	0.7327	0.6318
0.0000	1.0000	0.0000	0.7739	1.1540	0.8931
0.0000	0.0000	1.0000	0.8620	0.6413	0.5528
0.5033	0.1013	0.3954	0.8529	0.7000	0.5970
0.1977	0.1497	0.6526	0.8477	0.6752	0.5724
0.3354	0.3572	0.3074	0.8290	0.7286	0.6040
0.6430	0.2330	0.1240	0.8416	0.7319	0.6160
0.1520	0.6940	0.1540	0.7987	0.8524	0.6808
0.1055	0.6570	0.2374	0.8013	0.8220	0.6587
0.2000	0.2211	0.5789	0.8409	0.6853	0.5763
0.4964	0.3000	0.2036	0.8350	0.7307	0.6101
0.1950	0.0521	0.7529	0.8573	0.6657	0.5707
0.2965	0.5498	0.1537	0.8120	0.7892	0.6408

4.7 The Predictive Capabilities of the Different Viscosity Models

In this section, the viscosity-composition experimental data were utilized in subjecting the literature viscosity models to critical testing. The following models were tested:

1. The McAllister Three-Body Model
2. The Pseudo-Binary McAllister Model
3. The GCSP (with different values for r)
4. The GC-UNIMOD
5. The Modified GCSP
6. The Allan and Teja Correlation

Each of the above listed models is briefly presented and discussed in a separate section in this chapter. Comparison of the performance of each model and its agreement with the experimental data are discussed in terms of %AAD (percent absolute average deviation), and %MAX (per cent maximum deviation of the model from experimental data). Following are the definitions of the % AAD and %MAX:

$$\%AAD = \frac{1}{m}\left[\sum_{i=1}^{m}\frac{\left|X_i^{exp} - X_i^{cal}\right|}{X_i^{exp}}\right] x100 \tag{4.1}$$

where m is the number of experimental points, and X is the kinematic viscosity. The %MAX which is the maximum deviation of the predicted data is defined as

$$\%MAX = \max\left[\frac{X_i^{exp} - X_i^{cal}}{X_i^{exp}}\right]x100 \tag{4.2}$$

4.7.1 The Generalized McAllister three-body Model

Asfour and Nhaesi (2000a) extended the McAllister three-body collision model to n-component systems. The model they developed is given by

$$\ell n\nu_m = \sum_{i=1}^{n} X_i^3 \ell n(\nu_i M_i) + 3\sum_{i=1}^{n}\sum_{i=1}^{n} X_i^2 X_j \ell n(\nu_{ij} M_{ij})$$

$$+ 6\sum_{i=1}^{n}\sum_{i=1}^{n}\sum_{k=1}^{n} X_i X_j X_k \ell n(\nu_{ijk} M_{ijk}) - \ell n(M_{avg}) \tag{4.3}$$

The model, given by equation (4.3), contains binary and ternary interaction parameters. In order to calculate the values of the binary parameters, Asfour and Nhaesi (2000a) suggested the following equation for the binary parameters in case of regular solution systems:

$$\frac{\nu_{12}}{(\nu_1^2 \nu_2)^{1/3}} = 0.8735 + 0.0715\frac{(ECN_2 - ECN_1)^2}{(ECN_1^2\, ECN_2)^{1/3}} \tag{4.4}$$

and

$$\nu_{21} = \nu_{12}\left(\frac{\nu_2}{\nu_1}\right)^{1/3} \tag{4.5}$$

For the ternary parameters, the following equation was proposed by Nhaesi and Asfour (2000a):

$$\frac{\nu_{123}}{(\nu_1 \nu_2 \nu_3)^{1/3}} = 0.9637 + 0.0313\frac{(ECN_3 - ECN_1)^2}{ECN_2} \tag{4.6}$$

With the help of equations (4.4), (4.5), and (4.6), the binary and ternary parameter values can be predicted from the molecular and pure component properties. Inserting the values of the predicted parameters into equation (4.3) along with the pure component viscosities, and the appropriate mole fractions, one would be able to calculate viscosity of the mixture over the entire composition range. It should be noted here that the ECN's in equations (4.4) and (4.6) represent the *effective carbon numbers* of the components constituting a mixture. The *effective carbon numbers* are calculated from the following equation, which was proposed by Nhaesi and Asfour (1998):

$$\ell n(\nu) = A + B(N) \qquad (4.7)$$

where ν is the kinematic viscosity of the component of a multi-component system at 308.15 K, and N is its effective carbon number. The values of the constants A and B were reported by those authors as follows: A = - 1.943 and B = 0.193. Table 4.11 lists the effective carbon number values of each component used in this study.

It should be noted here that Nhaesi and Asfour (1998) reported an ECN value for cyclohexane of 8.9. That value was a misprint since the correct value calculated from equation (4.7) is 10.01.

Using the data from all the systems tested during this study, the predictive capability of the McAllister three-body model, equation (4.3), was tested. The results are given in Tables 4.13 through 4.17. For the quinary systems, there is an overall % AAD of 1.48, and a maximum deviation of 12.12%. For the quaternary systems investigated in this study, the overall AAD is 3.96% and the maximum deviation is 13.68%. Finally, the ternary systems showed an overall %AAD of 3.85, and a maximum deviation of 13.32%.

Table 4.12: ECN of pure components

Pure Component	**ECN**
Hexane	6.0
Heptane	7.0
Octane	8.0
Ethylbenzene	7.9
Cyclohexane	10.01
Toluene	7.2

From all the data presented so far it is clear that the generalized McAllister three-body collision model shows an excellent predictive capability. This confirms the results reported earlier by Nhaesi and Asfour (2000a). However, one should note here that deviations of the model from experimental data seem to increase as the mole fraction of cyclohexane increases in solution. Would that be a direct result of a higher value of the ECN calculated from equation (4.7)? During the course of this study lower values for the ECN of cyclohexane were arbitrarily employed. The lowest % maximum deviation and %AAD were obtained at a value of ECN of cyclohexane of 7.5. This shows that equation (4.7) actually over-predicts the ECN for cyclohexane. Is this true for other “planar” type molecules similar to cyclohexane; like cyclopentane and cycloheptane? This will have to be investigated in a separate and independent study.

4.7.2 "Pseudo-Binary" McAllister three-body model

Wu and Asfour (1992) proposed the pseudo-binary concept in order to deal with the problem of the arbitrary selection of a reference fluid required for the generalized corresponding states principle (GCSP). The idea is to treat a multi-component mixture ($n > 2$) as a binary mixture, one of the components is considered as component 1, and all the other components together represent pseudo-component 2’. Nhaesi and Asfour (2000b) utilized the pseudo-binary concept with the McAllister model for n-component systems.

The ECN of the pseudo-component was calculated by using the following equation:

$$(\mathrm{ECN})_{2'} = \sum_{i=2}^{n} X_i (\mathrm{ECN})_i \tag{4.8}$$

The Arrhenius equation of liquid viscosity was utilized by Nhaesi and Asfour (2000b) to evaluate the viscosity of the pseudo-component. The equation is as follows

$$\ell n \nu_{2'} = \sum_{i=2}^{n} X_i \ell n \nu_i \tag{4.9}$$

The molecular weight of the pseudo-component, $M_{2'}$, was calculated by Nhaesi and Asfour (2000b) by using the following mixing rule:

$$\ell n M_{2'} = \sum_{i=2}^{n} X_i \ell n M_i \tag{4.10}$$

It should be noted here also that Nhaesi and Asfour (2000b) pointed out that the mole fraction X_i in equations (4.7), (4.8), and (4.9) was normalized according to the following equation:

$$X_i = \frac{X_i}{\sum_{i=2}^{n} X_i} \tag{4.11}$$

The values of the $(\mathrm{ECN})_{2'}$ and $\nu_{2'}$ are substituted into equations (4.4) and (4.5) to calculate the binary interaction parameters of the McAllister model. Substitution of the result obtained from equations (4.8) and (4.9) into the McAllister model, equation (2.63), would result in the value of the kinematic viscosity of the liquid mixture. It should be noted here that according to this approach, there are only two interaction parameters no matter how many components the mixture actually contains.

Tables 4.18 to 4.22 give the results of using the experimental data to test the pseudo binary McAllister model.

Table 4.13: Results of testing the McAllister three-body model by using the experimental viscosity data of the quinary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	2.01	12.12
	298.15	1.63	10.82

Table 4.14: Results of testing the McAllister three-body model by using the experimental viscosity data of the quaternary systems containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	4.47	13.68
	298.15	3.98	12.36
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	1.78	4.36
	298.15	2.35	5.71

Table 4.15: Results of testing the McAllister three-body model by using the experimental viscosity data of the quinary system containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	1.63	10.66
	298.15	1.33	9.50

Table 4.16: Results of testing the McAllister three-body model using the experimental viscosity data of the quaternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	3.62	12.31
	298.15	3.12	10.96
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15	1.82	4.68
	298.15	1.99	4.97
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15	4.09	12.66
	298.15	3.70	11.73
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	4.69	13.46
	298.15	4.02	12.28
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	4.40	11.91
	298.15	3.95	10.46

Table 4.17: Results of testing the McAllister three-body model by using the experimental viscosity data of the ternary systems containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	3.66	8.11
	298.15	3.82	8.21
Octane(1)-Heptane(2)- Toluene(3)	293.15	3.02	6.30
	298.15	3.24	6.49
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	4.04	11.48
	298.15	3.37	10.07
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	2.92	5.37
	298.15	3.16	5.63
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	3.00	8.72
	298.15	2.68	7.74
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	6.13	13.18
	298.15	5.39	11.91
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	4.04	9.82
	298.15	3.31	8.27
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	2.16	5.25
	298.15	2.49	5.48
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	6.35	13.32
	298.15	5.61	12.03
Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)	293.15	4.47	11.59
	298.15	4.07	10.62

Table 4.18: Results of testing the pseudo-binary McAllister model by using the experimental viscosity data of the quinary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	2.34	10.64
	298.15	2.05	9.35

Table 4.19: Results of testing the pseudo-binary McAllister model by using the experimental viscosity data of the quaternary systems containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	4.80	16.83
	298.15	4.48	15.48
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	1.72	4.27
	298.15	1.28	4.42

Table 4.20: Results of testing the pseudo-binary McAllister model by using the experimental viscosity data of the quinary system containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	2.39	10.21
	298.15	2.17	9.06

Table 4.21: Results of testing the pseudo-binary McAllister model by using the experimental viscosity data of the quaternary systems containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	5.10	16.98
	298.15	4.73	15.58
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15	2.08	6.00
	298.15	2.00	5.50
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15	4.68	16.36
	298.15	4.29	15.39
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	6.49	16.76
	298.15	5.70	14.53
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	5.95	15.47
	298.15	5.32	13.98

Table 4.22: Results of testing the pseudo binary McAllister model using the experimental viscosity data on the ternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	3.35	8.19
	298.15	3.47	8.30
Octane(1)-Heptane(2)- Toluene(3)	293.15	2.68	6.80
	298.15	2.83	6.99
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	3.82	13.33
	298.15	3.23	11.90
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	1.86	3.92
	298.15	2.10	4.68
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	3.11	8.11
	298.15	2.90	7.14
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	6.51	15.02
	298.15	5.77	13.73
Octane(1)-Ethylbenzene(2)-cyclohexane(3)	293.15	4.27	10.85
	298.15	3.61	9.28
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	1.18	2.91
	298.15	1.51	3.08
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	6.30	15.48
	298.15	5.56	14.24
Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)	293.15	4.33	13.51
	298.15	3.91	12.52

Comparing the %ADDs and % maximum deviations reported so far, it is clear that the three-body collision McAllister model consistently predicted viscosities over the entire composition range for all systems investigated better than the pseudo-McAllister model, though the differences between the predictive capabilities of the two models are not very significant. However, it should be remembered that the calculation procedures followed in case of the McAllister pseudo-binary model are more convenient than those followed in the case of the three-body collision McAllister model.

4.7.3 The Generalized Corresponding States Principle (GCSP)

As pointed-out earlier in Chapter 2, the GCSP is superior to the TRAPP method, and this is the reason for selecting it in the comparative study reported herein. When the GCSP is used in the calculations, it requires the knowledge of the critical pressures and temperatures of the components constituting the mixture. Such values are available from literature sources such as Reid *et al.* (1987). The pseudo critical properties of the mixtures involved are normally calculated by following the procedures and equations suggested by Wong *et al.* (1984) and given earlier by equations (2.27) through (2.33) in Chapter 2 of this document.

As indicated earlier in Chapter 2, the selection of the reference component in calculations involving the GCSP significantly influences the %AADs obtained. This was made clear by Wu and Asfour (1992) and Wu et al. (1998). There is no rule that guarantees *apriori* selection that will result in the lowest possible %AAD. Therefore, calculations were made for all possible selections and the lowest % AADs are reported in Tables 4.23 through 4.27.

4.7.4 The GC-UNIMOD Method

Cao *et al.* (1993) reported a predictive model which they called the GC-UNIMOD. In their research, they employed an extensive database for testing their model. That database contained a variety of binary, ternary and quaternary liquid mixtures. The results of their tests indicated that the model showed a good predictive capability. Monnery *et al.* (1995), in their review supported the results reported by Cao *et al.* (1993).

As was indicated earlier in Chapter 2, the Cao *et al.* (1993) model consists of two parts, a *combinatorial* part and a *residual* part, as shown by equation (2.87). Nhaesi (1998) indicated that the residual part had an insignificant effect on the predictive capability of the model. Therefore, in this study, the model was applied (i) with the two parts, and (ii) with the residual part set to zero, in order to test its effect on the predictive capability with the systems investigated in this study.

