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AN EXPERIMENTAL STUDY OF THE VISCOMETRIC AND VOLUMETRIC PROPERTIES OF C_8 - C_{15} N-ALKANE BINARY AND TERNARY SYSTEMS AT SEVERAL TEMPERATURES

by

Jiangning Wu

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ABSTRACT

The viscosities and densities of five ternary C_8 - C_{15} n-alkane liquid systems and their corresponding eight binary subsystems have been measured at 293.15, 298.15, 308.15 and 313.15 K over the entire composition range.

The experimental and literature viscosity data were used to test and modify some existing viscosity predictive models.

The technique proposed earlier by Asfour *et al.* (1991) for the prediction of the dependence of viscosities of binary n-alkane systems on composition has been extended to cover ternary mixtures.

A pseudo-binary mixture model has been developed in this study to modify the Generalized Corresponding States Principle (GCSP) proposed earlier by Teja and Rice (1981) for either prediction or correlation of the viscosity of liquid mixtures. The proposed modification of the GCSP method has the following advantages for n-alkane liquid mixtures having more than two components: (i) for the prediction of mixture viscosities, it does not require the selection procedure of reference fluids, thus eliminating the possible significant errors resulting from such a selection; (ii) for the correlation of mixture viscosities, it reduces the number of the binary interaction coefficients to one, no matter how many components are in a system. Obviously, this results in substantial cost and time savings.

Two equations are proposed in this study as supplements to the original GCSP to make it more efficient; viz. (i) the first equation is concerned with the prediction of the

interaction coefficient of the binary n-alkane systems from the pure component properties. (ii) the second equation provides a technique for the appropriate reference fluid selection when the original GCSP method is used for the viscosity prediction of ternary n-alkane liquid systems.

Some literature excess property models have also been subjected to testing by using the excess property data calculated from the experimental viscometric and volumetric properties obtained in this study.

DEDICATION

To my parents for their love, encouragement and support.

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CHAPTER 1

INTRODUCTION

1.1 General

Viscosity is defined as the shear force per unit area divided by the velocity gradient (Reid *et al.*, 1977). Viscosity of liquid mixtures is a very important transport property. This is because theoretically, it provides a clear insight into the behavior of liquid molecules and practically, it is required for the solution of many engineering problems involving heat transfer, mass transfer and fluid flow.

The characteristics of the liquid state involve (i) strong interactions among molecules, and (ii) disordered molecular arrangement. Because the effective treatment of liquid structure is difficult, theoretical analysis of the viscosity of liquid mixtures has lagged far behind the viscosity theories of gases which have negligible interactions amongst molecules due to large intra-molecular spaces. A reliable and generally valid theory for the quantitative prediction of liquid mixture viscosities has not been established yet (Eyring and Jhon, 1969; Reid *et al.*, 1977; Asfour *et al.*, 1991).

N-alkanes are major components of many industrial liquids, unfortunately, reliable viscometric data on n-alkane mixtures, especially mixtures of more than two components, are very scarce in the literature. Data on such systems are needed for their own value as well as for the theoretical analysis of liquid viscosities.

The semi-empirical McAllister model (McAllister, 1960) which is based on Eyring's theory of absolute reaction rates is regarded, by many investigators, as the best correlating technique available for binary and ternary non-polar liquid systems (Reid *et al.*, 1977). However, the major drawback of the model is that it contains adjustable parameters which must be determined by using experimental viscosity-composition data. Asfour *et al.* (1991) reported a new technique for predicting the values of the parameters of the model from the pure component properties for binary n-alkane mixtures, but for the ternary parameter of this model, there has been no comparable prediction technique.

The McAllister model would have too many undetermined parameters if extended to systems of four or more components because of the higher order of molecular interactions. Therefore, it may not be possible to extend the technique reported by Asfour *et al.* (1991) to cover systems containing more than three compounds. For such mixtures, the Generalized Corresponding States Principle (GCSP) is valid. The GCSP method proposed first for thermodynamic properties (Teja, 1980; Teja and Sandler, 1980) was extended to the viscosity of liquid mixtures by Teja and Rice (1981). It is based on the Corresponding States Principle and the properties of two reference fluids. This method can easily be extended to mixtures having unrestricted number of components. It is predictive or correlating depending on whether the binary interaction coefficients are set equal to unity or not. The disadvantages of this method are: (i) if it is used as a predictive technique and the number of components is more than two, different selections of reference fluids could give significantly different results for the viscosity prediction of n-alkane liquid mixtures and there is no reliable way to choose the appropriate reference fluids, (ii) if it is used as a correlation technique by taking the binary interaction coefficients as adjustable parameters for multicomponent mixtures, too many binary interaction coefficients would require many costly and time consuming experimental data to determine.

1.2 Objectives

The objectives of this study are as follows:

- To determine the viscosities of five selected ternary n-alkane liquid systems over the entire composition range at 293.15, 298.15, 308.15 and 313.15 K. Figure 1.1 illustrates the ternary systems investigated in this study. Viscosities of the corresponding eight binary subsystems are also to be measured over the same temperature ranges. All the obtained data will be employed in subjecting some literature models to critical testing. In addition, such data will be used for providing possible modifications of existing predictive models to enhance their predictive capabilities.
- To determine the densities of all systems indicated earlier over the same composition and temperature ranges to convert the measured kinematic viscosities to absolute viscosities required by some of the models to be tested.
- To extend the technique reported by Asfour *et al.* for the prediction of McAllister parameters to ternary n-alkane liquid mixtures.

- To modify the Generalized Corresponding States Principle by proposing a pseudo-binary mixture model in order to overcome its shortcomings in both prediction and correlation of multicomponent mixture viscosities.
- To test some literature excess property models by using the excess property data calculated from the gathered experimental data.

System	n-Octane	n-Decane	n-Undecane	n-Tridecane	n-Pentadecane
1	x		x	x	
2	x		x		x
3			x	x	x
4	x			x	x
5		x		x	x

Figure 1.1. Ternary n-Alkane Systems Investigated

CHAPTER 2

LITERATURE SURVEY

2.1 General

Viscosity of liquid mixtures has attracted much attention in the literature (Partington, 1951; Reid *et al.*, 1977) because of its theoretical and practical importance. Viscosity is a property which illustrates the nature of the problem encountered in the theory of transport properties (Pryde, 1966). Viscosity is also a property the knowledge of which is required in many engineering applications which involve heat transfer, mass transfer and fluid flow. It is well known that the three important dimensionless groups, the *Reynolds number*, the *Prandtl number* and the *Schmidt number*, all have viscosity as a main parameter. Unfortunately, reliable viscometric data on liquid mixtures at different temperatures are very scarce in the literature.

The investigation of viscosities of fluids started in 1877 by Arrhenius. More than 50 empirical or semi-empirical equations have appeared in the literature till 1977 (Irving, 1977). New models have continued to appear since 1977. Most of the reported equations describe the viscosity of binary liquid mixtures, however, some of them may be extended to mixtures of three or more components. Usually the extended equations require more adjustable parameters, but the accuracy of prediction is far from satisfactory. A general and reliable theory for the accurate prediction of viscosities of liquid mixtures has not been established yet (Reid *et al.*, 1977; Asfour, *et al.*, 1991).

In this chapter, the effects of pressure and temperature on liquid viscosity are discussed first. This is followed by a survey of the literature viscosity models for liquid mixtures. The last part of this chapter contains the terminology of some viscosity related properties.

2.2 The Effects of Pressure and Temperature on Liquid Viscosity

Viscosity of liquids increases if the pressure on the liquid is increased, because the molecules are pushed closer together, more work needs to be done to open up a vacant site for the molecule to jump into. But since the viscosity of liquids below the normal boiling point is not particularly affected by moderate pressures (Reid, *et al.*, 1977), the investigation of liquid viscosity is usually carried on under atmospheric pressure.

The viscosity of liquids decreases with temperature. As pointed out by Andrade (1954) "the most striking feature of liquid viscosity is the very marked way in which it decreases with rise of temperature, whereas the viscosity of gases increases with temperature". The Andrade correlation (Andrade, 1930) is given by

$$\eta = A\, e^{B/T} \tag{2.1}$$

It indicates an exponential decrease in viscosity with temperature. Although a number of variants of eq. (2.1) has been proposed since 1930, eq. (2.1) is still the most widely used correlation for showing the effect of temperature on liquid viscosity (Reid *et al.*, 1977).

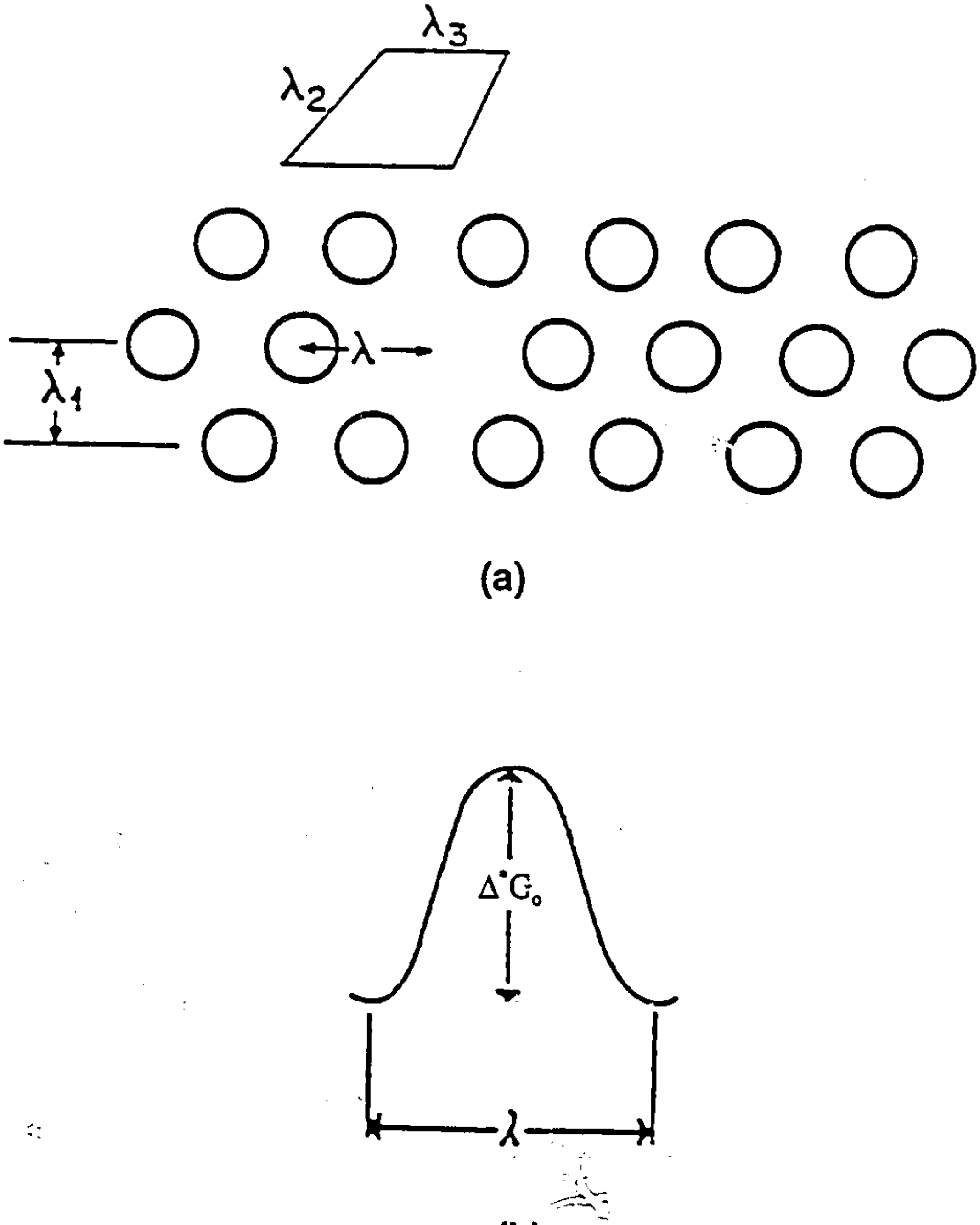
2.3 Survey of the Literature Viscosity Models for Liquid Mixtures

2.3.1 The Eyring theory of viscosity of liquid

A number of existing viscosity models is based on Eyring's absolute rate theory (Eyring, 1936, 1957). This theory is introduced as follows:

By considering viscous flow as a chemical reaction in which the elementary process is the passing of a single molecule from one equilibrium position to another over a potential barrier, Eyring combined the theory of absolute reaction rates, which was originally devised by himself (Eyring, 1935) and has been used successfully in chemical kinetics, with the hole theory and applied them to the study of liquid mixtures (Hischfelder *et al.* 1954; Eyring, 1936; Ewell and Eyring, 1937; Glasstone, 1941; Tabor, 1969; Eyring, 1957). The model for the viscous flow of a liquid is shown in Figure 2.1. The center to center distance between two layers of molecules in a liquid is λ_1, the space where a molecule is not shown is known as a hole. The average area occupied by a molecule is $\lambda_2\lambda_3$ and the center to center distance between a molecule and a hole is λ which is the distance that a molecule jumps