Results of testing reported in Tables 4.28 through 4.37 show that the residual part had an insignificant effect on the predictive capability of the model, which confirms the results that Nhaesi (1998) reported in his study on a different set of liquid mixtures. In addition, the predictive capability of the GC-UNIMODE method is certainly significantly lower than those of the McAllister three-body model, the pseudo-binary McAllister model.

Table 4.23: Results of testing the GCSP model by using the experimental viscosity data of the quinary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	13.41	27.99
	298.15	12.87	26.89

Table 4.24: Results of testing the GCSP model by using the experimental viscosity data of the quaternary systems containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	19.19	53.24
	298.15	18.71	52.02
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	15.19	38.52
	298.15	13.83	32.27

Table 4.25: Results of testing the GCSP model by using the experimental viscosity data of the quinary system containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	8.06	15.01
	298.15	7.78	14.48

Table 4.26: Results of testing the GCSP model by using the experimental viscosity data of the quaternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	9.76	22.97
	298.15	9.17	22.44
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15	7.36	15.01
	298.15	7.26	14.74
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15	7.33	15.29
	298.15	7.06	14.94
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	6.91	16.47
	298.15	7.05	15.97
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	8.62	16.71
	298.15	8.32	16.26

Table 4.27: Results of testing the GCSP model by using the experimental viscosity data of the ternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	9.54	25.50
	298.15	9.45	25.11
Octane(1)-Heptane(2)- Toluene(3)	293.15	8.86	22.59
	298.15	8.75	22.27
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	9.51	34.59
	298.15	9.23	33.63
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	3.97	7.18
	298.15	3.69	7.05
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	9.18	25.64
	298.15	8.83	24.90
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	10.79	16.22
	298.15	9.71	14.86
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	5.01	13.32
	298.15	4.79	12.57
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	4.45	11.67
	298.15	4.29	11.89
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	5.72	14.83
	298.15	5.49	14.42
Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)	293.15	10.32	24.47
	298.15	10.12	24.08

Table 4.28: Results of testing the GC-UNIMOD model by using the experimental viscosity data of the quinary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	7.86	12.98
	298.15	7.20	11.83

Table 4.29: Results of testing the GC-UNIMOD model by using the experimental viscosity data of the quaternary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	9.86	16.17
	298.15	9.19	14.94
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	4.08	4.80
	298.15	3.19	4.68

Table 4.30: Results of testing the GC-UNIMOD model by using the experimental viscosity data of the quinary system containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	7.30	11.86
	298.15	6.83	10.88

Table 4.31: Results of testing the GC-UNIMOD model by using the experimental viscosity data of the quaternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	9.28	15.57
	298.15	8.62	14.34
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15	4.26	6.72
	298.15	4.01	6.24
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15	8.14	12.84
	298.15	7.65	12.08
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	10.83	15.68
	298.15	9.73	13.55
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	10.98	14.74
	298.15	10.09	13.34

Table 4.32: Results of testing the GC-UNIMOD model by using the experimental viscosity data of the ternary systems containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	2.24	3.32
	298.15	2.03	3.19
Octane(1)-Heptane(2)- Toluene(3)	293.15	2.57	4.24
	298.15	2.29	3.96
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	8.37	13.50
	298.15	7.58	12.17
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	3.35	4.51
	298.15	3.03	4.10
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	4.78	8.36
	298.15	4.37	7.63
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	10.68	15.21
	298.15	9.82	14.08
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	9.30	12.58
	298.15	8.47	11.21
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	4.65	6.52
	298.15	4.21	5.55
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	10.21	12.84
	298.15	9.38	11.80
Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)	293.15	8.80	14.43
	298.15	8.04	13.50

Table 4.33: Results of testing the GC-UNIMOD model (residual = 0) by using the experimental viscosity data of the quinary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	8.70	13.86
	298.15	8.03	12.69

Table 4.34: Results of testing the GC-UNIMOD model (residual = 0) by using the experimental viscosity data of the quaternary systems containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	9.75	15.89
	298.15	9.08	14.67
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	4.79	5.53
	298.15	3.88	5.17

Table 4.35: Results of testing the GC-UNIMOD model (residual = 0) using the experimental viscosity data on the quinary system containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	8.13	12.71
	298.15	7.65	11.72

Table 4.36: Results of testing the GC-UNIMOD model (residual = 0) by using the experimental viscosity data of the quaternary systems containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15 298.15	9.17 8.52	15.30 14.07
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15 298.15	4.93 4.69	6.98 6.50
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15 298.15	9.10 8.59	14.25 13.46
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15 298.15	11.55 10.43	16.54 14.44
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15 298.15	11.93 11.02	15.78 14.35

Table 4.37: Results of testing the GC-UNIMOD model (residual = 0) by using the experimental viscosity data of the ternary systems containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	3.41	4.99
	298.15	3.19	4.81
Octane(1)-Heptane(2)- Toluene(3)	293.15	3.68	5.57
	298.15	3.39	5.22
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	9.51	14.99
	298.15	8.70	13.62
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	3.98	5.37
	298.15	3.66	4.94
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	4.78	8.36
	298.15	4.37	7.63
Heptane(1)-Ethylbenzene(2)-	293.15	10.52	14.94
Cyclohexane(3)	298.15	9.67	13.81
Octane(1)-Ethylbenzene(2)-	293.15	9.16	12.34
Cyclohexane(3)	298.15	8.34	10.98
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	5.08	7.54
	298.15	4.64	6.56
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	11.14	13.97
	298.15	10.29	12.90
Ethylbenzene(1)-Cyclohexane(2)-	293.15	9.14	14.97
Toluene(3)	298.15	8.39	14.03

4.7.5 The Modified Generalized Corresponding States Principle (MGCSP)

The MGCSP method was originally developed by Wu and Asfour (1992) to predict the viscosities of n-alkane systems. The modification depends on using a pseudo-binary model, which was described earlier in Chapter 2. Nhaesi and Asfour (2000a), extended the method to cover regular solutions. They utilized the ECN concept that they developed (Nhaesi and Asfour 1998) along with equations (2.34) through (2.38) to perform the required calculations.

Tables 4.38 through 4.42 report the results of testing the predictive capability of the modified corresponding states principle. Generally, the method gives good predictions when compared with the other methods. However, its predictive capability is much lower than those of the McAllister three-body collision model and the pseudo-binary McAllister model.

4.7.6 The Allan and Teja Correlation

Allan and Teja proposed this Antoine-type correlation, equation (2.98) for estimating the viscosities of liquid systems. Allan and Teja (1991) correlated the constants A, B, and C contained in equation (2.98) with the carbon number of n-alkanes. The following mixing rule was suggested by the authors for calculating the effective carbon numbers of a mixture by using the effective carbon numbers of the constituents of the mixture:

$$\mathrm{ECN} = \sum_{i=1}^{n} X_i (\mathrm{ECN}_i) \tag{4.12}$$

Table 4.38: Results of testing the MGCSP model by using the experimental viscosity data on the quinary system containing hexane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	5.61	9.32
	298.15	5.41	9.22

Table 4.39: Results of testing the MGCSP model by using the experimental viscosity data of the quaternary systems containing hexane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	4.08	8.95
	298.15	4.01	8.73
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	14.09	23.62
	298.15	10.57	19.93

Table 4.40: Results of testing the MGCSP model by using the experimental viscosity data of the quinary system containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	5.49	9.36
	298.15	5.21	9.09

Table 4.41: Results of testing the MGCSP by using the experimental viscosity data of the quaternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	3.66	7.78
	298.15	3.63	7.58
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15	9.86	20.27
	298.15	9.54	19.51
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15	5.84	14.86
	298.15	5.60	14.29
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	11.54	16.88
	298.15	11.37	16.63
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	11.92	16.69
	298.15	11.66	16.44

Table 4.42: Results of testing the MGCSP model by using the experimental viscosity data of the ternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	2.16	10.41
	298.15	2.09	10.20
Octane(1)-Heptane(2)- Toluene(3)	293.15	4.99	16.92
	298.15	4.83	16.40
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	8.68	18.11
	298.15	8.46	17.26
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	14.33	20.50
	298.15	14.07	19.90
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	2.85	11.22
	298.15	2.86	10.57
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	5.76	12.02
	298.15	5.66	12.05
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	6.38	12.27
	298.15	6.30	12.11
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	13.21	20.26
	298.15	13.07	19.88
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	7.43	16.79
	298.15	7.27	16.35
Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)	293.15	11.14	22.18
	298.15	10.87	21.78

Table 4.43: Results of testing the Allan and Teja correlation by using the experimental viscosity data of the quinary system containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	7.83	24.52
	298.15	7.64	23.90

Table 4.44: Results of testing the Allan and Teja correlation by using the experimental viscosity data of the quaternary systems containing hexane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	6.81	20.96
	298.15	6.76	20.64
Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)	293.15	13.86	28.59
	298.15	9.56	23.90

Table 4.45: Results of testing the Allan and Teja correlation by using the experimental viscosity data of the quinary system containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)	293.15	7.69	24.52
	298.15	7.46	23.90

Table 4.46: Results of testing the Allan and Teja correlation by using the experimental viscosity data of the quaternary systems containing heptane.

System	**Temperature, K**	**%AAD**	**%MAX**
Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)	293.15	6.77	20.96
	298.15	6.60	20.64
Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)	293.15	8.97	24.52
	298.15	8.82	23.90
Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)	293.15	7.01	24.52
	298.15	6.87	23.90
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	9.67	24.52
	298.15	9.35	23.90
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)	293.15	9.25	24.52
	298.15	9.02	23.90

Table 4.47: Results of testing the Allan and Teja correlation by using the experimental viscosity data of the ternary systems containing heptane.

System	Temperature, K	%AAD	%MAX
Octane(1)-Heptane(2)-Ethylbenzene(3)	293.15	2.98	20.96
	298.15	3.03	20.64
Octane(1)-Heptane(2)- Toluene(3)	293.15	4.50	24.52
	298.15	4.41	23.90
Octane(1)- Cyclohexane(2)-Toluene(3)	293.15	7.35	24.52
	298.15	7.21	23.90
Octane(1)-Ethylbenzene(2)- Toluene(3)	293.15	12.43	24.52
	298.15	12.27	23.90
Octane(1)-Heptane(2)-Cyclohexane(3)	293.15	7.46	13.17
	298.15	7.49	13.11
Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	7.85	20.96
	298.15	7.70	20.64
Octane(1)-Ethylbenzene(2)-Cyclohexane(3)	293.15	7.34	20.96
	298.15	7.17	20.64
Heptane(1)-Ethylbenzene(2)-Toluene(3)	293.15	11.52	24.52
	298.15	11.42	23.90
Heptane(1)- Cyclohexane(2)-Toluene(3)	293.15	8.55	24.52
	298.15	8.37	23.90
Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)	293.15	11.30	24.52
	298.15	11.10	23.90

The AADs obtained from this method are relatively high; the highest was 12.53%, for the system octane(1) – ethylbenzene(2) – toluene (3) at 293.15 K.

4.8 Summary of all the previous comparisons

Figures 4.1 to 4.4 provide a comparison of the %AADs of all the viscosity models when those models were tested by using the viscosities of the systems containing hexane at 293.15 and 298.15, respectively.

It is clear from those four figures that the generalized McAllister model (G-McA) gave the lowest %AAD at both temperatures. The pseudo-binary McAlliester model (Ps-B-McA) was a close one. All other models showed close values of the %AAD, except the GCSP which gave the largest deviation of all models.

Figures 4.5 to 4.10 provide a comparison of the %AADs of all viscosity models when those models were tested by using the viscosity data of the systems containing heptane at 293.15 and 298.15 K.

Again, the generalized McAllister model gave the best predictions, except for the ternary systems at 293.15 and 298.15 K. The pseudo-binary McAllister model gave slightly better predictions, as indicated in Figures 4.9 and 4.10. The differences were generally insignificant. All other models gave close %AADs with each other. However, the %AADs given by the generalized McAllister and the pseudo-binary McAllister models are significantly lower than the %AADs of the other models.

Finally, Figures 4.11 and 4.12 provide 3-D bar charts that give overall comparison of all models in case of the systems containing hexane and those containing heptane, respectively. Again, it is clear that the Generalized McAllister model gave the best predictions followed by the pseudo-binary McAllister model. All other models gave significantly higher %AADs then those two models.

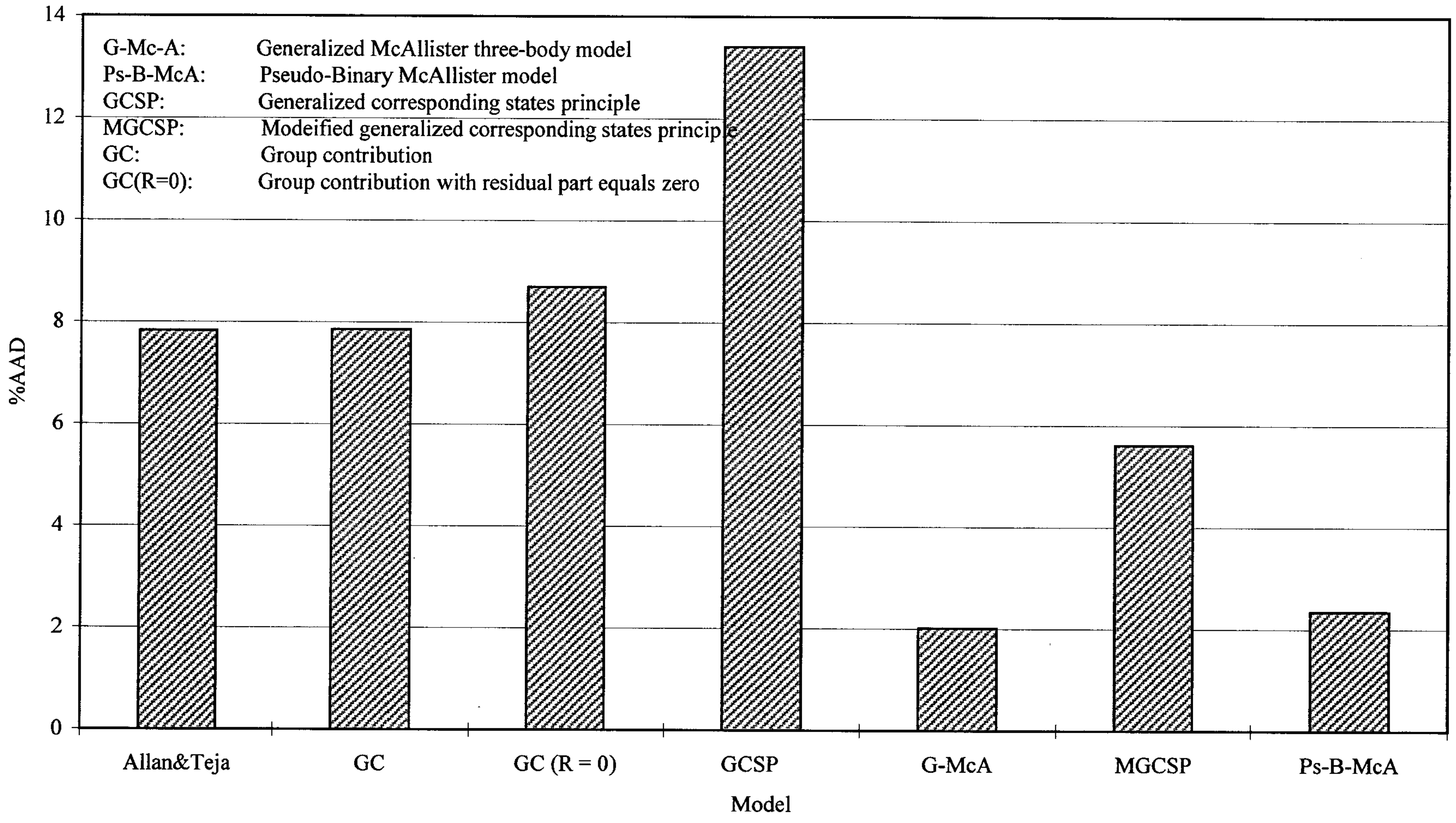

Figure 4.1: Model Comparison for Quinary System containing Hexane at 293.15 K

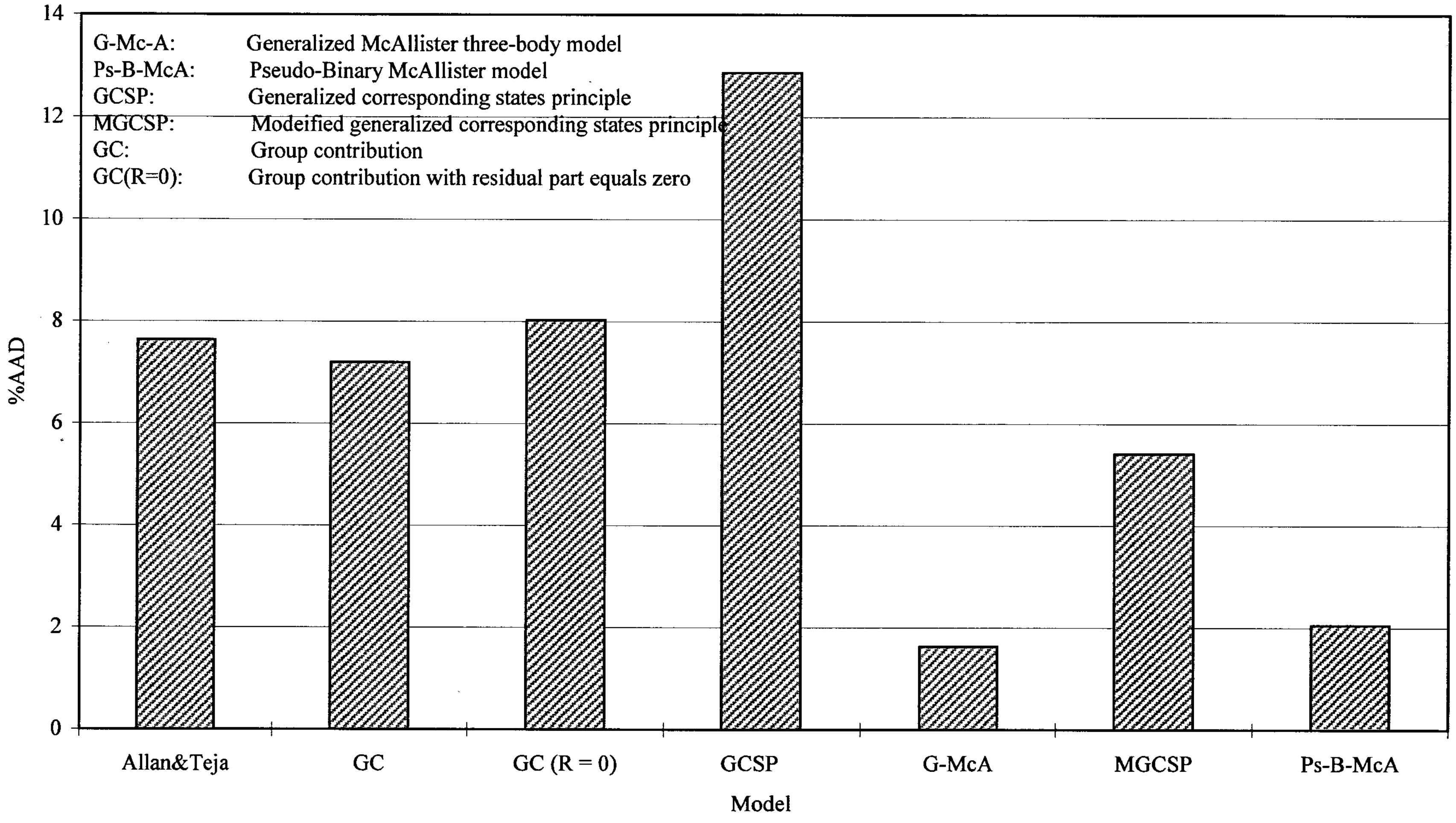

Figure 4.2: Model Comparison for Quinary System containing Hexane at 298.15 K

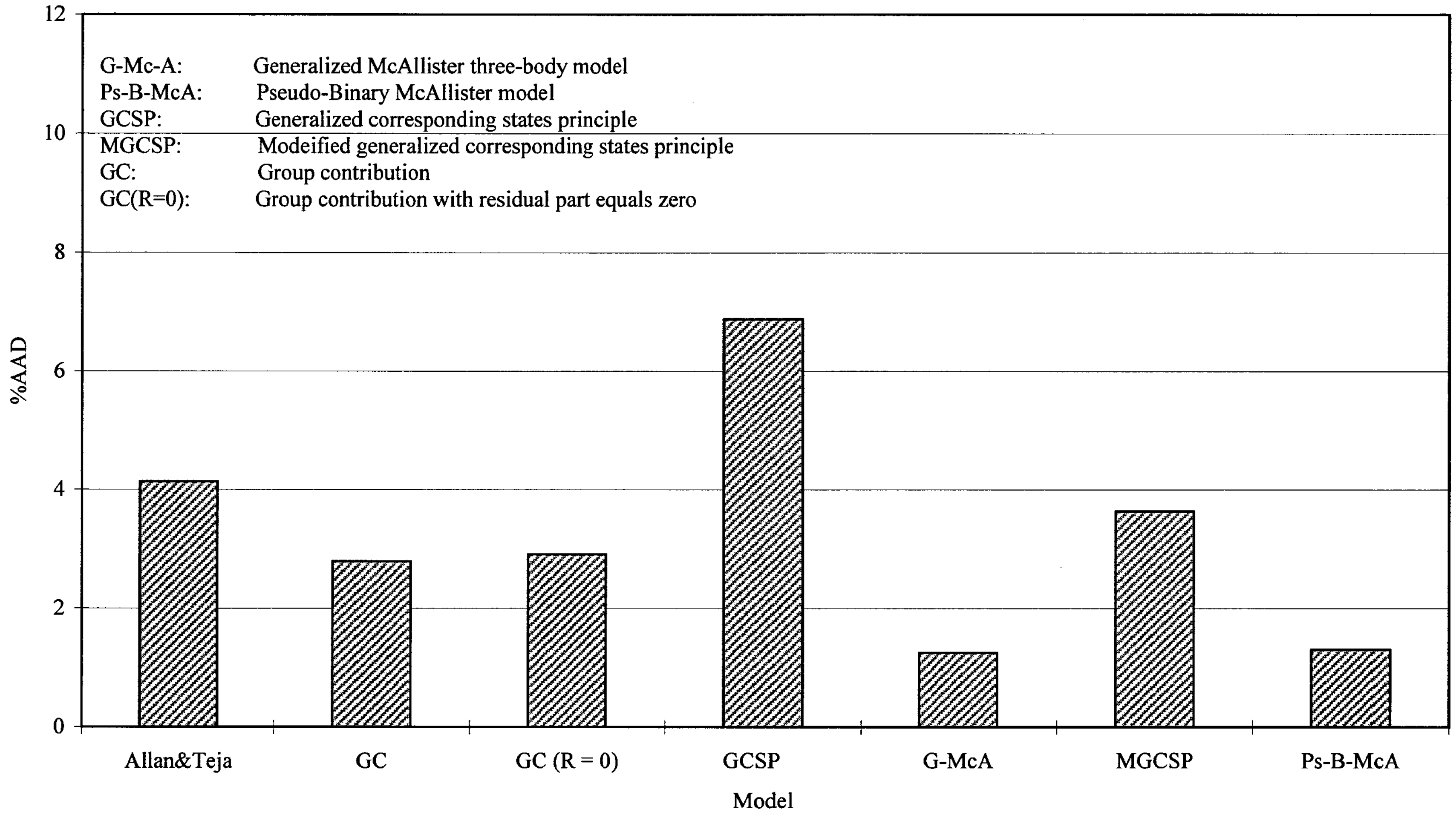

Figure 4.3: Model Comparison for Quaternary Systems containing Hexane at 293.15 K

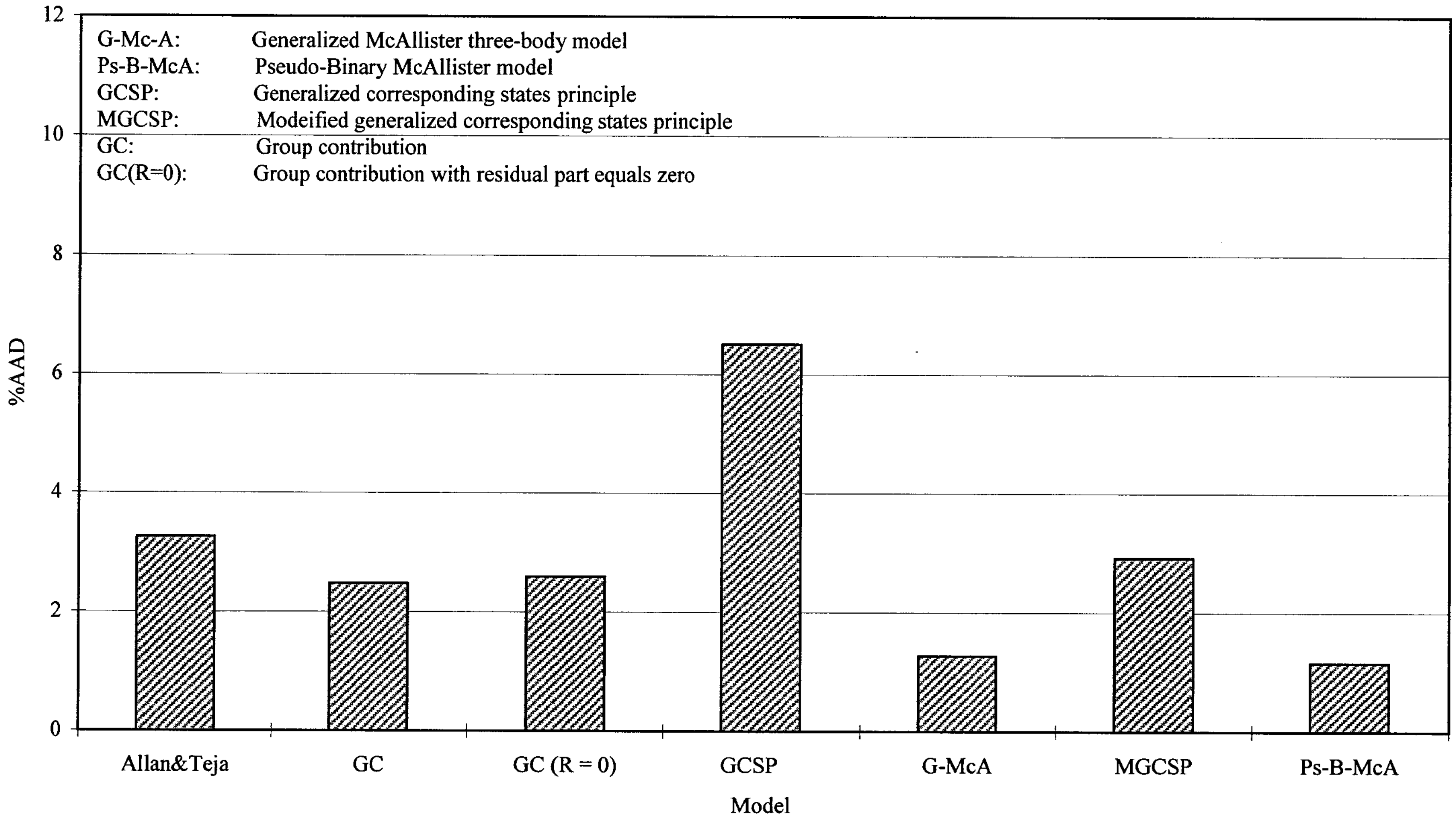

Figure 4.4: Model Comparison for Quaternary Systems containing Hexane at 298.15 K