When no force is acting on the liquid, the jumping of the molecules to a neighbouring site is a result of thermal activation, the rate of jumping to left and right is equal so that there is no net flow. The potential energy barrier, $\Delta^{*}G_o$, which must be crossed by the jumping molecule in this case is shown in Figure 2.1(b). The frequency



(a)

(b)

Figure 2.1. The Eyring Molecular Model of Liquid Viscosity

for both forward and backward molecule jumps when no force acting is

$$r_o = \frac{kT}{h} \exp\left(-\frac{\Delta^* G_o}{kT}\right) \tag{2.2}$$

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

When a shear stress, *f*, is applied forward to the liquid, the viscous flow occurs. The force on the molecule is ($f \lambda_2 \lambda_3$). Since work is defined as force multiplied by the distance it acts through and the distance to reach the top of the barrier is ($\lambda/2$), the work done in carrying the molecule to the top of the potential barrier (on the other side of the barrier peak the molecules is assumed to give up all its energy as heat) is

$$work = f \lambda_2 \lambda_3 \frac{\lambda}{2} \tag{2.3}$$

The potential barrier is different for viscous flow from the non-flow situation. In this case, the forward rate of a molecule jumping is

$$r_{forward} = \frac{kT}{h} \left[\exp\left(-\frac{\Delta^* G_o - f \lambda_2 \lambda_3 \frac{\lambda}{2}}{kT}\right)\right] \tag{2.4}$$

and the backward rate of the molecule jumping is

$$r_{backward} = \frac{kT}{h} \left[\exp\left(-\frac{\Delta^* G_o + f \lambda_2 \lambda_3 \frac{\lambda}{2}}{kT}\right)\right] \tag{2.5}$$

Therefore, the net rate of jumping is

$$r = \frac{kT}{h}\,[\exp(-\,\frac{\Delta^{*}G_o}{kT})\,[\exp(\frac{f\lambda_2\lambda_3\lambda}{2kT})\;-\;\exp(-\frac{f\lambda_2\lambda_3\lambda}{2kT})] \tag{2.6}$$

The last term of eq. (2.6) can be simplified to give

$$r = \frac{kT}{h}\,\exp(-\,\frac{\Delta^{*}G_o}{kT})\,[2\frac{f\lambda_2\lambda_3\lambda}{2kT}] = \frac{f\lambda_2\lambda_3\lambda}{h}\,\exp(-\,\frac{\Delta^{*}G_o}{kT}) \tag{2.7}$$

Because the absolute viscosity η is defined as the shear stress divided by the velocity gradient and the velocity gradient across the two molecular layers separated by a distance λ_1 is

$$\begin{aligned} \textit{velocity gradient} &= \frac{\textit{velocity difference}}{\lambda_1} \\ &= \frac{(\textit{distance per jump}) \times (\textit{number of jumps per sec.})}{\lambda_1} \\ &= \frac{\lambda\, r}{\lambda_1} \end{aligned} \tag{2.8}$$

Then the viscosity η is

$$\eta = f\,\frac{\lambda_1}{\lambda r} \tag{2.9}$$

Inserting the value of r from equation (2.7) gives

$$\eta = \frac{h\lambda_1}{\lambda_2\lambda_3\lambda^2}\,\exp(\frac{\Delta^{*}G_o}{kT}) \tag{2.10}$$

If $\lambda_1 = \lambda$ and treating $\lambda_2\lambda_3\lambda_1$ as the molecular average volume, v, equation (2.10) becomes

$$\eta = \frac{h}{v} \exp\left(\frac{\Delta^{*}G_{o}}{kT}\right) \tag{2.11}$$

and

$$\eta = \frac{hN_{o}}{V} \exp\left(\frac{\Delta^{*}G}{RT}\right) \tag{2.12}$$

where N_o is Avogadro's number, V the molar volume of the liquid and $\Delta^{*}G$ the molar activation energy of viscous flow. The barrier energy $\Delta^{*}G_o$ may also be considered as the energy to create a necessary hole for a molecule to drop in.

If $\lambda_1 \neq \lambda$, then eq. (2.10) becomes

$$\begin{aligned} \eta &= \frac{h\lambda_{1}^{2}}{\lambda_{1}\lambda_{2}\lambda_{3}\lambda^{2}} \exp\left(\frac{\Delta^{*}G_{o}}{KT}\right) \\ &= \frac{hN_{o}}{V}\left(\frac{\lambda_{1}}{\lambda}\right)^{2} \exp\left(\frac{\Delta^{*}G}{RT}\right) \end{aligned} \tag{2.13}$$

For the kinematic viscosity ν

$$\nu = \frac{\eta}{\rho} = \eta\frac{V}{M} = \left(\frac{\lambda_{1}}{\lambda}\right)^{2} \frac{hN_{o}}{M} \exp\left(\frac{\Delta^{*}G}{RT}\right) \tag{2.14}$$

2.3.2 McAllister's model

McAllister (1960) proposed a semi-empirical model based on Eyring's absolute reaction rate theory for the viscosity of nonpolar binary liquid mixtures.

For a mixture of molecules of types 1 and 2, the two-dimensional, three-body interaction was first assumed and a number of different encounters as shown in Figure 2.2 were proposed. It is clear from Figure 2.2 that there are six types of interactions in a binary mixture, they are 1-1-1. 1-2-1, 1-1-2, 2-1-2, 1-2-2 and 2-2-2. The following assumptions are also made: (i) the probability for the interactions is dependent only on the concentration, not on the free energy of activation, (ii) the rate of each individual interaction is proportional to the energy of activation in much the same way as the reaction rate of a chemical reaction, and (iii) the free energies of activation for viscosity are additive on a number fraction or mole fraction. Therefore, for a mixture, the free energy of activation $\Delta^{*}G$ can be expressed in general as

$$\Delta^{*}G = \sum_{i=1}^{2}\sum_{j=1}^{2}\sum_{k=1}^{2} x_i x_j x_k \, \Delta^{*}G_{ijk} \tag{2.15}$$

where x is the mole fraction. Two additional assumptions are

$$\Delta^{*}G_{121} \doteq \Delta^{*}G_{112} \equiv \Delta^{*}G_{12} \tag{2.16}$$

$$\Delta^{*}G_{212} \doteq \Delta^{*}G_{122} \equiv \Delta^{*}G_{21} \tag{2.17}$$

Substituting eqs. (2.16) and (2.17) into (2.15) yields

$$\Delta^{*}G = x_1^3 \Delta^{*}G_1 + 3x_1^2 x_2 \Delta^{*}G_{12} + 3x_1 x_2^2 \Delta^{*}G_{21} + x_2^3 \Delta^{*}G_2 \tag{2.18}$$

For each type of energy of activation considered here a corresponding kinematic viscosity may be assigned.

For the mixture

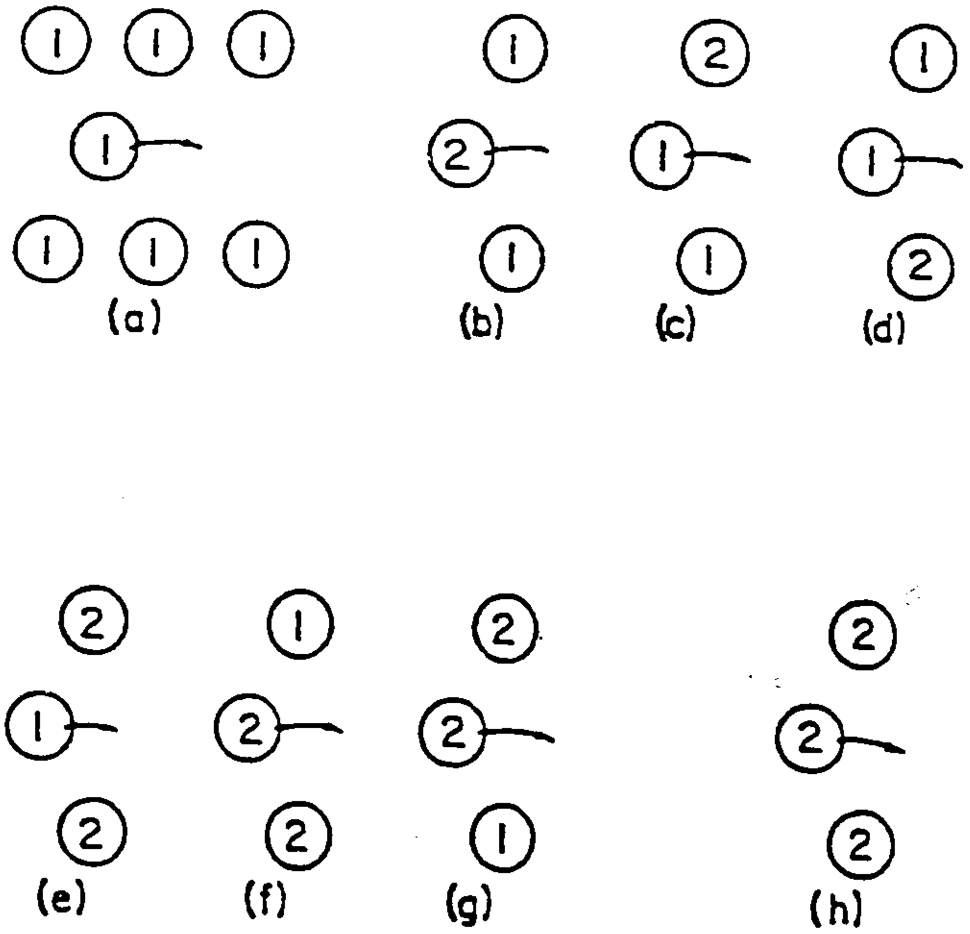


Figure 2.2. Types of Viscosity Interactions in a Binary Mixture, The McAllister Three-body Model (McAllister, 1960)

$$\nu = \frac{hN_o}{M} e^{\Delta^{*}G/RT} \tag{2.19}$$

where

$$M = \sum_i x_i M_i \tag{2.20}$$

For the pure component i

$$\nu_i = \frac{hN_o}{M_i} e^{\Delta^{*}G_i/RT} \tag{2.21}$$

For interactions

$$\nu_{ij} = \frac{hN_o}{M_{ij}} e^{\Delta^{*}G_{ij}/RT} \tag{2.22}$$

where

$$M_{ij} = (2M_i + M_j)/3 \tag{2.23}$$

Substituting eq. (2.18) into (2.19) results in

$$\nu = \frac{hN_o}{M} e^{(x_1^3\Delta^{*}G_1 + 3x_1^2x_2\Delta^{*}G_{12} + 3x_1x_2^2\Delta^{*}G_{21} + x_2^3\Delta^{*}G_2)/RT} \tag{2.24}$$

By taking logarithms of eqs. (2.19), (2.21), (2.22) and (2.24), combining to eliminate the free energies of activation, rearranging and condensing, the well-known McAllister's three-body model is as follows:

$$\begin{aligned}\ln \nu &= x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 \\ &- \ln(x_1 + x_2 \frac{M_2}{M_1}) + 3x_1^2 x_2 \ln(\frac{2 + M_2/M_1}{3}) \\ &+ 3x_1 x_2^2 \ln(\frac{1 + 2M_2/M_1}{3}) + x_2^3 \ln(\frac{M_2}{M_1})\end{aligned} \tag{2.25}$$

Equation (2.25) contains two adjustable parameters which have to be determined from mixture viscosity data by the method of least squares. The three-body model works well when the two types of molecules in the mixture are different in size (radius) by less than a factor of 1.5.

McAllister pointed out that when the size of one component molecule is much larger than that of the other component molecule, the three-body interactions considered above may not be realistic, four-body interaction has to be assumed. By techniques analogous to the method given earlier, McAllister derived his four-body model as follows:

$$\begin{aligned}\ln \nu &= x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} \\ &+ 4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln(x_1 + x_2 \frac{M_2}{M_1}) \\ &+ 4x_1^3 x_2 \ln(\frac{3 + M_2/M_1}{4}) + 6x_1^2 x_2^2 \ln(\frac{1+M_2/M_1}{2}) \\ &+ 4x_1 x_2^3 \ln(\frac{1+3M_2/M_1}{4})\end{aligned} \tag{2.26}$$

McAllister (1960) tested his three-body model, eq. (2.25), on various binary systems.

A methanol-toluene system was tested using experimental data over a temperature range 20.00°C - 60.11°C, except for three data points at 25°C and one at 37.8°C, all the calculated viscosity values agreed with the experimental data with a maximum difference of ± 0.5%. When the benzene-toluene system was tested by eq. (2.25) at 25°C, the maximum deviation is 0.06% and the average difference is 0.02%. Testing the cyclohexane-heptane system at 37.8°C resulted in an average percentage difference of ± 0.2% and a maximum difference of 0.5%. Testing the polar system, acetone-water by eq. (2.25) resulted in an average deviation of 6.4% and a maximum deviation of 15.8%. The conclusion is that even though the McAllister's three-body model was highly oversimplified, the accuracy obtained for non-polar binary liquid mixtures is sufficiently good (McAllister, 1960). The weakness is that it contains adjustable parameters which have to be determined by using mixture viscosity data. Collection of such data is costly and time consuming.