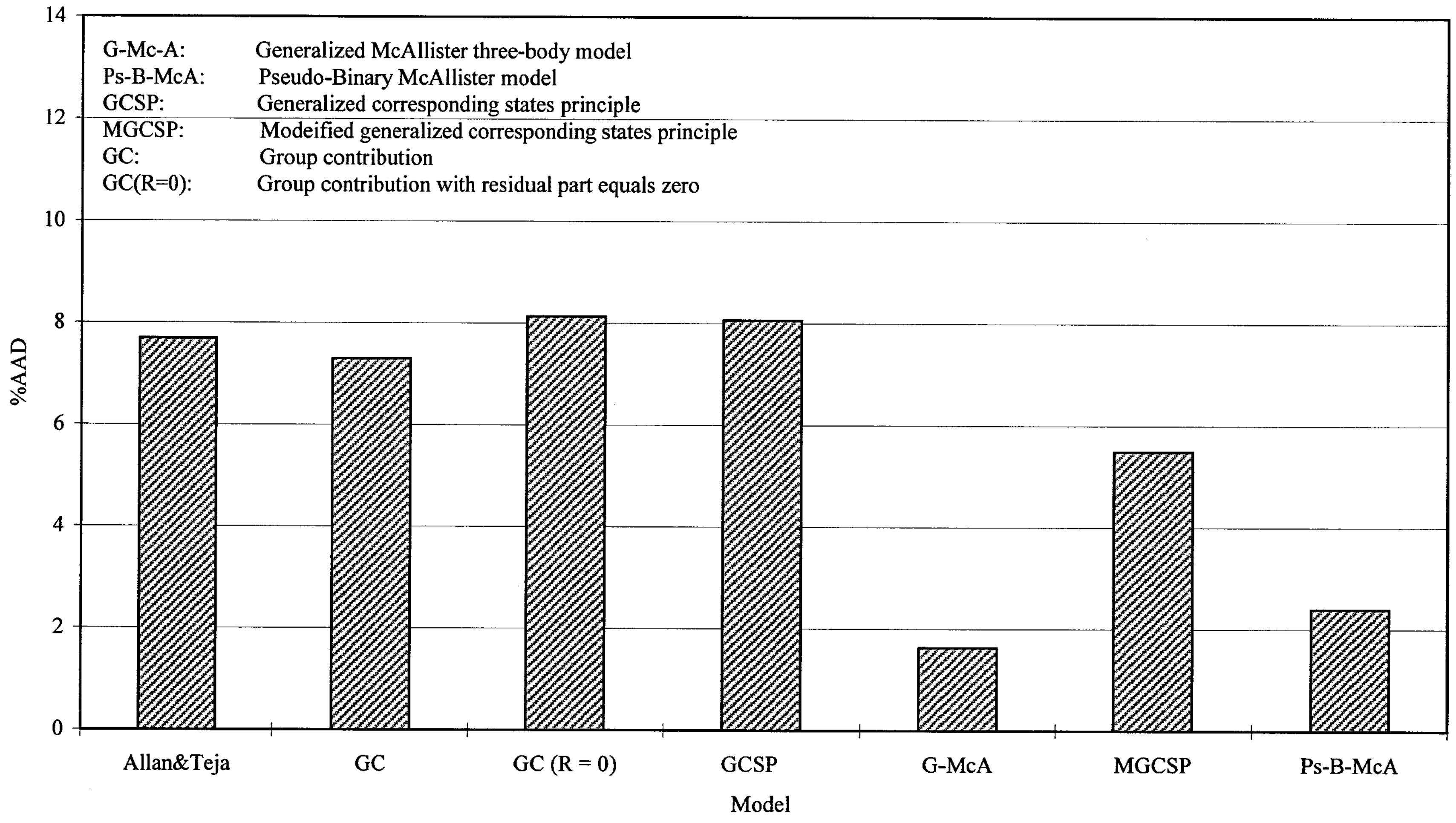

Figure 4.5: Model Comparison for Quinary System containing Heptane at 293.15 K

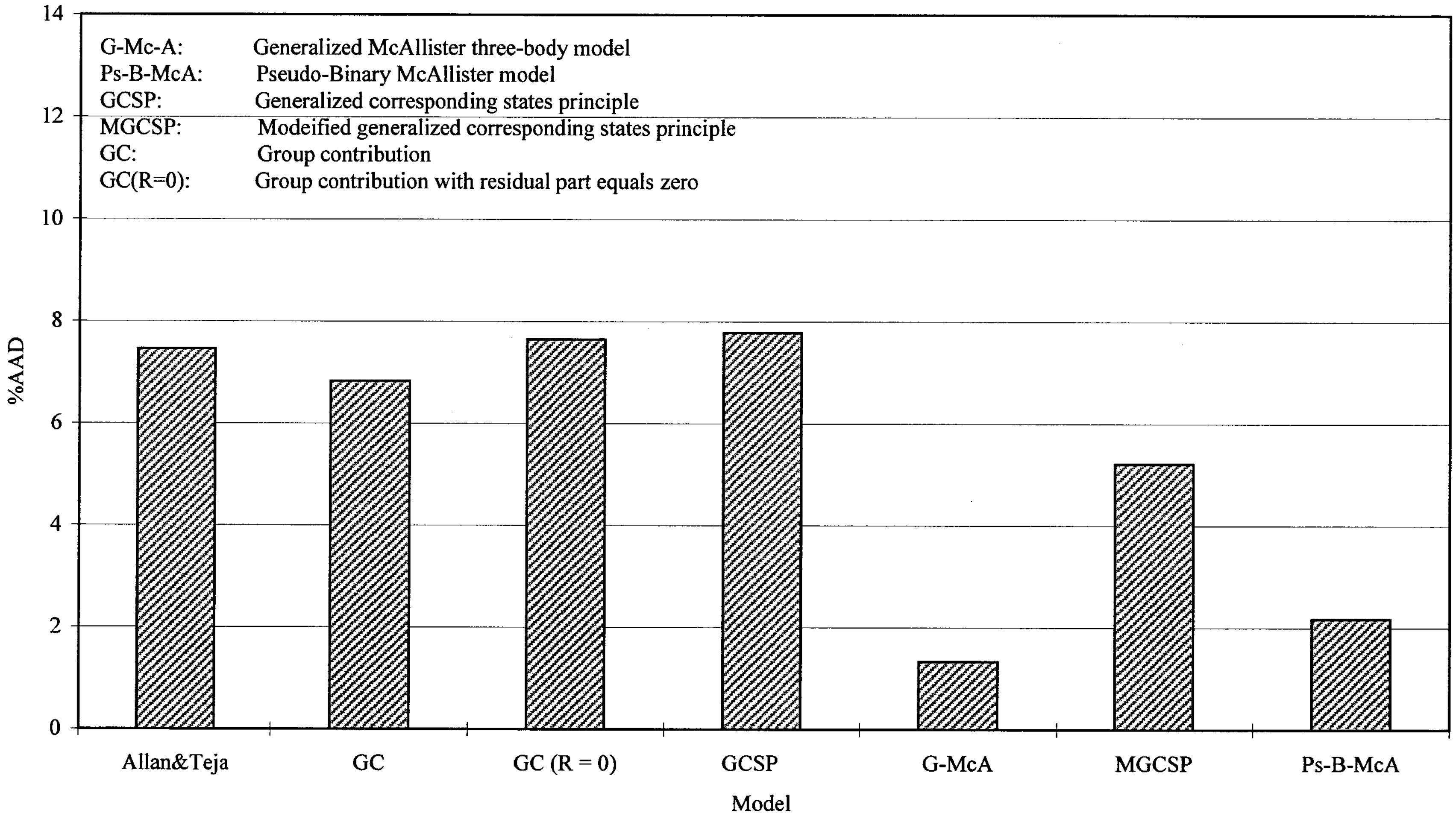

Figure 4.6: Model Comparison for Quinary System containing Heptane at 298.15 K

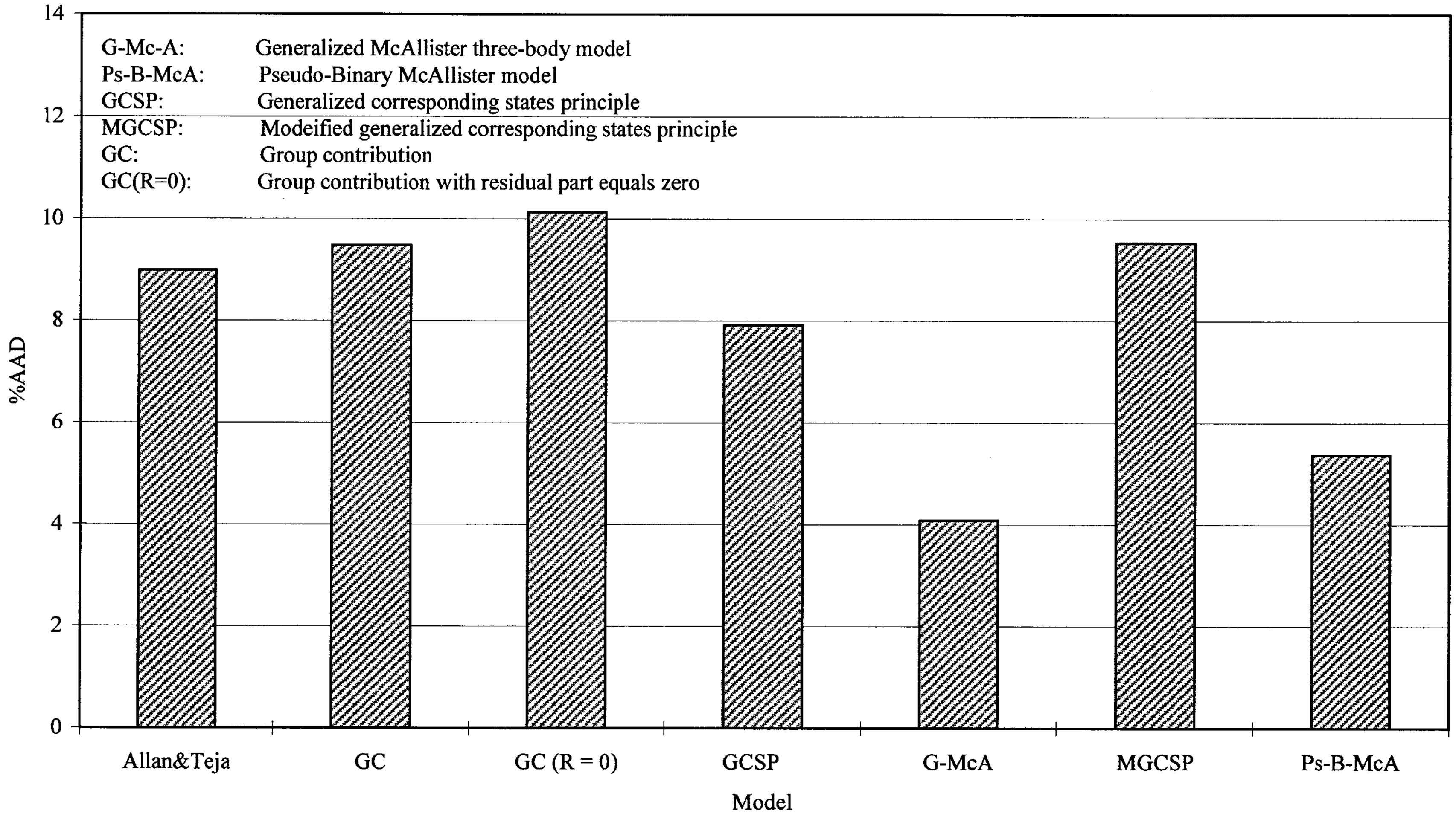

Figure 4.7: Model Comparison for Quaternary Systems containing Heptane at 293.15 K

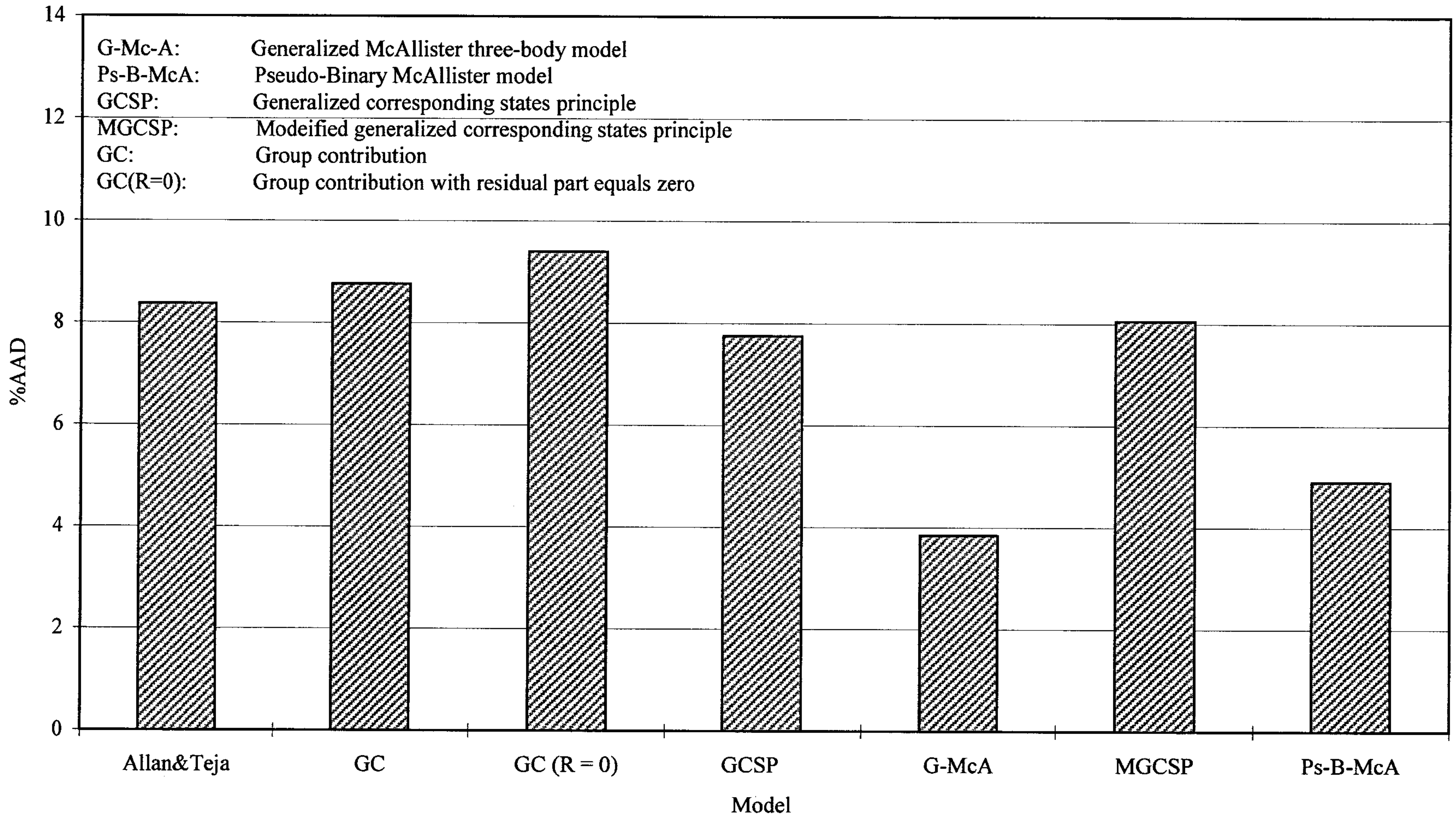

Figure 4.8: Model Comparison for Quaternary Systems containing Heptane at 298.15 K

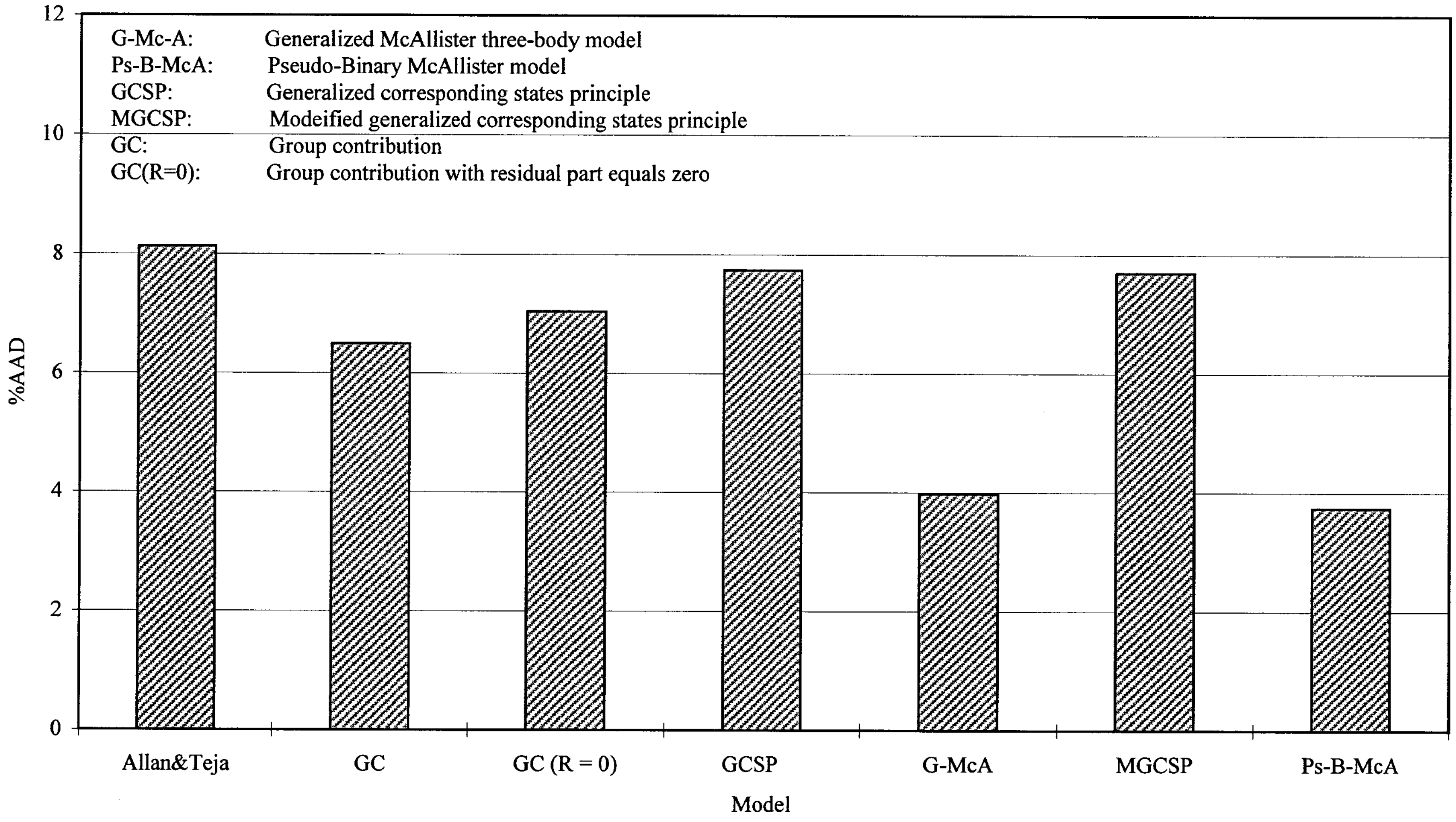

Figure 4.9: Model Comparison for Ternary Systems containing Heptane at 293.15 K

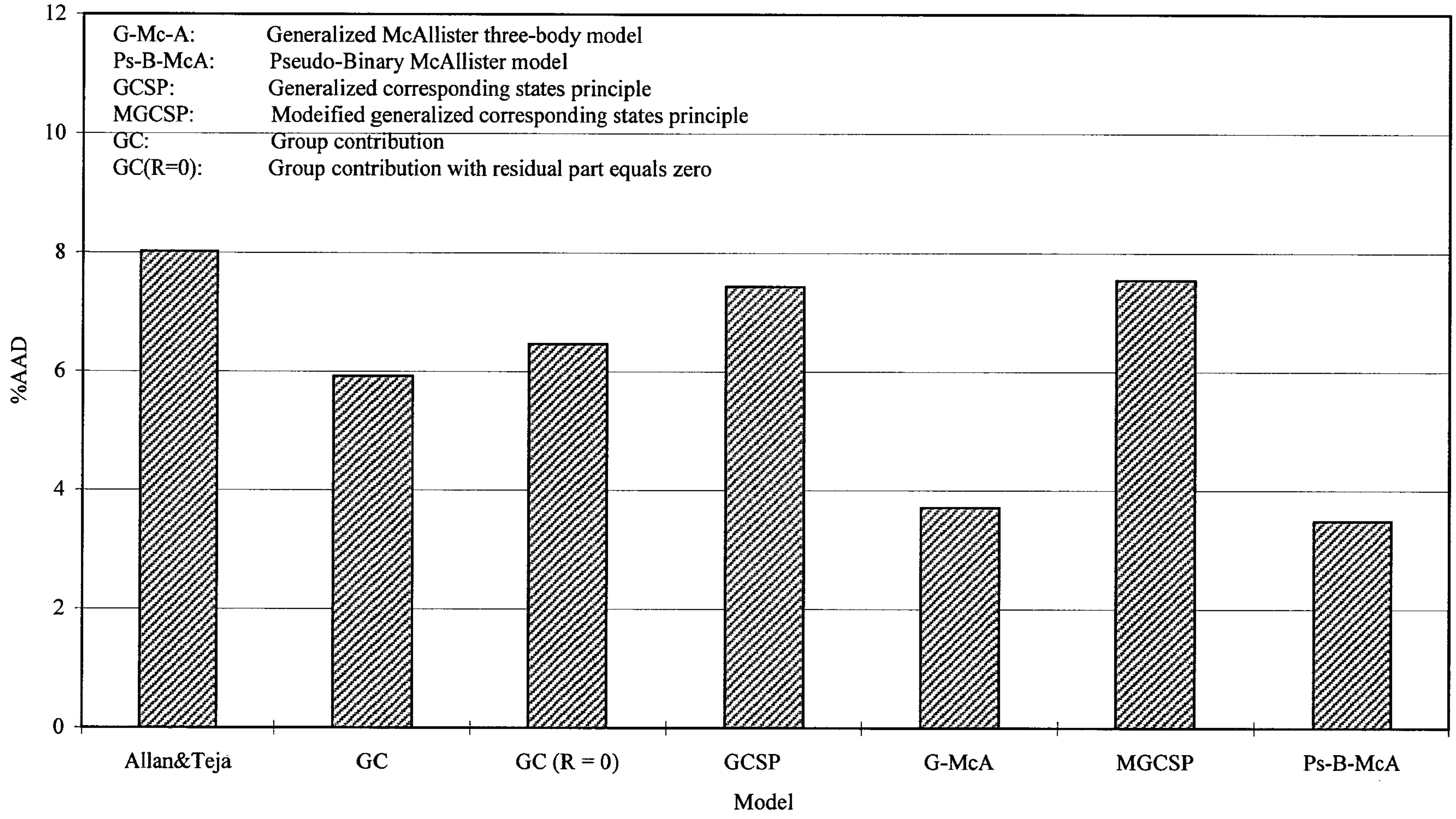

Figure 4.10: Model Comparison for Ternary Systems containing Heptane at 298.15 K

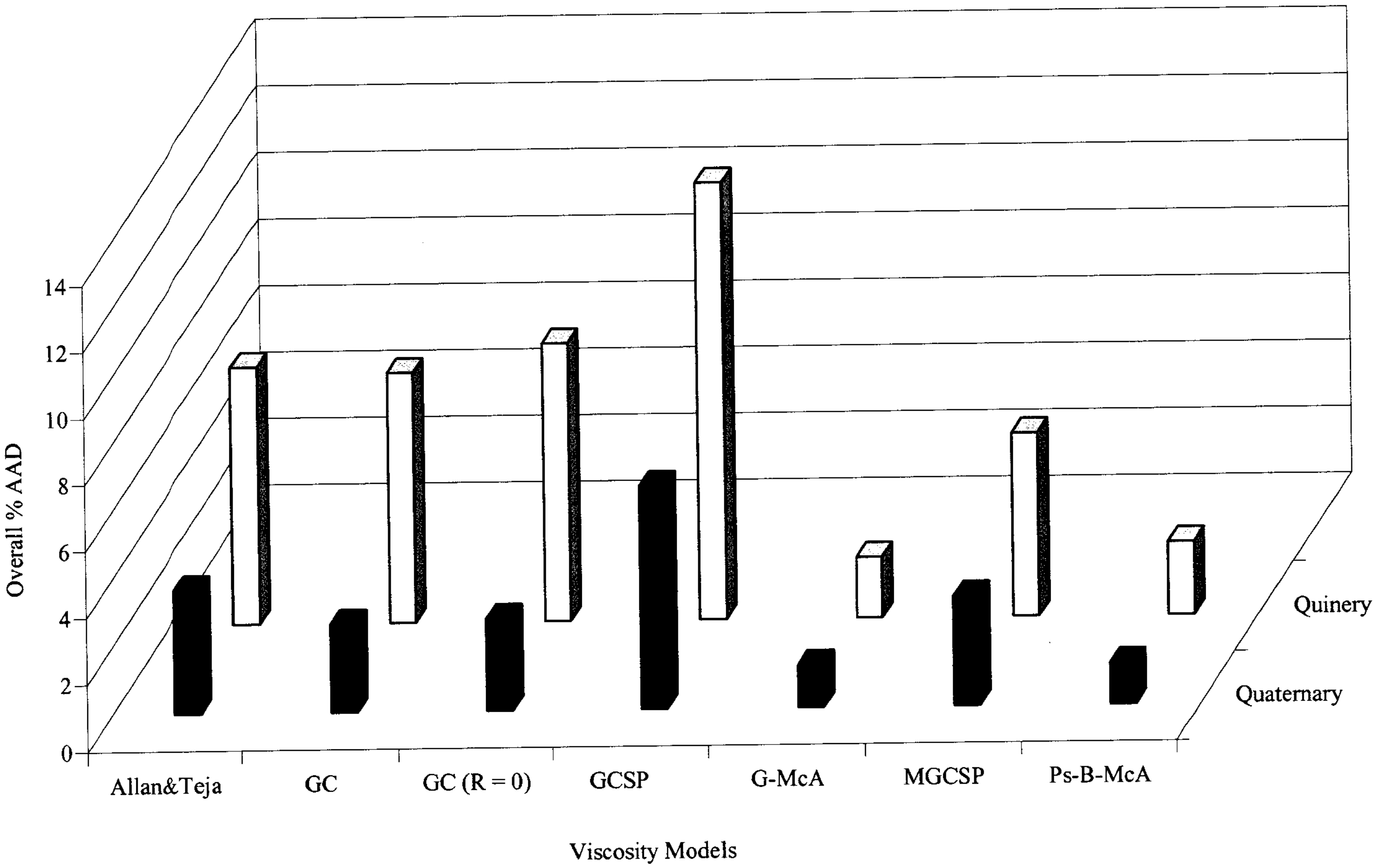

Figure 4.11: Results of Testing Various Viscosity Models by Using Data of Hexane-containing Regular Liquid Systems at 293.15 and 298.15 K