Recently, Asfour *et al.* (1991) proposed a new technique which can predict the binary parameters of the McAllister's three-body and four-body model very well for n-alkane liquid mixtures from the properties of the pure components of the mixture.

The equations suggested by Asfour *et al.* for predicting the McAllister three-body parameters for n-alkane binary systems are as follows:

$$\frac{\nu_{12}}{(\nu_1^2 \nu_2)^{1/3}} = 1 + 0.044 \frac{(N_2 - N_1)^2}{(N_1 N_2)^{1/3}} \tag{2.27}$$

$$\nu_{21} = \nu_{12} \, (\nu_1/\nu_2)^{1/3} \tag{2.28}$$

where N_1 and N_2 are the carbon atoms/per molecule of components 1 and 2, respectively. And their equations for predicting the McAllister four-body parameters for n-alkane binary mixtures are

$$\frac{\nu_{1122}}{(\nu_1^2 \nu_2^2)^{1/4}} = 1 + 0.03 \frac{(N_2 - N_1)^2}{(N_1 N_2)^{1/4}} \tag{2.29}$$

$$\nu_{1112} = \nu_{1122} \, (\nu_1/\nu_2)^{1/4} \quad \textit{and} \quad \nu_{2221} = \nu_{1122} \, (\nu_2/\nu_1)^{1/4} \tag{2.30}$$

The predicted values of the parameters are in a very close agreement with those calculated from experimental data.

The McAllister model was extended to ternary mixtures by Chandramouli and Laddha (1963) and by Kalidas and Laddha (1964). In the study of the viscosity of a ternary mixture, molecules of types 1, 2 and 3 and their different interactions are considered. These are shown in Figure 2.3. Still, the two-dimensional, three-body interactions were assumed here, and arbitrarily, the size (radius) of the three types of molecules did not differ by a factor of 1.5 with respect to each other. The extension of the McAllister model is

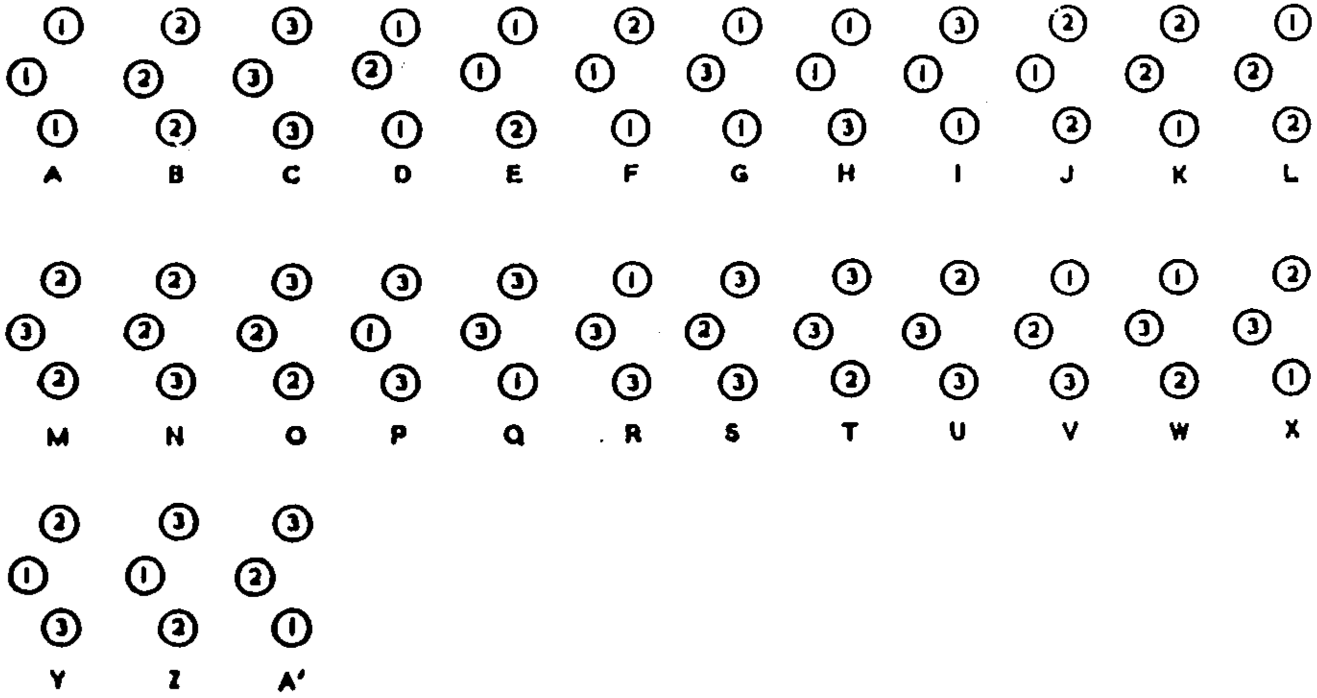


Figure 2.3. Interaction Model for Ternary System Consisting Molecules of Type 1, 2 and 3 (Kalidas *et al.*, 1964)

$$\begin{aligned}\ln\nu = {} & x_1^3\ln\nu_1 + x_2^3\ln\nu_2 + x_3^3\ln\nu_3 + 3x_1^2x_2\ln\nu_{12} + 3x_1^2x_3\ln\nu_{13} \\ & + 3x_2^2x_1\ln\nu_{21} + 3x_2^2x_3\ln\nu_{23} + 3x_3^2x_1\ln\nu_{31} + 3x_3^2x_2\ln\nu_{32} \\ & + 6x_1x_2x_3\ln\nu_{123} - \ln(x_1M_1 + x_2M_2 + x_3M_3) + x_1^3\ln M_1 \\ & + x_2^3\ln M_2 + x_3^3\ln M_3 + 3x_1^2x_2\ln(2M_1+M_2)/3 \\ & + 3x_1^2x_3\ln(2M_1+M_3)/3 + 3x_2^2x_1\ln(2M_2 + M_1)/3 \\ & + 3x_2^2x_3\ln(2M_2 + M_3)/3 + 3x_3^2x_1\ln(2M_3 + M_1)/3 \\ & + 3x_3^2x_2\ln(2M_3 + M_2)/3 + 6x_1x_2x_3\ln(M_1 + M_2 + M_3)/3\end{aligned} \tag{2.31}$$

where ν_{123} involves the interactions of three different molecules each of type 1, 2 and 3. The relationship between ν_{123} and its corresponding activation energy is

$$\nu_{123} = \left(\frac{hN_o}{M_{123}}\right) e^{\Delta^* G_{123}/RT} \tag{2.32}$$

where

$$M_{123} = (M_1 + M_2 + M_3)/3 \tag{2.33}$$

In eq. (2.31), there are six binary parameters: ν_{12}, ν_{21}, ν_{13}, ν_{31}, ν_{23} and ν_{32}, and one ternary parameter, ν_{123}. The binary constants can be calculated by measurements of viscosities of binary mixtures or can be predicted by the technique reported by Asfour *et al.* (1991). Therefore, eq. (2.31) contains only one undetermined parameter, ν_{123}. Application of eq.(2.31) to acetone-methanol-ethylene glycol mixture at 30°C gave an average deviation of ± 0.95% and a maximum deviation of ± 1.81% (Kalidas and Laddha, 1964), the accuracy supported the proposed model which is the extension of McAllister's three-body model to ternary mixtures.

It can be noted that although the extended McAllister's model, eq. (2.31), gives reasonably accurate results, yet its main drawback is that the value of the parameter, ν_{123}, has to be determined by ternary viscosity-composition data. Moreover, the McAllister model would be very complicated and would possess more undetermined adjustable parameters if it is extended to mixtures having more than three components. This is because of the higher order interactions among the components. Therefore, the McAllister model is only practical for binary and ternary mixtures.

2.3.3 Heric's model

Heric's model is also based on Eyring's absolute rate theory for viscosities.

Eyring's viscosity equation, eq. (2.13), can be written as

$$\frac{\Delta^{*}G}{RT} = \ln\frac{\eta V}{hN_{o}}\left(\frac{\lambda}{\alpha}\right)^{2} \tag{2.34}$$

where α is equivalent to λ_1.

If for the binary mixture it is assumed that (Katti and Chaudhri, 1964)

$$\frac{\Delta^{*}G}{RT} = x_1\frac{\Delta^{*}G_1}{RT} + x_2\frac{\Delta^{*}G_2}{RT} \tag{2.35}$$

and

$$\ln\frac{\lambda}{\alpha} = x_1\ln\frac{\lambda_1}{\alpha_1} + x_2\ln\frac{\lambda_2}{\alpha_2} \tag{2.36}$$

then

$$\ln\eta V = x_1\ln\eta_1 V_1 + x_2\ln\eta_2 V_2 \tag{2.37}$$

but for regular solutions, $\Delta^{*}G$ would not be a linear function of $\Delta^{*}G_1$ and $\Delta^{*}G_2$ (Guggenheim, 1952; Rowlinson, 1959), therefore, Katti and Chaudhri's (1964) application of the Eyring viscosity theory based upon the regular solution two-body model is given by

$$\ln\eta V = x_1\ln\eta_1 V_1 + x_2\ln\eta_2 V_2 + x_1 x_2\frac{W_{visc}}{RT} \tag{2.38}$$

where W_{visc} was defined as the interaction energy for the activation of viscous flow.

The Katti and Chaudhri's approach was then modified by replacing $x_1x_2W_{visc}$ with a power series in concentration and extended to multicomponent systems by Heric (1966) as well as Heric and Brewer (1967) into the following form

$$\ln v = \sum_{i=1}^{n} x_i \ln v_i + \sum_{i=1}^{n} x_i \ln M_i - \ln\sum_{i=1}^{n} x_i M_i + \delta_{i...n} \tag{2.39}$$

$$\delta_{i...n} = \sum_{\substack{i=1\\ i<j}}^{n} x_i x_j\left[A_{ij} + B_{ij}(x_i - x_j) + C_{ij}(x_i - x_j)^2 + \cdots\right] \tag{2.40}$$

or alternatively,

$$\delta_{1\ldots n} = \sum_{\substack{i=1\\i<j}}^{n} x_i x_j (A'_{ij} + B'_{ij} x_i + c'_{ij} x_i^2 + \cdots) \tag{2.41}$$

eqs. (2.40) and (2.41) are based upon an assumption that only binary interaction occurs, it leads to a relatively large error when applied to ternary mixtures. Thus, for ternary systems, eqs. (2.40) and (2.41) were expanded by Heric and Brewer (1969) to the following forms:

$$\begin{aligned}\delta_{123} &= \sum_{\substack{i=1\\i<j}}^{3} x_i x_j [A_{ij} + B_{ij}(x_i - x_j) + C_{ij}(x_i - x_j)^2 + \cdots] \\ &\quad + x_1 x_2 x_3 (A_{123} + B_{123} x_1 + C_{123} x_2)\end{aligned} \tag{2.42}$$

and

$$\begin{aligned}\delta_{123} &= \sum_{\substack{i=1\\i<j}}^{3} x_i x_j (A'_{ij} + B'_{ij} x_i + C'_{ij} x_i^2 + \cdots) \\ &\quad + x_1 x_2 x_3 (A'_{123} + B'_{123} x_1 + C'_{123} x_2)\end{aligned} \tag{2.43}$$

where eqs. (2.40) and (2.42) are called symmetric because the series reduces to a single term in A_{ij} at $x_i = x_j = 0.5$ whereas eqs.(2.41) and (2.43) are called asymmetric.

Testing eqs. (2.39) and (2.40) with data from 14 binary non-electrolyte systems at 25°C, a standard error of 0.07% – 0.41% in kinematic viscosity was found (Heric and Brewer, 1967). For ternary mixtures, the asymmetric Heric model gave better results than the symmetric one, its standard error of kinematic viscosity estimation is 0.28% – 0.50% whereas that of the symmetric model is 0.26% – 2.4% when both approaches applied to 11 ternary non-electrolyte systems at 25°C (Heric and Brewer, 1969).

In comparing Heric's model with McAllister's model, the former is not superior to the latter in accuracy. The major shortcoming of Heric's model is that it requires a varied number of adjustable parameters and usually the number has to be large enough to achieve the required accuracy.

2.3.4 Rowley's Model

Wei and Rowley (1984a, 1984b, 1985) proposed a method for the prediction of non-aqueous liquid mixture viscosities based on a local composition model and Eyring's viscosity theory. They substituted the following relationship between the activation energy of viscous flow, $\Delta^{*}G$, and the excess free energy of mixing, G^E,

$$\Delta^{*}G = \Delta^{*}G^{i} - \sigma G^{E} \tag{2.44}$$

into Eyring's viscosity theory, eq. (2.12), and regrouping G^E in terms of S^E and H^E to yield

$$\eta V = (\eta V)_{id} \exp\left(\frac{\sigma S^{E}}{R}\right) \exp\left(\frac{-\sigma H^{E}}{RT}\right) \tag{2.45}$$

where $\Delta^{*}G^{i}$ represents the activation energy of an ideal mixture, S^E and H^E are excess entropy and excess enthalpy of mixing, respectively, and σ is a proportionality factor. The excess entropy term was grouped with $(\eta V)_{id}$ in eq.(2.45) to yield a local shear viscosity and this group was labelled $(\eta V)_{loc}$. Thus

$$\eta V = (\eta V)_{loc} \exp\left(\frac{-\sigma H^E}{RT}\right) \tag{2.46}$$

Defining a property, ξ, as

$$\xi \equiv \ln(\eta V) \tag{2.47}$$

then eq. (2.46) has the form as

$$\xi = \xi_{loc} - \frac{\sigma H^E}{RT} \tag{2.48}$$

where ξ_{loc} is the contribution to ξ due to nonrandom mixing on the local level.

In order to use eq. (2.48) to predict mixture viscosity, the nonrandom two-liquid theory (NRTL) which was developed by Renon and Prausnitz (1968) and extended to thermal conductivities by Rowley (1982) has been used to compute ξ_{loc}.

In the NRTL model (Renon and Prausnitz, 1968), the local mole fractions are assumed to be related to the overall mole fractions as

$$\frac{x_{ij}}{x_{jj}} = \frac{x_i}{x_j} G_{ij} \tag{2.49}$$

$$G_{ij} = \exp\left(\frac{-\alpha A_{ij}}{RT}\right) \tag{2.50}$$

where x_{ij} represents the mole fraction of component i around a central molecule of type j, and α and A_{ij} are NRTL parameters obtainable from thermodynamic equilibrium data. It is noticeable that $A_{ij} \neq A_{ji}$ and $A_{ij} = 0$ whenever i = j.

The local volume fraction is defined by (Wei and Rowley, 1984a, 1984b, 1985)

$$\phi_{ij} = \frac{V_i x_{ij}}{\sum_{l=1}^{n} V_l x_{lj}} \qquad (2.51)$$

and the overall volume fraction is known as

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^{n} x_i V_i} \qquad (2.52)$$

Substituting eqs. (2.49) and (2.52) into (2.51) yields

$$\phi_{ij} = \frac{\phi_i G_{ij}}{\sum_{l=l}^{n} \phi_l G_{lj}} \qquad (2.53)$$

According to the two-liquid theory (Renon and Prausnitz, 1968; Rowley, 1982), a binary mixture can be considered as two hypothetical pure fluids shown in Figure 2.4. If $\xi^{(j)}$ represents the viscous effect of the hypothetical pure fluid of type j, then ξ_{loc} is assumed to be a volume fraction average of $\xi^{(j)}$ of the n hypothetical pure fluids which represent the n-component mixture,

$$\xi_{loc} = \sum_{j=1}^{n} \phi_j \xi^{(j)} \qquad (2.54)$$

The volume fractions were chosen to work here by Wei and Rowley because it produced a form for the binary interaction term consistent with that found for thermal conductivity (Rowley, 1982) and because it gives slightly better results than those given by mole

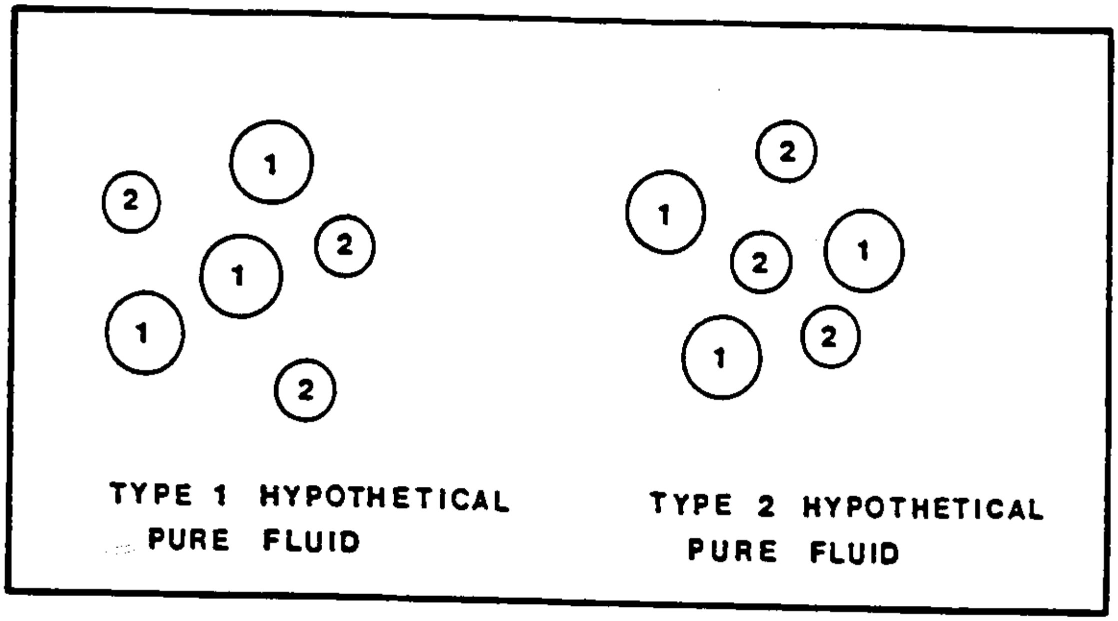


Figure 2.4. The NRTL Theory Model for a Binary Mixture (Rowley, 1982)

fractions (1984a, 1984b, 1985). $\xi^{(i)}$ in the above equation is assumed to be related to the local volume fractions by

$$\xi^{(j)} = \sum_{i=1}^{n} \phi_{ij} \xi_{ij} \tag{2.55}$$

Combination of eqs. (2.54) and (2.55) gives

$$\xi_{loc} = \sum_{j=1}^{n} \phi_j \sum_{i=1}^{n} \phi_{ij} \xi_{ij} \tag{2.56}$$

where ξ_{ij} equals ξ_{ji}. Substituting (2.53) into (2.56) yields

$$\xi_{loc} = \frac{\sum_{j=1}^{n} \phi_j \sum_{i=1}^{n} \phi_i G_{ij} \xi_{ij}}{\sum_{l=1}^{n} \phi_l G_{lj}} \tag{2.57}$$

In order to calculate the viscous interaction terms ξ_{ij}, setting

$$\xi_{loc} = \xi_{21} = \xi_{12} \tag{2.58}$$

at a particular composition defined by

$$\frac{x_{21}^{*} \exp(\xi_{21})}{x_{11}^{*} \exp(\xi_{11})} = \frac{x_{12}^{*} \exp(\xi_{12})}{x_{22}^{*} \exp(\xi_{22})} \tag{2.59}$$

Eq. (2.59) means that at the * composition, the ratio of viscous effects due to component 2 and those due to component 1 around a central molecule of component 1 is equivalent to the ratio of viscous effects due to component 1 and those due to component 2 around a central molecule of component 2.

Substituting eq. (2.58) into (2.56) gives a direct solution for ξ_{21}

$$\xi_{21} = \frac{\phi_1^* \phi_{11}^* \xi_1^o + \phi_2^* \phi_{22}^* \xi_2^o}{\phi_1^* \phi_{11}^* + \phi_2^* \phi_{22}^*} \tag{2.60}$$

where the pure component j value for ξ is represented with ξ_j^o, i.e. $\xi_{jj} = \xi_j^o$. In eq. (2.60), ϕ_{ii}^* can be calculated from eq. (2.53) and ϕ_i^* can be calculated by the equation derived below.

From the definition of volume fraction, it is obvious that

$$\frac{\phi_1}{\phi_2} = \frac{V_1}{V_2}\frac{x_1}{x_2} \tag{2.61}$$

and eq. (2.49) gives

$$\frac{x_1}{x_2} = \left(\frac{\dfrac{x_{12}}{x_{22}}}{\dfrac{x_{21}}{x_{11}}}\frac{G_{21}}{G_{12}}\right)^{\frac{1}{2}} \tag{2.62}$$

Rearranging eq. (2.59) and replacing ξ_{jj} with ξ_j^o yield

$$\frac{\dfrac{x_{12}^*}{x_{22}^*}}{\dfrac{x_{21}^*}{x_{11}^*}} = \exp\left(\xi_2^o - \xi_1^o\right) \tag{2.63}$$

Substituting eqs. (2.62) and (2.63) into (2.61) at the * composition yields

$$\frac{\phi_1^*}{\phi_2^*} = \frac{V_1\sqrt{G_{21}}}{V_2\sqrt{G_{12}}}\exp\left(\frac{\xi_2^o - \xi_1^o}{2}\right) \tag{2.64}$$

subject to the constraint that

$$\sum_{i=1}^{n} \phi_i = 1 \tag{2.65}$$

eq. (2.64) becomes

$$\phi_1^* = \frac{V_1\sqrt{G_{21}}\exp(\frac{\xi_2^o}{2})}{V_1\sqrt{G_{21}}\exp(\frac{\xi_2^o}{2}) + V_2\sqrt{G_{12}}\exp(\frac{\xi_1^o}{2})} \tag{2.66}$$

Eq. (2.66) is used to calculate ϕ_i^*.

To complete Rowley's viscosity model, eqs. (2.48) and (2.57) can be combined and rearranged to yield

$$\xi = \sum_{i=1}^{n} \phi_i \xi_i^o + \sum_{i=1}^{n} \phi_i \frac{\sum_{j=1}^{n} \phi_j G_{ji}(\xi_{ji} - \xi_i^o)}{\sum_{l=1}^{n} \phi_l G_{li}} - \frac{\sigma H^E}{RT} \tag{2.67}$$

Eq. (2.67) can employed as follows: first ϕ_i^* is calculated from eq. (2.66) and ϕ_{ii}^* from eq. (2.53), then the binary interactions ξ_{ij} are calculated from eq. (2.60), G_{ij} and G_{ji} from eq. (2.50). The excess enthalpies of mixing can be obtained exclusively from Christensen *et al.* (1982) and the proportionality factor, σ, has been designated a value of 0.25 for non-aqueous systems by Wei and Rowley (1985). Using all the information given earlier, ξ can be computed by eq.(2.67) and the mixture viscosity is calculated as

$$\eta = \frac{\exp(\xi)}{V} \tag{2.68}$$

Rowley's viscosity model based on the NRTL theory has been tested by using data on 47 binary and seven ternary systems, a reasonable degree of accuracy was achieved in predicting the viscosities of mixtures (Wei and Rowley, 1985). This model can be used for any number of components without adjustable parameters, however, it requires pure component viscosities and binary equilibrium thermodynamic data as well as the data of excess enthalpy of mixing. Unfortunately, the binary equilibrium data and the H^E data are not always available. These disadvantages, of course, limit the use of the Rowley's model.

2.3.5 The Corresponding States Principle

The Corresponding States Principle, in the form originally stated by van der Waals, is based on the reduction of the variables using the critical constants. It is one of the most useful methods for predicting thermodynamic properties of fluids. It makes use of measured properties of one substance to predict the properties of other substances under conditions where no data exist and no satisfactory theoretical treatments may be applied (Hirschfelder, *et al.*, 1954). The Corresponding States Principle for transport properties of pure fluids was established by Helfand and Rice (1960), Preston *et al.* (1967), Tham and Gubbins (1969, 1970).

The basic assumption of the theory for viscosities is that the viscosity of mixtures or pure fluids can be equated to that of a hypothetical pure fluid, the viscosity of this hypothetical pure fluid is then evaluated via corresponding states with respect to a given reference fluid (Ely and Hanley,1981).

In the special case of corresponding states between two pure fluids α and o, the pure fluid α is defined to be in corresponding states with a reference fluid o if at the same reduced temperature T_R (=T/T_c) and volume V_R (=V/V_c) the reduced property X (where X may be the compressibility, or reduced transport properties: viscosity, diffusivity and thermal conductivity) is given by

$$X^{\alpha}(T_R, V_R) = X^{o}(T_R, V_R) \tag{2.69}$$

or alternatively (Teja, *et al.*, 1985)

$$X^{\alpha}(T_R, P_R) = X^{o}(T_R, P_R) \tag{2.70}$$

where P_R (= P/P_c) is the reduced pressure. It should be pointed out that correspondence defined in terms of the same reduced temperature and pressure is not necessarily equivalent to correspondence defined in terms of reduced temperature and volume, but they become identical when the critical compressibilities of the fluid of interest and the reference fluid are equal (Teja, *et al.*, 1985).

In case X represents reduced viscosity

$$(\eta\epsilon)^{\alpha} = (\eta\epsilon)^{o}(T_R, V_R) \tag{2.71}$$

where η is the viscosity at the temperature of interest and the reciprocal of ε is the viscosity at T_c (Reid *et al.*, 1977). ε is given by dimensional analysis as

$$\epsilon = M^{-1/2} V_c^{2/3} T_c^{-1/2} \tag{2.72}$$

or in the case of corresponding in terms of T_c and P_c

$$\epsilon = M^{-1/2} P_c^{-2/3} T_c^{1/6} \tag{2.73}$$

$\eta\varepsilon$ above hence represents the reduced viscosity.