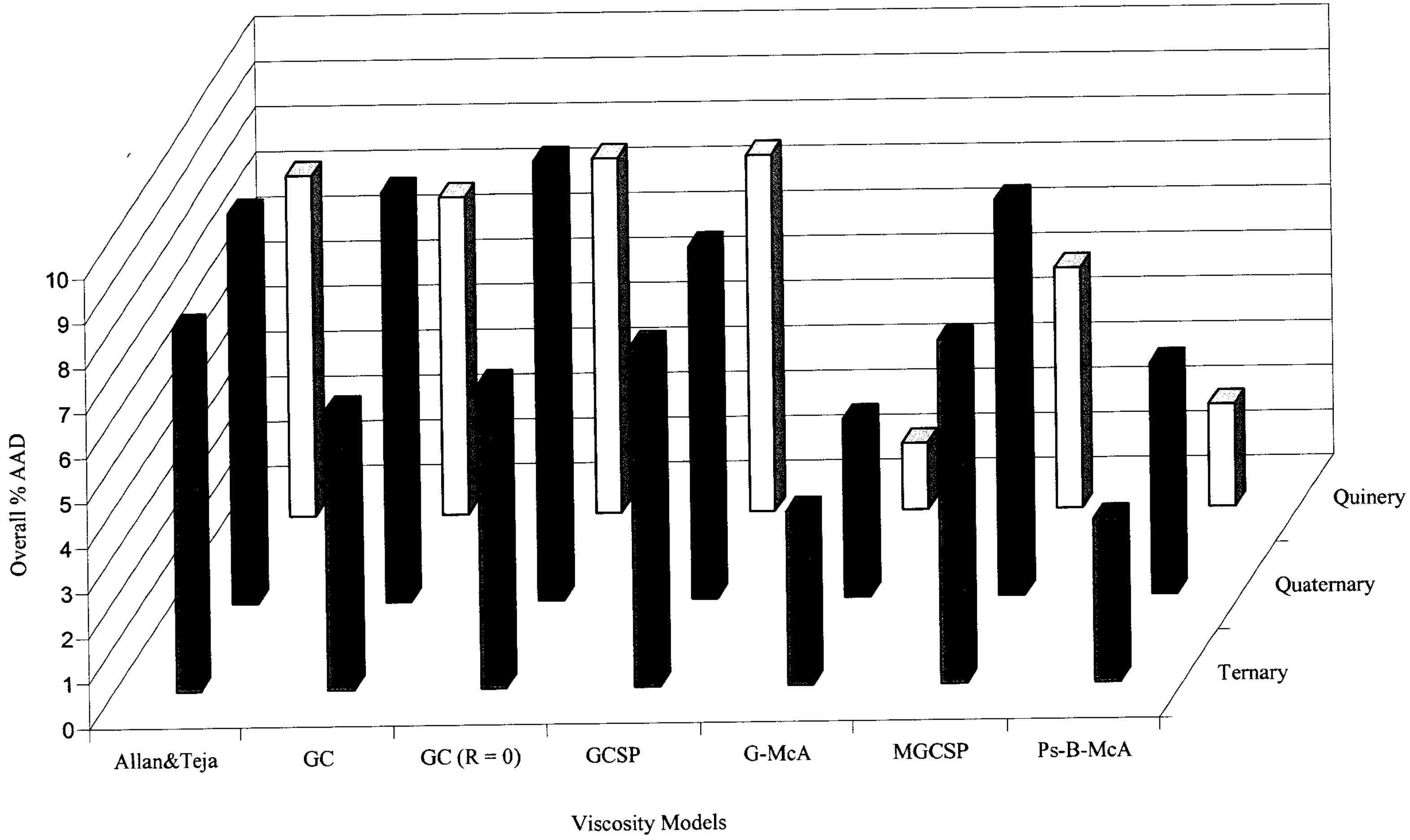

Figure 4.12: Results of Testing Various Viscosity Models by Using Data of Heptane-containing Regular Liquid Systems at 293.15 and 298.15 K

CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

The densities and viscosities of two quinary liquid systems and some of their quaternary and ternary subsystems at different temperatures were measured over the entire composition range. The data were used to test the predictive capability of some of the most widely used literature viscosity models. The following conclusions have been reached:

i) The densities and viscosities of the pure components constituting the liquid systems investigated in this study are in excellent agreement with those reported in the literature.

ii) The Generalized McAllister three-body collision model, as developed by Nhaesi and Asfour (2000a), exhibited the best overall predictive capability of all models. The overall average AAD is 2.5%.

iii) The pseudo-binary McAllister model, developed by Nhaesi and Asfour (2000b), ranks as second best in terms of its predictive capabilities. The overall average AAD is 2.9%

iv) The MGCSP, originally developed by Wu and Asfour (1992) for n-alkane solutions, and later modified by Nhaesi and Asfour (2000a), outperforms the original GCSP reported by Teja and Rice (1981). The overall average AADs are 6.1 % and 8.6%, respectively.

v) The GC-UNIMOD reported by Cao *et al.* (1993) gave an overall average AAD of 6.5%.

vi) The Allan and Teja correlation gave an overall average AAD is 7.2%.

5.2 Recommendations

The following suggestions are for further studies about this topic:

i) The results obtained show clearly that the McAllister three-body model is superior over the other models, but more viscosity data on regular liquid solutions should be obtained to be used in further investigation for the model.

ii) The effective carbon number of cyclohexane and similar "planar" type molecules should be further investigated since equation (4.7) seems to be over-estimating the ECN value of cyclohexane.

iii) Extending the McAlliester three-body model to be used for predicting the viscosities of associated solutions.

Nomenclature

a	group interaction energy parameter
AAD	average absolute deviation, %
B	constant generally used as a parameter
C	constant generally used as a parameter
D	constant generally used as a parameter
E	constant generally used as a parameter
ECN	effective carbon number
h	Plank's constant
i	index number
j	index number
k	index number
K	Boltzman's constant
M	molecular weight, g/mol
MAX	maximum deviation, %
N	number of components of the mixture, number of interaction parameters
$\overline{N}$	average number of carbon atoms of n-alkane liquid mixture
N	Avogadro's number
P	pressure, atm
q	area parameter of molecule
R	gas constant
r	rate at which a molecule moving under shear stress
r_0	rate at which a molecule moving as a result of thermal fluctuation

t	efflux time of the viscometer
T	temperatute, K
V	molar volume of the liquid, L/kmol
X	mole fraction, reduced transport property
x_{ij}	local mole fraction of component j around central molecule i
Z	compressibility factor

Greek Letters

ε	reciprocal of fluid viscosity at critical temperature
η	absolute viscosity
θ	shape factor
λ	center to center distance between a molecule and a hole in Eyring's theory
$\lambda_1, \lambda_2, \lambda_3$	intermolecular distance involved in Eyring's theory
ν	kinematic viscosity, m^2/s
ν_k^i	number of groups k in molecule
ν_{12}	McAllister three-body model binary interaction parameter
ν_{21}	McAllister three-body model binary interaction parameter
ν_{123}	McAllister three-body model interaction parameter
ν_{1112}	McAllister four-body model interaction parameter
ν_{2111}	McAllister four-body model interaction parameter
ξ_{ij}	binary interaction parameter in the Generalized Corresponding States Principle
ξ_{loc}	ξ based on local composition

ξ_i^c	pure component ξ value
ρ	density, kg/L
τ	oscillation period is seconds for the density meter
φ	shape factor
φ_i	fluidity, $1/\eta$
φ_{ji}	local volume fraction of component j around central molecule i
ω	Pitzer acentric factor

Subscripts

1,2,3,4,5	refer to various components in the mixture, respectively
123	refer to interaction between three molecules
c	critical properties
cm	pseudocritical properties
i,j	refer to ith and jth component in the mixture, respectively
ij	refer to interaction of type i-j
ijk	refer to interaction of type i-j-k
n	refers to nth component of the mixture
o	refers to the reference fluid
r	reduced properties
r1	refers to reference fluid 1
r2	refers to reference fluid 2

Superscripts

E	excess over the ideal solution property
o	refers to the reference fluid
r1	refers to reference fluid 1
r2	refers to reference fluid 2

Acronyms

AAD	Average absolute deviation, %
ASOG	Analytical Solution of Groups method
ECN	Effective carbon number
GC-UNIMOD	group contribution-viscosity thermodynamics model
GCSP	Generalized Corresponding States Principle
MAX	Maximum deviation, %
MGCSP	Modified Generalized Corresponding States Principle
TRAPP	TRAnsport Properties Prediction

References

ACS Applied Publications, "Physical properties of chemical compounds – II", number 22 of the Advances in Chemistry Series, American Chemical Society, 1959.

Abrams, D.S.; Prausnitz, J.M. "Statistical thermodynamics of liquid mixtures. A new expression for the excess Gibbs energy of partly and completely miscible systems", *AIChE J*, **21**, 116 (1975).

Allan, J.M.; Teja, A.S. "Correlation and prediction of the viscosity of defined and undefined hydrocarbon liquids", *Can. J. Chem. Eng.*, **69**, 986-991 (1991).

Asfour, Abdul-Fattah.; Cooper, E.F.; Wu, J; Zahran, R.R. "Prediction of the McAllister model parameters from pure components properties for liquid binary n-alkane systems", *I&ECRes*, **30**, 1666-1669 (1991).

Cao, W.; Fredenslund A.; Rasmussen, P. "Statistical thermodynamic model for viscosity of pure liquids and liquid mixtures", *Ind. Eng. Chem. Res*, **31**, 2603-2619 (1992).

Cao, W.; Knudsen, K.; Fredenslund A.; Rasmussen, P. "Simultaneous correlation of viscosity and vapor-liquid equilibrium", *Ind. Eng. Chem. Res*, **32**, 2077-2087 (1993a).

Cao, W.; Knudsen, K.; Fredenslund A.; Rasmussen, P. "Group-contribution viscosity predictions of liquid mixtures using UNIFAC-VLE parameters", *Ind. Eng. Chem. Res*, **32**, 2088-2092 (1993b).

Cooper, E.F., "Density and viscosity of n-alkane binary mixtures as a function of composition at several temperatures", M.A.Sc. Thesis, University of Windsor, Windsor, Canada, 1987.

Chandramouli, V.V.; Laddha, G.S. "Viscosity of ternary liquid mixtures", *Indian J. Technol.*, **1** (5), 199-203 (1963).

Derr, E.L.; Deal C.H. "Analytical solutions of groups: correlation of activity coefficients through structural group parameters", *I. Chem. E. Symp. Ser. No. 32* (Instn. Chem. Engrs., London) 3:40 (1969).

Derr E.L., Papadopoulos M. "Group interaction II. A test of group model on binary solutions of hydrocarbons", *J. Am. Chem. Soc.*, **81**, 2285 (1959).

Ely, J.F.; Hanley, H.J.M. "Prediction of transport properties. 1. Viscosity of fluids and mixtures", *I&ECFundam.*, **20**, 323-332 (1981).

Ewell, R.H.; Eyring, H. "Theory of the viscosity of liquids as a function of temperature and pressure", *J. Chem. Phys.*, **5**, 726 (1937).

Eyring, H. "Viscosity, plasticity and diffusion as examples of absolute reaction rates", *J. Chem. Phys.*, **4**, 283-291 (1936).

Fredenslund, A.; Jones, R.L.; Prausnitz, J.M. "Group-contribution estimation of activity coefficients in nonideal liquid mixtures", *AIChE J.*,**21**, 1086-1097 (1975).

Gomez-Diaz, D.; Mejuto, J. C.; Navaza, J. M. "Physicochemical Properties of Liquid Mixtures. Viscosity, Density, Surface Tension and Refractive Index of Cyclohexane + 2,2,4-Trimethylpentane Binary Liquid Systems from 25 °C to 50 °C" *J. Chem. Eng. Data,* **46 (3)**, 720-724 (2001).

Gregory, G.A., Letter to the editor, *Can J. Chem. Eng.*, **70**, 1037-1038 (1992).

Helfand, E.; Rice, S.A., "Principle of corresponding states for transport properties", *J. Chem. Phys.,* **32**, 1642 (1960).

Hafez, M. and S. Hartland "Densities and viscosities of binary systems toluene-acetone and and 4-methyl-2-pentanone- acetic acid at 20, 25, 35 and 45 °C", *J. Chem. Eng. Data,* **21 (2)**, 179-182 (1976).

Hwang, M. Wallace, W.B. "A corresponding states treatment for the viscosity of polar fluids", *I&EC Res.*, **26**, 1758-1766 (1987).

Joffe, J. "Combining rules for the third parameter in the pseudocritical method for mixtures", *I&EC Fundam.*, **10**, 532-533 (1971).

Kalidas, R.; Laddha, G.S. "Viscosity of ternary liquid mixtures", *J. Chem. Eng. Data,* **9**, 142-145 (1964).

Katayama, H. and Watanabe I. "Isobaric vapor-liquid equilibria of the n-heptane-toluene system at subatmospheric pressures", *J. Chem. Eng. Data.*, **25**, 107-110.

Langmuir, I. "The distribution and orientation of molecules", 3rd Colloid Symposium Monograph, The Chemical Catalog Company Inc., New York (1925).

Lee, B.I., Kesler, M.G. "A generalized thermodynamic correlation based on three-parameter corresponding states", *AIChE J.*, **21**, 510-527 (1975).

Letsou, A.; Stiel, L.I. "Viscosity of saturated nonpolar liquids at elevated pressures", *AIChE J.*, **19**, 409-411 (1973).