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

$$\frac{T^o}{T^\alpha} = \frac{T_c^o}{T_c^\alpha} \tag{2.74}$$

$$\frac{\rho^o}{\rho^\alpha} = \frac{\rho_c^o}{\rho_c^\alpha} \tag{2.75}$$

For fluid mixtures, the characterization parameters of the two-parameter corresponding states principle, T_c and V_c, must be replaced by appropriate pseudocritical parameters T_{cm} and V_{cm} which are dependent on composition. Therefore, the "van der Waals one-fluid" mixing rules (Ely and Hanly, 1981) appear as

$$V_{cm} = \sum_i \sum_j x_i x_j V_{cij} \tag{2.76}$$

$$T_{cm} V_{cm} = \sum_i \sum_j x_i x_j T_{cij} V_{cij} \tag{2.77}$$

There are various ways to define the cross coefficients T_{cij} and V_{cij}. The most common approach is to introduce binary interaction coefficients ξ_{ij} and η_{ij} which characterize the non-ideality of interaction between the binary pair. The binary interaction coefficients are introduced as follows (Wong *et al.*, 1984):

$$V_{cij} = \eta_{ij}[(V_{ci}^{1/3} + V_{cj}^{1/3})/2]^3 \tag{2.78}$$

and

$$T_{cij} = \xi_{ij}(T_{ci}T_{cj})^{1/2} \tag{2.79}$$

Equations (2.76) to (2.79) constitute the van der Waals one-fluid model for use with the two-parameter corresponding-states principle.

The values of the binary interaction parameters were normally set to unity in many cases for the prediction of viscosity (Wong *et al.*, 1984).

Equations (2.69), (2.70), (2.71), (2.74) and (2.75) are valid only for simple spherical molecules. The range of applicability of corresponding states can be broadened to non-spherical fluids in the two approaches described below (Teja and Thurner, 1986).

2.3.6 The extended corresponding states — The shape factor approach

In this approach, shape factors were incorporated into the calculation of transport properties by Mo and Gubbins (1974) and Ely and Hanley (1981). The two-parameter corresponding states formulism is maintained, except that eqs. (2.74) and (2.75) become

$$\frac{T^o}{T^\alpha} = \frac{T_c^o}{T_c^\alpha \theta_{\alpha,o}(T_R, V_R, \omega^\alpha)} \tag{2.80}$$

$$\frac{\rho^o}{\rho^\alpha} = \frac{\rho_c^o}{\rho_c^\alpha} \phi_{\alpha,o}(T_R, V_R, \omega^\alpha) \tag{2.81}$$

where θ and ϕ are the so-called shape factors (Leach *et al.*, 1968), which are functions of Pitzer's acentric factor, ω, of the reduced temperature and of the reduced volume.

The shape factors can be determined exactly for any pure fluid with respect to a reference fluid by simultaneous solution of the conformal-solution equations (Rowlinson and Watson, 1969):

$$Z^\alpha(T^\alpha, \rho^\alpha) = Z^o(T^o, \rho^o) \tag{2.82}$$

$$A^\alpha(T^\alpha, \rho^\alpha) = A^o(T^o, \rho^o) \tag{2.83}$$

where Z is the compressibility factor and A is the molar reduced residual Helmholtz free energy relative to the ideal gas at the same temperature and density. Leach *et al* (1968) have solved eqs. (2.82) and (2.83) for the pure normal paraffins C_1 - C_{15} with methane as reference fluid. Their results are generalized as

$$\theta_{\alpha,o}(T_R,\ V_R,\ \omega^\alpha) = 1 + (\omega^\alpha - \omega^o)\ F(T_R,\ V_R) \tag{2.84}$$

$$\phi_{\alpha,o}(T_R,\ V_R,\ \omega^\alpha) = [1 + (\omega^\alpha - \omega^o)\ G(T_R,\ V_R)]\ Z_c^o/Z_c^\alpha \tag{2.85}$$

where

$$F(T_R,\ V_R) = a_1 + b_1 lnT^+ + (c_1 + d_1/T^+)\ (V^+ - 0.5) \tag{2.86}$$

$$G(T_R,\ V_R) = a_2(V^+ + b_2) + c_2(V^+ + d_2) lnT^+ \tag{2.87}$$

and

$$T^+ = \min\{2,\ \max(T_R,\ 0.5)\} \tag{2.88}$$

$$V^+ = \min\{2,\ \max(V_R,\ 0.5)\} \tag{2.89}$$

The shape factors differ with the temperature and volume and their function is to define the state of the reference fluid that corresponds to the state of the fluid of interest.

When the shape factor approach applied to mixtures, the extension of van der Waals one-fluid model to a three-parameter corresponding states principles is used to calculate the pseudocriticals. This extension requires an additional equation to determine the mixture acentric factor. Lee and Kesler (1975), Joffe (1971), and Plocker *et al.* (1978) used a simple linear combination rule

$$\omega_m = \sum_i x_i \omega_i \tag{2.90}$$

Plocker *et al.*, however, have noted that the three-parameter model is successful only for mixtures containing small nonpolar molecules (1978).

The shape factor approach is based on the properties of one reference fluid, it requires accurate PVT and viscosity data of methane (the reference fluid), the critical properties (temperature, density or volume and pressure), acentric factor and molecular weight of each component of the mixture of interest. No viscosity data of the pure component and the mixture are required. The method is predictive, but if ξ_{ij} and η_{ij} are used as adjustable parameters, it is a correlating method. In principle, the number of mixture components is unrestricted. Ely and Hanley (1981) concluded that with this method the viscosity of a range of pure fluids: C_1 - C_{20}, aromatics and others, is predicted to within an absolute percent deviation of about 8% and a similar range of mixtures shows that the viscosity is represented to within about 7%.

A packaged program (TRAPP) for this approach developed by Ely and Hanley has been used extensively.

Since only one fluid, methane, can serve as reference fluid, the accuracy of this method has been limited.

2.3.7 The extended corresponding states — The Pitzer approach

In this approach, the classical two-parameter corresponding states principle was extended to the three-parameter generalized corresponding states principle (GCSP).

The two-parameter classical corresponding states equation

$$Z^{\alpha}(T_R, P_R) = Z^{o}(T_R, P_R) \tag{2.91}$$

was first written for non-spherical molecule thermodynamic properties by Pitzer *et al.* (1955) as a Taylor series expansion in the acentric factor

$$Z = Z^{o} + \omega Z^{1} \tag{2.92}$$

where the first term is the compressibility of a spherical reference fluid and the second term is a deviation function. Letsou and Stiel (1973) later extended this approach to viscosities of liquids by rewriting eq. (2.92) in the form

$$\ln(\eta\epsilon) = \ln(\eta\epsilon)^{o} + \omega\ln(\eta\epsilon)^{1} \tag{2.93}$$

For the purpose of convenient calculation, Lee and Kesler (1975) provided an analytical framework for the three-parameter corresponding states principle by writing eq. (2.92) as

$$Z = Z^{o} + \frac{\omega}{\omega^{r}}(Z^{r} - Z^{o}) \tag{2.94}$$

In eq. (2.94), Pitzer's original proposal of a Taylor series expansion of a thermodynamic property about that of a simple spherical reference fluid was still retained.

Teja *et al.* (Teja, 1980, Teja and Sandler,1980) proposed the following Generalized Corresponding States Principle (GCSP) for thermodynamic properties which no longer retains the simple spherical fluid as one of the reference fluids:

$$Z = Z^{r1} + \frac{\omega - \omega^{r1}}{\omega^{r2} - \omega^{r1}}(Z^{r2} - Z^{r1}) \tag{2.95}$$

where r1 and r2 represent two non-spherical fluids which are chosen so that they are similar to the pure component of interest or, in the case of mixtures, to the key components of interest. It is obvious that when $\omega^{r1} = 0$, i.e. one of the reference fluids is spherical, eq. (2.95) reduces to eq. (2.94). Therefore, the Lee-Kesler method is a special case of the GCSP.

In an analogous manner, Teja and Rice (1981) extended the GCSP for viscosity as follows

$$\ln(\eta\epsilon) = \ln(\eta\epsilon)^{r1} + \frac{\omega - \omega^{r1}}{\omega^{r2} - \omega^{r1}}[\ln(\eta\epsilon)^{r2} - \ln(\eta\epsilon)^{r1}] \tag{2.96}$$

where r1 and r2 again refer to two (non-spherical) reference fluids.

When extending eq. (2.96) to mixtures, mixing rules are needed. Teja and Rice (1981) first used the extended van der Waals one-fluid model to calculate the pseudocritical properties T_{cm} and V_{cm}. Later Teja and Thurner (1986) used the correspondence defined in terms of reduced temperature and pressure instead of reduced temperature and volume. One of the major inconveniences of using the van der Waals one-fluid model is that it gives mixing rules for T_{cm}, V_{cm} and ω_m, whereas the Corresponding States Theory is formulated in terms of T_{cm}, P_{cm} and ω_m. Therefore, Teja and Thurner used analogues to the van der Waals mixing rules proposed by Wong *et al.* (1984). They have the following forms

$$(T_{cm}^2/P_{cm}) = \sum_i \sum_j x_i x_j (T_{cij}^2/P_{cij}) \qquad (2.97)$$

$$(T_{cm}/P_{cm}) = \sum_i \sum_j x_i x_j (T_{cij}/P_{cij}) \qquad (2.98)$$

$$\omega_m (T_{cm}/P_{cm})^{2/3} = \sum_i \sum_j x_i x_j (T_{cij}/P_{cij})^{2/3} \omega_{ij} \qquad (2.99)$$

with

$$T_{cij} = \xi_{ij} (T_{cii} T_{cjj})^{1/2} \qquad (2.100)$$

$$(T_{cij}/P_{cij})^{1/3} = \frac{1}{2} [(T_{cii}/P_{cii})^{1/3} + (T_{cjj}/P_{cjj})^{1/3}] \qquad (2.101)$$

$$\omega_{ij} = \frac{1}{2} (\omega_{ii} + \omega_{jj}) \qquad (2.102)$$

and a simple mixing rule for mass is

$$M_m = \sum_i x_i M_i \qquad (2.103)$$

Eqs. (2.97) and (2.98) are equivalent to the van der Waals mixing rules if V_c is considered proportional to $Z_c T_c / P_c$ and Z_c is a quantity that only changes slightly from one species to another so that V_c is replaceable by T_c/P_c.

The mixing rules given by eqs. (2.97) and (2.98) have the advantage that they do not contain the critical volume, which is subject to greater experimental errors than critical temperature and pressure.

The GCSP method requires only the viscosities, the critical properties, the acentric factors of the two reference fluids (but no PVT data), and no mixture properties are required.

In eq. (2.100), ξ_{ij} is the binary interaction coefficient which can be calculated from experimental data, usually it is set equal to unity. In the case where ξ_{ij} is set to unity, the GCSP method is predictive. The flexibility of the method is that if the data of the key components of the mixture are not available, other similar fluids can be used as the reference fluids. It even can predict the viscosity of some undefined liquid mixtures very well (Teja *et al.*, 1985). In the case that a binary interaction coefficient is included in the calculations, the method becomes one of correlation rather than prediction.

Teja and Thurner (1986) compared the viscosity prediction of the two corresponding states methods for a number of binary mixtures. No binary interaction coefficients were used in either method. Their results are described below.

For a mixture of methane + n-butane over a temperature range 278 - 511 K and a pressure range 80 - 350 bar, the values of the average absolute deviation (AAD) for the two methods are shown in Table 2.1. From Table 2.1, in the temperature range 278 - 411 K and the pressure range 80 - 350 bar, the AAD between the experimental and the predicted viscosities obtained from the GCSP method is always less than that from TRAPP method. In the temperature range 411 - 511 K and the same pressure range, due to the extrapolation of the reference fluid data in GCSP method, the TRAPP method gave better results. However, when reference fluid data did not have to be extrapolated, then better predictions were obtained by the GCSP method.

For the mixture ethane + ethylene, the results of comparison are shown in Table 2.2. Over the temperature range 323 - 473 K and the pressure range 1 - 500/600 bar for several different compositions, still the GCSP method gave better results.

For the mixtures benzene + n-hexane and benzene + n-decane, the results of comparison are presented in Table 2.3. Experimental data were available at atmospheric pressure and over a limited temperature range. The GCSP method gave an average AAD of 4.0% for the benzene + n-hexane system and 8.8% for benzene + n-decane system. The TRAPP method, on the other hand, gave very large errors for the benzene + n-hexane system. This was due to the methane reference with shape factors predicting the wrong phase at 323 K. It is obvious that the ability of the GCSP method to use different reference fluids offers a tremendous advantage in the prediction of viscosities of components for which the shape factors are not valid.

It is demonstrated from the above comparison that the GCSP method can be used for the calculation of the viscosities of a variety of mixtures over a wide range of temperatures and pressures. In general, its predictions capability is better than those of the TRAPP method.