Lien, P. J.; Lin, H. M.; Lee, M. J.; Venkatesu, P. "Excess Molar Enthalpies of Dimethyl Carbonate with *o*-Xylene, *m*-Xylene, *p*-Xylene, Ethylbenzene, or Ethyl Benzoate at 298.15 K", *J. Chem. Eng. Data.*, **48 (1)**, 110-113.

McAllister, R.A. "The viscosity of liquid mixtures", *AIChE J.*, **6**, 427-431 (1960).

Mehrotra, Anil K.; Monnery W.D.; Svrcek W.Y. "A review of practical calculation methods for the viscosity of liquid hydrocarbons and their mixtures", *Fluid Phase Equil.*, **117**, 344-355 (1996).

Monnery W.D.; Svrcek W.Y., Mehrotra, Anil K "Viscosity, A critical review of practical predictive and correlative methods", *Can. J. Chem. Eng.*, **73**, 3-40 (1995).

Nayak, J. N.; Aralaguppi, M. I.; Aminabhavi, T. M. "Density, Viscosity, Refractive Index, and Speed of Sound in the Binary Mixtures of 1,4-Dioxane + Ethanediol, + Hexane, + Tributylamine, or + Triethylamine at (298.15, 303.15, and 308.15) K" *J. Chem. Eng. Data,* **48 (5)**, 1152-1156.

Nhaesi, A.H. "A study of the predictive models for the viscosity of multi-component liquid regular solutions", Ph.D. thesis, University of Windsor, Windsor, Canada, 1998.

Nhaesi, A.H., Asfour, Abdul-Fattah.A., "Predictive Models for the Viscosities of Multicomponent Liquid *n*-Alkane and Regular Solutions", *Can. J. Chem. Eng.*, **78**, 355-362 (2000a).

Nhaesi, A.H., Asfour, Abdul-Fattah.A., "Densities and Kinematic Viscosities of Ten Ternary Regular Liquid Systems at 293.15 and 298.15 K", *Can. J. Chem. Eng.*, **78**, 355-362 (2000b).

Perry, R.H.; Green, D. "Chemical Engineer's Handbook, 6th edition", McGraw-Hill, New York, 1984.

Pitzer, K.S.; Lippman, D.Z.; Curl, R.F.; Huggins, C.M.; Peterson, D.E. "The volumetric and thermodynamic properties of fluids. II. Compressibility factor, vapor pressure and entropy of vaporization", *J. Am. Chem. Soc.*, **77**, 3433-3440 (1995).

Ploecker, K.; Knapp, H.; Prausnitz, J.M. "Calculation of high-pressure vapor liquid equilibria from a corresponding-states correlation with emphasis on asymmetric mixtures", *I&EC Process Des. Dev.*, **17**, 324-332 (1977).

Prestone, G.T.; Chapman, T.W.; Prausnitz, J.M. "Transport properties of cryogenic liquids and their mixtures", Cryogenics, **7**, 274 (1967).

Redlich, O.; Derr, E.L.; Pierotti, G. "Group interaction I. A model for interaction in solution", *J. Am. Chem. Soc.*, **81**, 2283 (1959).

Reid, R.C.; Praunitz, J.M., Sherwood T.K. "The properties of gases and liquids", Mc-Graw Hill, New York, (1987).

Ritzoulis, G.; N. Papadopoulos, and D. Jannakoudakis, “Densities, viscosities and dielectric constants of acetonitrile + toluene at 15, 25 and 35 °C”, *J. Chem. Eng. Data,* **31**, 146-148 (1986).

Ronc, M.; Ratcliff, G.A.; Prediction of excess free energies of liquid mixtures by an analytic group solution model”, *Can. J. Chem. Eng.*, **49**, 875 (1971)

Rowlinson, J.S.; Watson, I.D. “The prediction of thermodynamic properties of fluids and fluid mixtures-I. The principle of corresponding states and its extensions”, *Chem. Eng. Sci.*, **24**, 1565-1574 (1969).

Sanni, S.A., and H.P. Hutchison, “Diffusivities and Densities fir Binary Liquid Mixtures”, *J. Chem. Eng. Data,* **18**, 317-322 (1973).

Teja, A.S.; Rice P. “Generalized corresponding states method for the viscosities of liquid mixtures”, *I&EC Fundam.*, **20**, 77-81 (1981).

Teja, A.S.; Thurner, P.A.; Pasumart, B. “Calculation of transport properties of mixtures synfuels process design”, *I&EC Process Des. Dev.*, **24**, 344-349 (1985).

Tham, M.J.; Gubbins, K.E. “Correspondence principle for transport properties of dense fluid”, *I&EC Fundam.*, **8**, 791 (1969).

“TRC Thermodynamics Tables Hydrocarbons”, Supplement number 92, Thermodynamics Research Center, Texas, 1986

Vitali, G. and Lobbia G.G. “Ultrasonic velocity in binary mixtures of alkyl-benzenes at 25 °C”, *J. Chem. Eng. Data,* **28**, 402-404 (1983).

Willman, B.; Teja, A.S. “Method for the prediction of pure-component vapor pressure in range 1 kpa to the critical pressure”, *Ind. Eng. Chem. Process Des. Dev.*,**24**, 1033-1036 (1985).

Wilson, G.M.; Deal C.H. “Activity coefficients and molecular structure”, *Ind. Eng. Chem. Fundamentals*, **1**, 20 (1962).

Wong, D.S.H.; Sandler, S.I.; Teja A.S. “Vapor liquid equilibrium calculations by use of a generalized corresponding states principle. I. New mixing Rules”, *I&EC Fundam.*,**23**, 38-44 (1984).

Wu, D.T. “Prediction of Viscosities of Liquid Mixtures by a Group Contribution Method. *Fluid Phase Equilibria*, **30**, 149-156 (1986).

Wu, J. "An experimental study of the viscometric and volumetric properties of C_8-C_{15} n-alkane binary and ternary systems at several temperatures". Ph.D. thesis, University of Windsor, Windsor, Canada, 1992.

Wu, J.; Asfour, Abdul-Fattah A. "Viscometric properties of n-alkane liquid mixtures", *Fluid Phase Equilibria*, **76**, 283-294 (1992).

Timmermans, J. "Physicho-cheimcal constants of pure organic compounds", Volume I, Intersceince, New York, 1950.

Timmermans, J. "Physicho-cheimcal constants of pure organic compounds", Volume II, Intersceince, New York, 1965.

APPENDICES

Appendix A

Raw Data of Viscosity and Density Measurements

Table A.1: Raw Data for the Quinary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)

X_1	X_2	X_3	X_4	X_5	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
						Temperature = 293.15 K		Temperature = 298.15 K	
0.0986	0.2921	0.4888	0.0493	0.0712	25A483	0.71738	348.98	0.71632	331.68
0.1967	0.1433	0.0518	0.1036	0.5046	25A498	0.71778	346.19	0.71672	329.65
0.1511	0.4904	0.0492	0.1006	0.2087	25A483	0.70671	315.78	0.70563	301.78
0.4932	0.1042	0.0563	0.1990	0.1473	25A498	0.70900	389.18	0.70797	368.56
0.1514	0.2555	0.1960	0.0531	0.3440	25A483	0.71625	340.19	0.71518	324.21
0.1986	0.1022	0.0476	0.4990	0.1526	25A498	0.71338	421.33	0.71231	398.03

Table A.2: Raw Data for the Quaternary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Cyclohexane(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.1019	0.4972	0.1537	0.2472	25A483	0.70612	336.66	0.70504	320.52
0.1562	0.1977	0.5956	0.0505	25A498	0.71907	362.24	0.71802	344.15
0.0747	0.2779	0.4496	0.1978	25A483	0.71638	364.98	0.71532	346.47
0.5816	0.0474	0.1887	0.1823	25A498	0.70913	407.69	0.70811	385.11
0.1029	0.0458	0.2476	0.6037	25A483	0.71761	473.78	0.71653	445.35
0.0530	0.6952	0.0955	0.1563	25A498	0.7017	295.41	0.70061	281.86
0.4429	0.1540	0.1950	0.2081	25A483	0.70904	398.79	0.70801	378.79
0.0508	0.0987	0.2852	0.5653	25A498	0.71802	440.61	0.71695	415.28

Table A.3: Raw Data for the Quaternary System: Octane(1)-Hexane(2)-Ethylbenzene(3)-Toluene(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.1013	0.4867	0.1688	0.2432	25A483	0.71016	306.85	0.70907	292.65
0.1477	0.1973	0.6085	0.0465	25A498	0.72017	359.05	0.71911	340.45
0.0508	0.2970	0.4658	0.1864	25A483	0.71951	341.28	0.71844	324.8
0.4652	0.0581	0.2162	0.2605	25A498	0.71435	369.86	0.71332	351.49
0.1054	0.0597	0.2008	0.6341	25A483	0.72637	368.85	0.72529	349.85
0.0575	0.5928	0.1029	0.2468	25A498	0.70744	284.14	0.70636	281.86
0.4513	0.1506	0.2029	0.1952	25A483	0.71172	366.37	0.71068	348.76
0.0523	0.1054	0.2870	0.5553	25A498	0.72669	355.15	0.72561	340.86

Table A.4: Raw Data for the Quinary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)-Toluene(5)

X_1	X_2	X_3	X_4	X_5	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
						Temperature = 293.15 K		Temperature = 298.15 K	
0.1109	0.2602	0.5324	0.0604	0.0360	25A483	0.70888	377.67	0.70785	358.17
0.2002	0.1371	0.0580	0.1022	0.5024	25A498	0.70859	359.73	0.70751	341.55
0.1446	0.4515	0.1479	0.3373	0.2145	25A483	0.69906	355.21	0.69802	337.92
0.4967	0.0917	0.0548	0.1982	0.1586	25A498	0.6998	399.78	0.69878	378.16
0.1450	0.2181	0.1763	0.0657	0.3949	25A483	0.70414	298.96	0.70519	313.39
0.2087	0.0904	0.0541	0.4948	0.1519	25A498	0.69979	348.07	0.70085	367.32

Table A.5: Raw Data for the Quaternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Cyclohexane(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.1110	0.4737	0.1459	0.2694	25A483	0.69830	385.73	0.69726	365.68
0.1611	0.1868	0.5878	0.0644	25A498	0.70971	379.38	0.70868	359.94
0.0600	0.2772	0.4558	0.2070	25A483	0.70748	388.38	0.70644	368.25
0.6018	0.0486	0.1930	0.1566	25A498	0.69941	409.55	0.69840	387.65
0.1086	0.0476	0.2520	0.5918	25A483	0.70792	474.73	0.70687	448.17
0.0582	0.6711	0.1134	0.1573	25A498	0.69539	351.31	0.69435	334.14
0.4618	0.1427	0.2064	0.1890	25A483	0.69994	410.81	0.69892	389.03
0.0546	0.0923	0.3040	0.5491	25A498	0.70892	447.7	0.70786	422.12

Table A.6: Raw Data for the Quaternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)-Toluene(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.0691	0.4838	0.1711	0.2760	25A483	0.70243	343.93	0.70139	327.5
0.1515	0.1760	0.6127	0.0597	25A498	0.71135	374.7	0.71032	355.25
0.0548	0.2728	0.5925	0.0798	25A483	0.71121	365.99	0.71017	348.43
0.4983	0.0474	0.1958	0.2584	25A498	0.70414	376.05	0.70311	356.81
0.1000	0.0452	0.1972	0.6576	25A483	0.71742	374.04	0.71637	353.53
0.0599	0.5567	0.1134	0.2700	25A498	0.70075	328.79	0.69971	313.28
0.4517	0.1317	0.2104	0.2062	25A483	0.70297	378.7	0.70195	360.1
0.0537	0.0923	0.3007	0.5533	25A498	0.7175	365.46	0.71645	346.7

Table A.7: Raw Data for the Quaternary System: Octane(1)-Heptane(2)-Cyclohexane(3)-Toluene(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.1124	0.4611	0.1661	0.2603	25A483	0.69949	359.31	0.69845	341.33
0.1523	0.0919	0.6046	0.1512	25A498	0.70408	456.4	0.70301	429.55
0.0539	0.2613	0.4757	0.2091	25A483	0.70324	413.21	0.70217	389.82
0.4017	0.0457	0.2010	0.3515	25A498	0.70359	387.5	0.70256	367.11
0.0962	0.1336	0.0603	0.7099	25A483	0.71284	357.23	0.71177	338.69
0.0532	0.7312	0.1053	0.1104	25A498	0.69476	354.09	0.69373	338.2
0.0515	0.0900	0.2988	0.5596	25A483	0.71228	387.41	0.7112	367.25
0.4458	0.2237	0.2596	0.0709	25A498	0.69655	399.85	0.69552	378.49

Table A.8: Raw Data for the Quaternary System: Heptane (1)- Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.0879	0.5126	0.1524	0.2471	25A483	0.71752	401.3	0.71646	380.67
0.1329	0.1135	0.4007	0.3528	25A498	0.71118	397.83	0.71011	376.94
0.0462	0.3050	0.4480	0.2007	25A483	0.71418	432.88	0.7131	412.25
0.5125	0.0543	0.3522	0.0809	25A498	0.69867	373.69	0.69761	356.31
0.1724	0.1802	0.0551	0.5924	25A483	0.71543	362.27	0.71436	344.58
0.1768	0.5958	0.1049	0.1225	25A498	0.71497	381.41	0.71393	362.29
0.1348	0.1134	0.6994	0.0524	25A483	0.70666	488.25	0.70557	458.11
0.0481	0.1077	0.3027	0.5415	25A498	0.71614	390.29	0.71506	369.2

Table A.9: Raw Data for the Quaternary System: Octane (1)- Ethylbenzene(2)-Cyclohexane(3)-Toluene(4)

X_1	X_2	X_3	X_4	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
					Temperature = 293.15 K		Temperature = 298.15 K	
0.1030	0.5168	0.1707	0.2094	25A483	0.71935	404.00	0.71831	383.02
0.1528	0.1096	0.3942	0.3434	25B350	0.71068	440.39	0.70962	416.7
0.0554	0.1031	0.6586	0.1829	25B349	0.71022	503.94	0.70914	473.02
0.4840	0.0562	0.2141	0.2457	25B350	0.70536	404.10	0.70433	383.66
0.1998	0.1475	0.0596	0.5930	25A483	0.71425	378.98	0.7132	359.54
0.0993	0.5996	0.1062	0.1949	25A498	0.71784	402.00	0.7168	380.73
0.0525	0.0986	0.6948	0.1541	25B349	0.70984	513.32	0.70876	481.7
0.0593	0.3498	0.2973	0.2936	25B350	0.71618	434.66	0.71512	411.12

Table A.10: Raw Data for the Ternary System: Octane(1)-Heptane(2)-Ethylbenzene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.5078	0.1080	0.3841	25A483	0.70266	393.37	0.70164	373.67
0.2283	0.1728	0.5989	25A498	0.71091	381.58	0.70987	362.10
0.3525	0.3404	0.3071	25B349	0.70000	384.81	0.69898	365.85
0.6567	0.2228	0.1204	25B350	0.69534	411.70	0.69433	390.69
0.7961	0.1232	0.0807	25A483	0.69467	406.73	0.69366	385.99
0.1030	0.6259	0.2712	25A498	0.69827	340.88	0.69724	324.94
0.4259	0.3229	0.2512	25B349	0.69852	389.16	0.6975	369.72
0.5053	0.3460	0.1487	25B350	0.69573	397.39	0.69471	377.90
0.2131	0.4347	0.3522	25A483	0.70110	362.90	0.70008	344.76
0.4167	0.5215	0.0617	25A498	0.69299	364.57	0.69197	346.74

Table A.11: Raw Data for the Ternary System: Octane(1)-Heptane(2)-Toluene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.5409	0.3113	0.1478	25B349	0.69515	389.96	0.69414	369.9
0.2286	0.5408	0.2306	25B350	0.69636	362.02	0.69532	343.85
0.3271	0.2149	0.4580	25A483	0.7032	357.51	0.70216	340.53
0.6400	0.0999	0.2600	25A498	0.69827	378.97	0.69725	360.86
0.7775	0.0624	0.1601	25B349	0.69616	412.78	0.69515	391.71
0.0911	0.1415	0.7674	25B350	0.71374	366.09	0.71267	347.73
0.2092	0.3478	0.4431	25A483	0.70253	346.37	0.70148	329.78
0.3323	0.4415	0.2262	25A498	0.69656	350.06	0.69553	333.13
0.2311	0.6804	0.0885	25B349	0.69268	361.54	0.69165	344.06
0.3418	0.0386	0.6196	25B350	0.7085	380.37	0.70745	361.5

Table A.12: Raw Data for the Ternary System: Octane(1)-Cyclohexane(2)-Toluene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.5017	0.1098	0.3885	25A483	0.70329	390.85	0.70227	370.74
0.1987	0.1600	0.6413	25A498	0.71239	373.45	0.71133	353.88
0.3443	0.3503	0.3054	25B349	0.70459	432.67	0.70354	409.5
0.6440	0.2343	0.1217	25B350	0.69799	445.84	0.69697	422.48
0.0666	0.8754	0.0580	25A483	0.7065	591.28	0.70541	550.28
0.1040	0.6699	0.2261	25A498	0.70798	479.2	0.7069	450.4
0.2056	0.2168	0.5777	25B349	0.71123	399.63	0.71017	378.85
0.4979	0.2989	0.2032	25B350	0.7008	443.33	0.69977	419.46
0.1953	0.0568	0.7478	25A483	0.71421	370.56	0.71315	351.6
0.3079	0.5445	0.1476	25A498	0.70295	459.49	0.70191	432.34

Table A.13: Raw Data for the Ternary System: Octane(1)-Ethylbenzene(2)-Toluene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.5013	0.1047	0.3940	25B349	0.70507	394.36	0.70405	375.27
0.1992	0.1814	0.6194	25B350	0.71529	391.42	0.71424	371.22
0.3460	0.3467	0.3073	25A483	0.71046	386.63	0.70943	367.26
0.6385	0.2291	0.1323	25A498	0.70172	389.81	0.70071	370.68
0.0796	0.8133	0.1071	25B349	0.72047	427.63	0.71942	402.94
0.1099	0.6535	0.2365	25B350	0.71922	419.94	0.71818	398.34
0.2050	0.2121	0.5829	25A483	0.71512	378.41	0.71407	358.87
0.2757	0.2803	0.4440	25A498	0.71268	376.9	0.71164	357.58
0.1962	0.0525	0.7513	25B349	0.71524	379.94	0.71418	360.18
0.3145	0.4845	0.2011	25B350	0.71177	412.6	0.71074	390.14

Table A.14: Raw Data for the Ternary System: Octane(1)-Heptane(2)-Cyclohexane(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.5030	0.1011	0.3960	25A483	0.69685	455.39	0.69582	428.75
0.1965	0.1606	0.6428	25A498	0.70005	479.47	0.69899	450.04
0.3557	0.3279	0.3164	25B349	0.69520	430.69	0.69418	407.43
0.6235	0.2089	0.1676	25B350	0.69402	434.83	0.69301	411.86
0.1119	0.8070	0.0810	25A483	0.69110	352.67	0.69006	335.17
0.1079	0.6529	0.2392	25A498	0.69322	367.38	0.69219	348.95
0.2042	0.2090	0.5868	25B349	0.69905	485.09	0.69800	456.62
0.5054	0.2819	0.2128	25B350	0.69423	431.12	0.69322	408.1
0.1534	0.0946	0.7520	25A483	0.70218	540.45	0.70112	505.39
0.3067	0.5225	0.1708	25A498	0.69300	377.48	0.69196	358.07

Table A.15: Raw Data for the Ternary System: Heptane(1)-Ethylbenzene(2)-Cyclohexane(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.4935	0.1008	0.4057	25B349	0.69871	410.53	0.69766	388.78
0.1995	0.1518	0.6487	25B350	0.70513	491.81	0.70406	462.78
0.3365	0.3542	0.3093	25A483	0.70578	392.74	0.70473	373.27
0.6629	0.2501	0.0869	25A498	0.69846	341.66	0.69742	325.41
0.1545	0.6886	0.1569	25B349	0.71504	414.56	0.71399	393.65
0.1038	0.6610	0.2352	25B350	0.71570	433.06	0.71465	409.88
0.1895	0.2488	0.5617	25A483	0.70697	449.47	0.70591	423.78
0.4709	0.2904	0.2388	25A498	0.70228	366.32	0.70123	348.22
0.1871	0.0550	0.7578	25B349	0.70381	527.84	0.70274	494.55
0.2951	0.5494	0.1555	25B350	0.70987	398.63	0.70882	378.52

Table A.16: Raw Data for the Ternary System: Octane(1)-Ethylbenzene(2)-Cyclohexane(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.4946	0.1011	0.4043	25A483	0.70009	457.58	0.69907	431.95
0.1974	0.1536	0.6491	25A498	0.70564	489.81	0.70458	461.42
0.3338	0.3512	0.3149	25B349	0.70644	447.36	0.7054	423.32
0.6095	0.2235	0.1670	25B350	0.70050	440.69	0.69948	417.39
0.1528	0.6993	0.1478	25A483	0.71536	419.48	0.71432	396.76
0.1021	0.6666	0.2313	25A498	0.71588	419.11	0.71484	396.69
0.1968	0.2249	0.5783	25B349	0.70679	496.88	0.70574	468.27
0.4966	0.2962	0.2072	25B350	0.70311	441.64	0.70209	418.20
0.1906	0.0570	0.7524	25A483	0.70419	539.21	0.70313	504.55
0.2981	0.5517	0.1502	25A498	0.71023	405.10	0.70920	384.49

Table A.17: Raw Data for the Ternary System: Heptane(1)-Ethylbenzene(2)-Toluene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.4975	0.1023	0.4002	25B349	0.70434	342.04	0.70328	326.8
0.2032	0.1572	0.6396	25B350	0.71505	368.32	0.71398	349.84
0.3418	0.3435	0.3146	25A483	0.71022	351.57	0.70917	335.12
0.6473	0.2337	0.1190	25A498	0.70007	329.59	0.69903	313.81
0.1296	0.7199	0.1505	25B349	0.71851	404.41	0.71747	383.61
0.0949	0.6681	0.2370	25B350	0.7198	410.65	0.71875	389.56
0.2052	0.2205	0.5742	25A483	0.71509	356.92	0.71403	339.78
0.5008	0.2995	0.1997	25A498	0.70479	336.27	0.70374	319.76
0.1935	0.0565	0.7500	25B349	0.71531	360.25	0.71424	342.24
0.3012	0.5445	0.1543	25B350	0.71202	380.8	0.71098	362.43

Table A.18: Raw Data for the Ternary System: Heptane(1)-Cyclohexane(2)-Toluene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15	
0.4920	0.1114	0.3966	25A483	0.70254	339.25	0.70148	323.68
0.1927	0.1501	0.6572	25A498	0.71268	350.63	0.71161	333.05
0.3297	0.3651	0.3052	25B349	0.70422	394.32	0.70315	373.85
0.6326	0.2396	0.1279	25B350	0.69639	374.79	0.69534	356.27
0.1464	0.7008	0.1529	25A483	0.70586	484.06	0.70478	454.52
0.1019	0.6700	0.2282	25A498	0.70797	463.98	0.70688	435.62
0.1946	0.2259	0.5794	25B349	0.71136	378.34	0.71029	358.73
0.4936	0.3023	0.2041	25B350	0.69974	385.21	0.69868	365.86
0.1894	0.0538	0.7568	25A483	0.71447	350.21	0.71339	332.56
0.3008	0.5466	0.1525	25A498	0.70259	418.07	0.70152	394.97

Table A.19: Raw Data for the Ternary System: Ethylbenzene(1)-Cyclohexane(2)-Toluene(3)

X_1	X_2	X_3	Viscometer Number	Density Meter Reading	Efflux Time (s)	Density Meter Reading	Efflux Time (s)
				Temperature = 293.15 K		Temperature = 298.15 K	
0.5033	0.1013	0.3954	25B349	0.72187	420.79	0.7208	398.16
0.1977	0.1497	0.6526	25B350	0.72094	410.86	0.71987	388.99
0.3354	0.3572	0.3074	25A483	0.71752	426.46	0.71645	403.46
0.6430	0.2330	0.1240	25A498	0.71981	418.58	0.71876	396.4
0.1520	0.6940	0.1540	25B349	0.71196	517.35	0.71087	484.38
0.1055	0.6570	0.2374	25B350	0.71244	504.67	0.71135	473.4
0.2000	0.2211	0.5789	25A483	0.7197	401.14	0.71862	379.48
0.4964	0.3000	0.2036	25A498	0.7186	420.25	0.71754	395.79
0.1950	0.0521	0.7529	25B349	0.72268	400.6	0.72161	378.76
0.2965	0.5498	0.1537	25B350	0.71441	483.08	0.71333	454.53

Appendix B

Estimated Experimental Errors

B.1 Density Measurements

The equation used with the density meter is:

$$\rho = \frac{A\tau^2}{1 - B\tau^2} - C \tag{B.1}$$

when equation (B.1) is differentiated with respect to the period of vibration, T, equation (B.2) is obtained.

$$d\rho = \frac{2A\tau}{\left(1 - B\tau^2\right)^2} d\tau \tag{B.2}$$

During the study, two measuring cells were used as mentioned in chapter 3, A DMA 602 and a DMA 602T. The maximum fluctuation in any of them in density meter readings is $1x10^{-5}$ second.

For the DMA 602 measuring cell, the maximum value of τ was 0.72669 seconds at 293.15 K, substituting this value into equation (B.2) with the respected values for A, B and C, the maximum predicted error is calculated as:

$$d\rho = \frac{2(3.951236)(0.72669)}{[1 + 0.04134048 \times (0.72669)^2]^2} x10^{-5} = 5.5 \times 10^{-5} \text{ kg/L}$$

As for the DMA 602T measuring cell, the maximum value for T was 0.72268 seconds at 293.15 K. Following the same procedure, the maximum predicted error is

$$d\rho = \frac{2(3.939776)(0.72268)}{[1 + 0.0365093 \times (0.72268)^2]^2} x10^{-5} = 5.5 \times 10^{-5} \text{ kg/L}$$

B.2 Viscosity Measurement

From Chapter 3, the viscometer equation is:

$$\nu = Ct - \frac{E}{t^2} \tag{B.3}$$

Differentiating equation (B.3) with respect to the efflux time, t, gives

$$d\nu = \left(C + \frac{E}{3t^3} \right) dt \tag{B.4}$$

For each system at each temperature, at least three measurements of efflux time were taken. The error in measuring the efflux time did not exceed $\pm 0.1\%$.

In order to calculate the maximum expected error in kinematic viscosity, dt can be assumed to be equal to 0.1% of the maximum efflux time measured by every viscometer.

Table B.1 shows the maximum predicted error in the kinematic viscosity for each viscometer used.

Table B.1: Estimated errors in measuring kinematic viscosity

Viscometer Number	Efflux Time (s)	dt	dν
25A483	591.28	0.59	1.066
25A498	489.81	0.49	0.906
25B349	517.35	0.52	0.917
25B350	504.67	0.5	0.872

VITA AUCTORIS

NAME	Omar Medhat El-Hadad
PLACE OF BIRTH	Alexandria, Egypt
YEAR OF BIRTH	1979
EDUCATION	Alexandria University, Alexandria, Egypt 1996-2001 B.Sc (Chemical Engineering) University of Windsor, Windsor, Canada 2002-2004 M.A.Sc. (Environmental Engineering)