The use of two reference fluids in this method slightly improves the accuracy compared to TRAPP as shown above. But for multicomponent systems, the effects of the selection of reference fluids on the prediction capability are unknown because all systems tested in the literature are binary. On the other hand, if this method is used as a correlation technique, the number of the binary interaction coefficients will increase rapidly with the number of the components of the mixture.

Table 2.1

Viscosity Predictions for System Methane(1) - n-Butane(2) (x_1 = 0.396)
by the Two Corresponding States Methods

No. of Points	Temperature K	Pressure Range bar	GCSP AAD	TRAPP AAD
12	278	80 - 350	2.42	5.46
12	311	80 - 350	2.64	4.07
11	344	80 - 350	2.57	6.22
11	378	80 - 350	1.69	8.96
11	411	80 - 350	4.66	8.08
12	444	80 - 350	7.79	6.84
12	478	80 - 350	13.01	5.91
12	511	80 - 350	18.24	5.84

Table 2.2

Viscosity Predictions for system Ethane(1) - Ethylene(2)
by the Two Corresponding States Methods

x_1	No. of Points	Temperature range K	Pressure range bar	GCSP AAD	TRAPP AAD
0.354	60	323-473	1-600	5.97	5.96
0.699	56	323-473	1-500	8.25	8.97
0.813	60	323-473	1-600	6.67	7.21
			Overall	6.93	7.34

Table 2.3

Viscosity Predictions for Systems Benzene(1) - n-Hexane(2) and Benzene(1) - n-Decane(2) by the Two Corresponding States Methods

x_1	No. of Points	Temperature Range K	Pressure bar	GCSP AAD	TRAPP AAD	TRAPP* AAD
			Benzene(1) - n-Hexane(2)			
0.930	3	298 - 323	1.0	1.59	18.2	19.8
0.818	3	298 - 323	1.0	1.48	43.3	16.0
0.735	3	298 - 323	1.0	4.92	46.1	20.1
0.595	3	298 - 323	1.0	4.78	42.3	14.5
0.437	3	298 - 323	1.0	7.42	40.3	11.5
			Overall	4.04	38.0	16.4
			Benzene(1) - n-Decane(2)			
0.5	3	298 - 323	1.0	8.79	6.00	

* 323 K data omitted

2.3.8 Proposed modification of the Generalized Corresponding States Principle

Wu and Asfour (1991b) proposed a pseudo-binary mixture model to modify the GCSP method. The proposed modification of the GCSP method has the following two advantages over the original GCSP for n-alkane multicomponent mixtures: (i) the selections of reference fluids which could cause significant errors in predicting viscosity have been avoided, and (ii) the large numbers of the binary interaction coefficients which are essential for minimizing the deviations in the original GCSP for the viscosity correlation have been reduced to one no matter how many components are involved in a system.

In this model, the basic assumption is that the multicomponent mixture ($n \geq 3$) is considered as a binary system consisting of pure component 1 and pseudopure component 2', where component 2' is a mixture of components 2, 3, ...n. The reference fluids in this case are r1 = 1 and r2 = 2'. The required properties of component 2', the acentric factor, the critical properties and the reduced viscosity, are calculated from the corresponding properties of the pure liquid n-alkanes according to the principle of congruence (Brønsted and koefoed, 1946). The details of this modification are described in Chapter 5 later.

2.4 Viscosity Related Properties

2.4.1 Density

Because the kinematic viscosity of fluids, ν, is the variable actually measured, the density of these fluids needs to be measured at the same time to convert the kinematic viscosity to absolute viscosity, η, as follows

$$\eta = \nu \rho \tag{2.104}$$

Density can be measured with high precision.

2.4.2 Excess volume of mixing

Excess volume of mixing is defined as (Eyring *et al.*, 1969; Murrell and Boucher, 1982)

$$V^E = \Delta V_{mix} = V - \sum_i x_i V_i \tag{2.105}$$

i.e.

$$V^E = \frac{\sum_i x_i M_i}{\rho} - \sum_i \frac{x_i M_i}{\rho_i} \tag{2.106}$$

2.4.3 Excess viscosity of mixing

The excess of a property is defined as the difference between the real liquid property and the ideal liquid property (Eyring *et al.*, 1969). Therefore, the expression of excess viscosity is as follows:

$$\eta^E = \eta - \sum_i x_i \eta_i \tag{2.107}$$

2.4.4 Excess free energy of viscous flow

Eyring's rate process theory, eq. (2.34), gives the absolute viscosity as

$$\eta = \left(\frac{\alpha}{\lambda}\right)^2 \frac{hN_o}{V} e^{\frac{\Delta^{*}G}{RT}} \tag{2.108}$$

which can be rewritten as

$$\frac{\Delta^{*}G}{RT} = \ln(\eta V) + 2\ln\left(\frac{\lambda}{\alpha}\right) - \ln(hN_o) \tag{2.109}$$

For ideal solutions it was suggested (Reed and Taylor, 1959) that the excess molar free energy of viscous flow be given by

$$\Delta^{*}G^{i} = \sum_i x_i \Delta^{*}G_i \tag{2.110}$$

By definition (Heric and Brewer, 1967)

$$\Delta^{*}G^{E} = \Delta^{*}G - \Delta^{*}G^{i} \tag{2.111}$$

substituting eq. (2.109) into (2.111) yields

$$\frac{\Delta^{*}G^{E}}{RT} = \ln(\eta V) + 2\ln(\frac{\lambda}{\alpha}) - \ln(hN_{o}) - \sum_{i} x_{i}[\ln(\eta_{i}V_{i}) + 2\ln(\frac{\lambda_{i}}{\alpha_{i}}) - \ln(hN_{o})] \tag{2.112}$$

because

$$\ln(\frac{\lambda}{\alpha}) = \sum_{i} x_{i}\ln(\frac{\lambda_{i}}{\alpha_{i}}) \tag{2.36}$$

$\Delta^{*}G^{E}$ is related to viscosities and molar volumes by

$$\Delta^{*}G^{E} = RT[\ln(\eta V) - \sum_{i} x_{i}\ln(\eta_{i}V_{i})] \tag{2.113}$$

CHAPTER 3

EXPERIMENTAL EQUIPMENT AND PROCEDURES

3.1 Materials

The n-alkane samples used in this study were supplied by Aldrich Chemical Company. These are n-octane, n-decane, n-undecane, n-tridecane and n-pentadecane. The stated purity of the reagents is 99+ mol% with the exception of n-undecane which has a stated purity of 99 mol%. Gas chromatographic analysis and refractive index measurements confirmed that the purities of these chemicals were better than their stated values. Therefore, they were used without further purification. Table 3.1 lists the results of the refractive index measurements of the pure reagents. The results of gas chromatographic analysis are presented in Appendix D.

The chemicals used in viscometer calibration were Cannon Viscosity Standards: N.4, N.8, N1.0 and S3 obtained from Cannon Instrument Company.

The additional chemicals used in density meter calibration were n-hexane, toluene, benzene and carbon tetrachloride. They were all supplied by Aldrich Chemical Company and all have 99+ mol% stated purity. Also used in density meter calibration is double distilled water prepared in this laboratory.

Table 3.1

Values of Refractive Index of Pure Components

	Temperature = 293.15K		Temperature = 298.15 K	
Compound	Experimental Value[+]	Literature Value[*]	Experimental Value[+]	Literature Value[*]
n-Octane	1.3976	1.39743	1.39507	1.39505
n-Decane	1.41187	1.41189	1.40963	1.40967
n-Undecane	1.41723	1.41725	1.41503	1.41507
n-Tridecane	1.42577	1.42560	1.42347	1.42346
n-Pentadecane	1.4319	1.43188	1.42987	1.42979

[*] TRC Tables, 1988

[+] Refractometer used is an ABBE Refractometer type 1 supplied by ATAGO Company.

3.2 Preparation of Solutions

Ternary and binary solutions were prepared by weighing the individual components on a Mettler HK 160 electronic balance. The measuring range of the balance is 0 to 160 g. It has a reproducibility of ± 1 x 10^{-7} Kg. To avoid evaporation losses, the procedure suggested by Asfour (1980) was followed. According to that procedure, 0.03 L glass vials with teflon discs and aluminium seals were used which were all supplied by Chromatographic Specialities Ltd. The pure components were injected into the sealed vials by 0.01 L hypodermic glass syringes fitted with G24 stainless steel needles.

3.3 Viscosity Measurement

3.3.1 Equipment

Cannon-Ubbelohde capillary viscometers designed by Ubbelohde in 1935 and improved by Cannon *et al.* (1960) were used to measure the viscosities of the pure components and the mixtures at each temperature level indicated above. These viscometers have the following advantages (Asfour, 1980):

(i) no significant variation of viscometer constants with temperature,
(ii) exact filling volume control is not required,
(iii) they are capable of high precision in routine use.

In this study, two viscometers of size 25A (range 0.5 - 2.0 x 10^{-6} m^2/s), two viscometers of size 50B (range 0.8 - 4.0 x 10^{-6} m^2/s) and one viscometer of size 75J (range 1.6 - 8.0 x 10^{-6} m^2/s) were used. They were supplied by Cannon Instrument Company.

The temperature control of the viscosity measurement was achieved by a model M1-18M constant temperature bath also obtained from Cannon Instrument Company. The control range of the bath is from approximately 10°C above ambient to 100°C. The temperature fluctuation is ± 0.01°C below 60°C using water as bath medium and ± 0.02°C above 60°C using oil as bath medium. Mercury-in-glass thermoregulator is used by turning a magnetic cap to the expected temperature and the real temperature inside the bath is checked by a calibrated thermometer graduated to 0.01°C. A cooling coil is inside the bath which if connected with the refrigerating circulator can extend the working temperature range down. The control panel of this constant temperature bath has a pilot light, on-off switches for preheater, stirrer motor and a switch used to set heater control for operation below or above 54.4°C (130°F). Below 54.4°C, a relay operates a 200 watt heater to maintain control, above 54.4°C, the same relay operates a 700 watt heater.

3.3.2 Operation of viscometers

A liquid sample was introduced through tube 1, Figure 3.1, to fill bulb A until the level of the liquid was in between upper and lower etched marks on bulb A, with the viscometer mounted in vertical position. The viscometer supported by a plastic holder

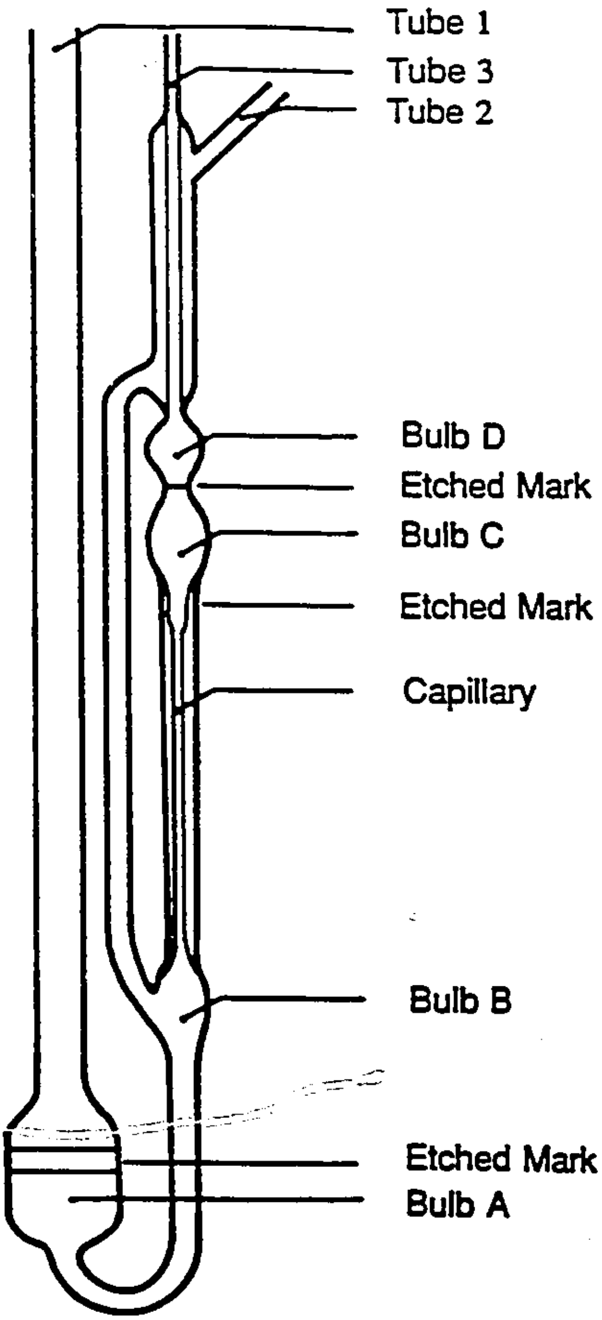


Figure 3.1. The Cannon-Ubbelohde Viscometer

was then placed in the model M1-18M constant temperature bath as shown in Figure 3.2. After the liquid sample reached thermal equilibrium with the bath, the viscosity measurement was started. Approximately 30 minutes is required for thermal equilibrium to be attained. Covering the opening of tube 2 with an on end clamped silicon rubber tubing, the liquid was sucked through the capillary from tube 3 by a pipette. Care must be taken so that no air bubbles are entrapped with the capillary. Once the liquid level crossed bulb D, the pipette was removed from tube 3 and the clamp in the silicon rubber tubing was also removed. The efflux time was determined by measuring the time taken by the liquid meniscus to travel from the upper to the lower etched marks on bulb C. At least three measurements were taken for each sample and the reproducibility was within ± 0.1%.

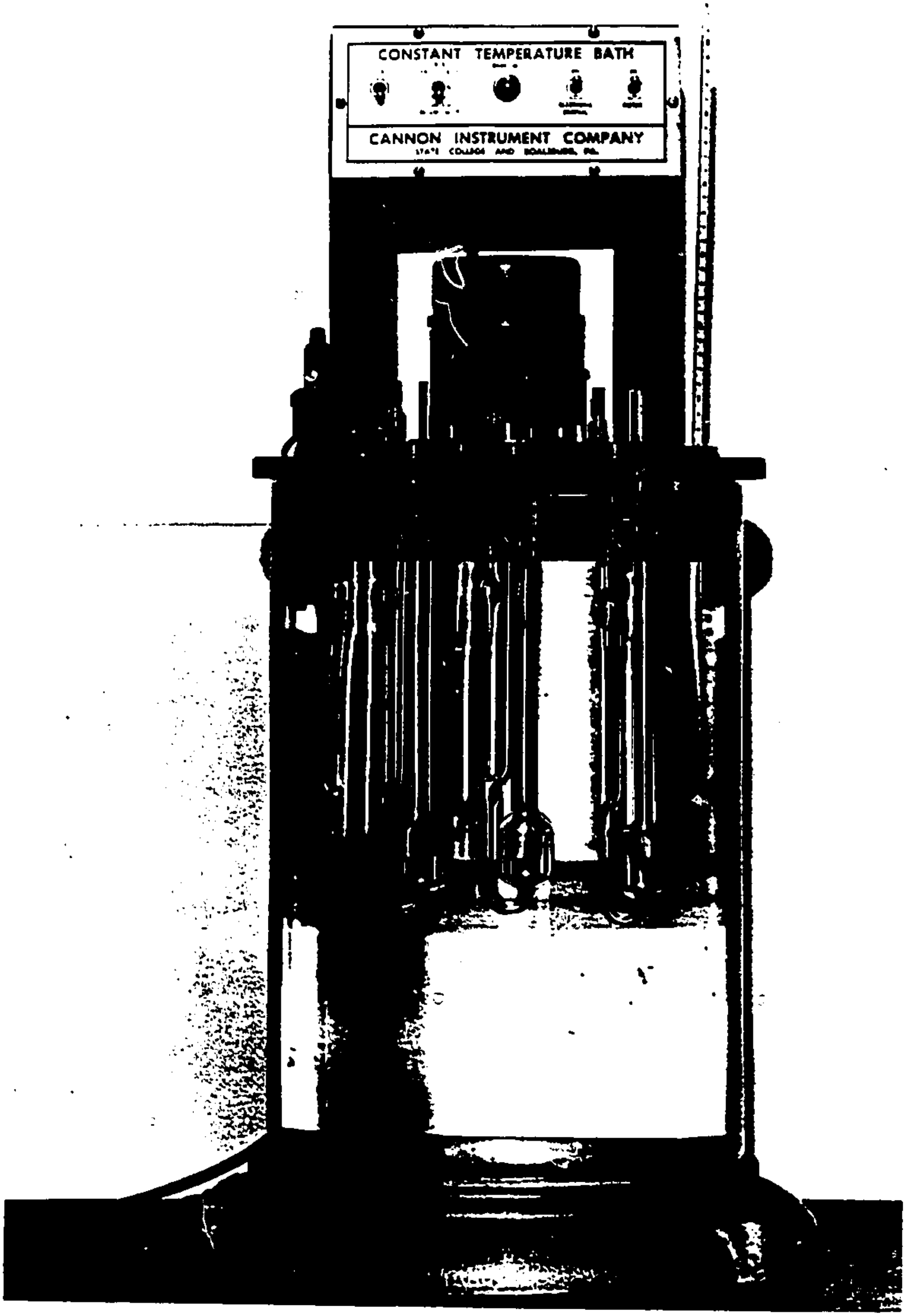
3.3.3 Viscosity equation

Cannon *et al.* (1960) proposed the following equation for calculating liquid viscosity from measured efflux time

$$\nu = Ct - \frac{E}{t^n} \tag{3.1}$$

where C and E are calibration constants determined by fitting viscosity - efflux time experimental data to eq.(3.1) using the method of least squares, and t is efflux time. The value of n depends on the shape of the capillary ends. The viscometers used in this study all have trumpet shaped ends, therefore, n is equal to 2. Thus the following viscosity

Figure 3.2. Pictorial View of the Constant Temperature Bath



VISCOSITY MEASURING APPARATUS

equation was used

$$\nu = Ct - \frac{E}{t^2} \tag{3.2}$$

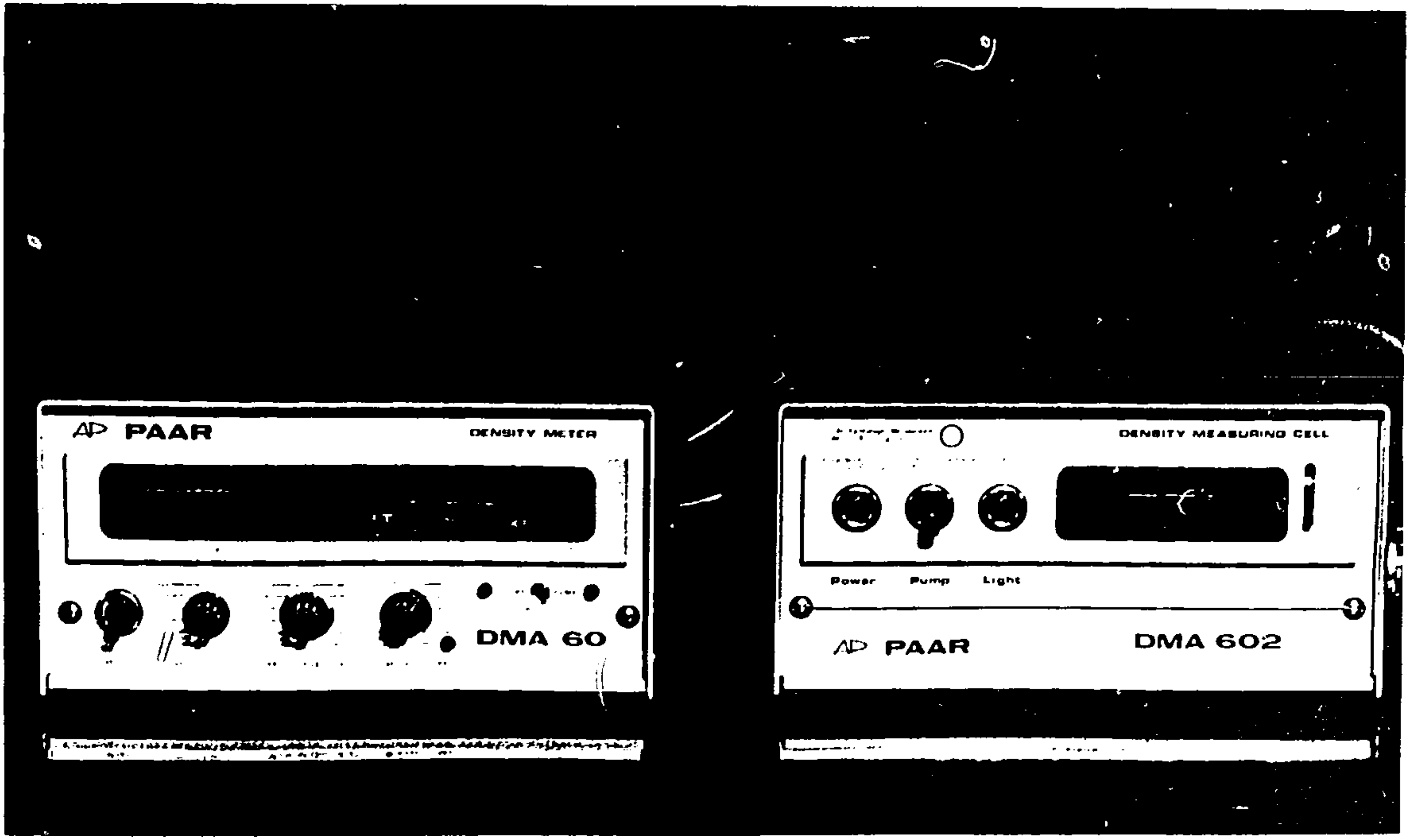
3.4 Density Measurement

3.4.1 Equipment

A high precision density meter (Anton Paar model DMA 60 with a measuring cell DMA 602) was used in this study. The density meter utilizes the oscillating sample tube method. Figure 3.3 shows a pictorial view of the system. Separating the remote cell and the electronic readout unit permits a better thermostating of the sample cell. It also offers a greater flexibility of the system configuration.

Density determination is based on measuring the period of oscillation of a hollow vibrating U-shaped sample tube, which is filled with sample liquid or through which the sample liquid flows continuously. The U-shaped sample tube (oscillator) is made of borosilicate glass (Duran 50) and is inside the DMA 602 remote measuring cell. This tube is fused into a dual-walled, thermostated glass cylinder. The space between the U-shaped sample tube and the inner wall of the dual wall cylinder is filled with a gas of high thermal conductivity to facilitate a rapid temperature equilibration of the sample inside the oscillator with the thermostat liquid which flows through the dual wall cylinder

Figure 3.3. Pictorial View of the Precision Density Meter



PRECISION DENSITY METER

around the sample tube. The oscillator is electronically excited in an undamped harmonic fashion. The direction of oscillation is perpendicular to the plane of the U-shaped sample tube. The frequency of the oscillator is only influenced by that fraction of the volume of liquid or gas which is actually in the vibrating part of the sample tube. It is essential to ensure that the oscillator (sample tube) is completely filled, overfilling does not affect the measurement. The remote cells generate frequency analog square wave signals in synchronization with the zero amplitude positions of the oscillating sample tube. These signals are transmitted into the DMA 60 processing unit through an optical isolator for the purpose of electrical separation (between remote cell and DMA 60). The period of vibration of the U-shaped sample tube is displayed to the eighth decimal place period meter with selectable resolution. With the simple relationship between the density of the sample and the natural frequency of the filled oscillator it is possible to use the method for the determination of the density of samples which are injected into or flowing through the oscillator.

The range of density that can be measured by this instrument is 0.5 to 2.0 g/cm^3. The sample size is 0.7 cm^3. The stated precision of the density meter is ± 1.5 x 10^{-6} kg/L. The achievable accuracy of density measurement depends on the achievable temperature stability. As a result of the temperature dependent changes of the oscillator, a temperature variation of ± 0.01°C will yield an uncertainty in the density determination of approximately ± 3 x 10^{-6} g/ cm^3. The temperature dependent density change of the sample must be added to this value.

DMA 602 can work within a temperature range of -10°C to + 70°C (controlled by ultrathermostat) and a pressure range from vacuum up to 10 bar. It takes power from standard 110 and 220 V alternating current permitting a fluctuation of ± 10%, within a frequency range of 40 - 60 Hz. It can operate under ambient condition of 0 to 45°C.

To maintain the sample fluid inside the sample tube at a desired temperature DMA 602 has to be coupled to a circulating thermostatic bath with the two sockets at the back of the instrument.

The DMA 60 processing unit is powered by alternating current with the ranges of voltage and frequency as same as the DMA 602 remote cell.

N4 electronic temperature controller is a high precision instrument supplied by Haake, West Germany. It has a precision of ± 0.01°C with the set-point ranging from 30°C to 300°C. Deionized water is used as the circulating fluid. Since the lowest set point permitted by the instrument is 30°C, a refrigerating circulator is required to couple with it to obtain temperature control at 20 and 25°C in the DMA 602 measuring cell.

As suggested earlier by Asfour (1980), the density meter is placed in a plywood box with a plexiglass front door. The temperature inside the box was controlled at the working temperature, i.e. 20, 25, 35 and 40 ± 0.1°C. The box is fitted with an exhaust fan, a 200 watt household bulb (two bulbs were used for temperatures at 35 and 40°C), a transmission cooler which is connected to a refrigerating circulator and a precise thermonitor. The Sargent-Welch Thermonitor (model ST) was supplied by Sargent-Welch Scientific Co., U.S.A. This thermonitor is connected to the household bulb. The transmission cooler provides necessary cooling inside the box. When the temperature in the box becomes lower than the set-point the bulb lights up to bring the temperature to the required level. When the temperature approaches the set-point the power supplied by the thermonitor decreases until it is completely cut-off as the temperature reaches the set-point. The exhaust fan provides the required air circulation. The temperature in the box is monitored by a mercury-in-glass thermometer graduated to 0.1°C. Controlling the temperature in the box in this way has been proven to give extremely reliable results (Asfour, 1980).

3.4.2 Procedure

After thorough cleaning with ethanol and drying with the pump installed in DMA 602, the U-shaped sample tube was filled with the liquid sample which was injected by a 0.002 L glass hypodermic syringe. The ends of the sample tube were covered by Teflon cocks. The injection procedure should be performed very carefully to prevent entrapping any air bubbles and to ensure fully wetting of the wall of the sample tube. At least 20 minutes should be allowed for the sample to reach thermal equilibrium at the designated temperature. The "start" button was pressed and the DMA 60 unit would display the readings of the oscillation period of the sample tube. At least ten readings were taken for one sample, then the average was recorded and the density was calculated from the reading of period.

3.4.3 Density meter equation

The equation used in this study for calculating sample density from the vibrating period of the oscillator was suggested by the supplier of the density meter

$$\rho = \frac{AT^2}{1 - BT^2} - C \tag{3.3}$$

where A, B and C are calibration constants which have to be determined from known density values by the method of least squares. T is the period of oscillation.

CHAPTER 4

EXPERIMENTAL RESULTS

4.1 General

Five selected ternary n-alkane liquid systems and their corresponding eight binary subsystems were investigated. The range of the n-alkanes is from C_8 to C_{15}, which represents liquid n-alkanes. The investigated systems are listed in Table 4.1. In Table 4.1, the component 1, 2 and 3 are arranged according to an ascending order of the carbon numbers. Transport properties and thermodynamic properties of these systems are not available in the literature.

The densities and viscosities of both pure components and mixtures were measured over the entire composition range at four temperature levels: 293.15, 298.15, 308.15 and 313.15 K. These data represent very valuable addition to the literature. They are needed both for their own value and for the testing and modifying of various models which are used to predict volumetric and viscometric properties of n-alkane liquid mixtures. The excess properties of both binary and ternary systems calculated from the experimental data are also reported in this chapter.

4.2 Calibration of the Density Meter

The density meter was calibrated at 293.15, 298.15, 308.15 and 313.15 K,

TABLE 4.1

Investigated Ternary Systems and Binary Subsystems

TERNARY SYSTEMS
n-Octane(1)-n-Undecane(2)-n-Tridecane(3)
n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)
n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)
n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)
n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)
BINARY SUBSYSTEMS
n-Octane(1)-n-Undecane(2)
n-Octane(1)-n-Tridecane(2)
n-Octane(1)-n-Pentadecane(2)
n-decane(1)-n-Pentadecane(2)
n-Undecane(1)-n-Pentadecane(2)
n-Tridecane(1)-n-Pentadecane(2)
n-Decane(1)-n-Tridecane(2)
n-Undecane(1)-n-Tridecane(2)

respectively, by selected liquids of known density, the density range of these calibration liquids covers the density range of interest. The results of calibration are reported in Table 4.2. The three parameters in the density equation were determined from the density of the calibration liquids by the non-linear regression.

The accuracy of the density meter depends on that of the density of the standard liquids used for calibration. Therefore, these liquids were selected very carefully to provide density values as accurate as possible.

4.3 Calibration of the Viscometers

Five different size Cannon-Ubbelohde type viscometers were used in this study, two were 25A type, two were 50B type and one was 75J type. The measuring ranges of these viscometers have been reported in Chapter 3.

These viscometers were calibrated with Calibration Standards purchased from Cannon Instrument Company at the temperatures of interest. The range of the standards covers the viscosity range of interest. At least five measurements were performed for each standards to get an average reading, the reproducibility is ± 0.1%. The two parameters in the viscosity equation were calculated by the least squares method.

The results of calibration along with the standard deviations of fit are listed in Table 4.3.

Table 4.2

Calibration Data for the Density Meter

Substance	Density, kg/L	Density Meter Reading, sec
	Temperature = 293.15 K	
n-Hexane	0.65925 (TRC Tables, 1988)	0.332153
n-Octane	0.70267 (TRC Tables, 1988)	0.336094
n-Decane	0.73012 (TRC Tables, 1988)	0.338585
Toluene	0.8669 (TRC Tables, 1986)	0.350805
Benzene	0.8790 (TRC Tables, 1986)	0.351880
Double Distilled Water	0.9982343 (Perry, 1950)	0.362166
Carbon Tetrachloride	1.5940 (Timmermans, 1950)	0.409697

Parameter	Value
A	16.352364
B	-0.024420
C	1.139585

Standard Diviation of the Fit = 2.92 x 10^{-4} kg/L

Substance	Density, kg/L	Density Meter Reading, sec
	Temperature = 298.15 K	
n-Hexane	0.65471 (TRC Tables, 1988)	0.331633
n-Octane	0.69862 (TRC Tables, 1988)	0.335622
n-Decane	0.72635 (TRC Tables, 1988)	0.338144
Toluene	0.86220 (Timmermans, 1950)	0.350292
Benzene	0.87366 (Timmermans, 1950)	0.351309
Double Distilled Water	0.99707 (Perry, 1950)	0.361959
Carbon Tetrachloride	1.58445 (Timmermans, 1950)	0.408848

Parameter	Value
A	16.343878
B	-0.019993
C	1.138456

Standard Diviation of the Fit = 2.83 x 10^{-4} kg/L

Table 4.2 (cont'd)

Calibration Data for the Density Meter

Substance	Density, kg/L	Density Meter Reading, sec
Temperature = 308.15 K		
N.4	0.6473 (Viscosity Standard)	0.330700
n-Octane	0.69042 (Garcia *et al.*, 1986)	0.334679
n-Decane	0.71915 (Inglese *et al.*, 1983)	0.337252
N1.0	0.7768 (Viscosity Standard)	0.342533
S3	0.8295 (Viscosity Standard)	0.347226
Benzene	0.86296 (Timmermans, 1965)	0.350164
Double Distilled Water	0.9940610 (Perry, 1950)	0.361490

Parameter	Value
A	16.249367
B	0.002962
C	1.130187

Standard Deviation of the Fit = 3.05 x 10^{-4} kg/L

Substance	Density, kg/L	Density Meter Reading, sec
Temperature = 313.15 K		
n-Octane	0.6863 (TRC Tables, 1988)	0.334206
n-Decane	0.7150 (TRC Tables, 1988)	0.336808
Toluene	0.8485 (TRC Tables, 1986)	0.348754
Benzene	0.8577 (TRC Tables, 1986)	0.349595
Double Distilled Water	0.9922497 (Perry, 1950)	0.361233
Carbon Tetrachloride	1.55498 (Timmermans, 1965)	0.406289

Patameter	Value
A	16.318019
B	-0.010458
C	1.134018

Standard Deviation of Fit = 2.26 x 10^{-4} kg/L

Table 4.3

Calibration Data for the Viscometers

Temperature = 293.15 K

Liquid	Efflux Time sec	Standard Viscosity 10^{-6} m²/s	Calculated Viscosity 10^{-6} m²/s	Parameters C 10^{-9} m²/s²	Parameters E 10^{-6} m²s	Standard Deviation 10^{-6} m²/s
			Viscometer 25A483			
N.4	261.03	0.4709	0.4710	1.80136	-54.6259	4.43x10^{-4}
N.8	431.52	0.7780	0.7776			
N1.0	720.61	1.298	1.2982			
			Viscometer 25A498			
N.4	255.60	0.4709	0.4712	1.84180	-26.8300	1.04x10^{-3}
N.8	421.84	0.7780	0.7771			
N1.0	704.96	1.298	1.2984			
			Viscometer 50B158			
N.8	182.07	0.7780	0.7787	4.27295	-25.6315	2.27x10^{-3}
N1.0	303.22	1.298	1.2959			
S3	1004.10	4.290	4.2905			
			Viscometer 50B159			
N.8	200.46	0.7780	0.7786	3.88383	-2.73914	1.91x10^{-3}
N1.0	333.75	1.298	1.2962			
S3	1104.69	4.290	4.2904			
			Viscometer 75J29			
N.8	89.29	0.7780	0.7790	8.76835	-30.8842	3.15x10^{-3}
N1.0	147.86	1.298	1.2951			
S3	489.35	4.290	4.2907			

Table 4.3 (cont'd)

Calibration Data for the Viscometers

Temperature = 298.15 K

Liquid	Efflux Time sec	Standard Viscosity 10^{-6} m^2/s	Calculated Viscosity 10^{-6} m^2/s	Parameters		Standard Deviation 10^{-6} m^2/s
				C 10^{-9} m^2/s^2	E 10^{-6} m^2·s	
			Viscometer 25A483			
N.4	250.29	0.4508	0.4510	1.80140	-9.54884	8.14×10^{-4}
N.8	408.15	0.7360	0.7353			
N1.0	670.77	1.208	1.2084			
			Viscometer 25A498			
N.4	244.87	0.4508	0.4509	1.84205	8.70242	4.63×10^{-4}
N.8	399.37	0.7360	0.7356			
N1.0	655.91	1.208	1.2082			
			Viscometer 50B158			
N.8	172.35	0.7360	0.7365	4.26988	-16.1933	1.36×10^{-3}
N1.0	282.57	1.208	1.2068			
S3	886.98	3.787	3.7873			
			Viscometer 50B159			
N.8	189.76	0.7360	0.7366	3.88215	4.16643	1.67×10^{-3}
N1.0	310.79	1.208	1.2065			
S3	975.59	3.787	3.7874			
			Viscometer 75J29			
N1.0	137.83	1.208	1.2080	8.76487	0.34293	
S3	432.07	3.787	3.7870			

Table 4.3 (cont'd)

Calibration Data for the Viscometers

Temperature = 308.15 K

Liquid	Efflux Time sec	Standard Viscosity 10^{-6} m²/s	Calculated Viscosity 10^{-6} m²/s	Parameters C 10^{-9} m²/s²	Parameters E 10^{-6} m²·s	Standard Deviation 10^{-6} m²/s
Viscometer 25A483						
N.4	230.96	0.4154	0.4156	1.79564	-48.20034	8.46x10^{-4}
N.8	368.07	0.6620	0.6613			
N1.0	589.33	1.058	1.0584			
Viscometer 25A498						
N.4	226.10	0.4154	0.4156	1.83638	-21.49524	8.14x10^{-4}
N.8	360.02	0.6620	0.6613			
N1.0	576.29	1.058	1.0584			
Viscometer 50B158						
N1.0	248.25	1.058	1.0580	4.2465	-234.51722	
S3	711.06	3.020	3.0200			
Viscometer 50B159						
N1.0	273.05	1.058	1.0580	3.8591	-320.69093	
S3	782.44	3.020	3.0200			
Viscometer 75J29						
N1.0	121.27	1.058	1.0580	8.71520	-15.78132	
S3	346.51	3.020	3.0200			

Table 4.3 (cont'd)

Calibration Data for the Viscometers

Temperature = 313.15 K

Liquid	Efflux Time sec	Standard Viscosity 10^{-6} m²/s	Calculated Viscosity 10^{-6} m²/s	Parameters C 10^{-9} m²/s²	Parameters E 10^{-6} m²·s	Standard Deviation 10^{-6} m²/s
			Viscometer 25A483			
N.4	221.99	0.3989	0.3983	1.79375	-3.90757	2.33×10^{-3}
N.8	352.09	0.6296	0.6316			
N1.0	553.74	0.9943	0.9933			
			Viscometer 25A498			
N.4	217.41	0.3989	0.3992	1.83504	-10.40324	9.85×10^{-4}
N.8	342.59	0.6296	0.6288			
N1.0	542.05	0.9943	0.9947			
			Viscometer 50B158			
N1.0	233.33	0.9943	0.9943	4.24290	-235.32122	
S3	641.88	2.724	2.7240			
			Viscometer 50B159			
N1.0	256.72	0.9943	0.9943	3.85660	-279.49511	
S3	706.18	2.724	2.7240			
			Viscometer 75J29			
N1.0	114.01	0.9943	0.9943	8.70956	-16.74097	
S3	312.74	2.724	2.7240			

4.4 Binary System Data

Figures 4.1 to 4.4 show the density isotherms of all the binary mixtures at the four temperature levels investigated.

The experimental density-composition data and the calculated excess volume-composition data of the eight binary systems are presented in Tables 4.4 to 4.11.

The viscosity isotherms of all the binary mixtures were shown in Figures 4.5 to 4.8. It is obvious that viscosity is not a simple function of composition.

The experimental viscosity-composition data and the excess properties-composition data, i. e. the excess viscosity and excess energy of viscous flow, calculated from viscosity data are listed in Tables 4.12 to 4.19.

4.5 Ternary System Data

The experimental density and viscosity data of the five ternary systems are listed in Tables 4.20 to 4.24 whereas their corresponding excess volume, excess viscosity and excess energy of viscous flow data are reported in Appendix B, Tables B.1 to B.5.

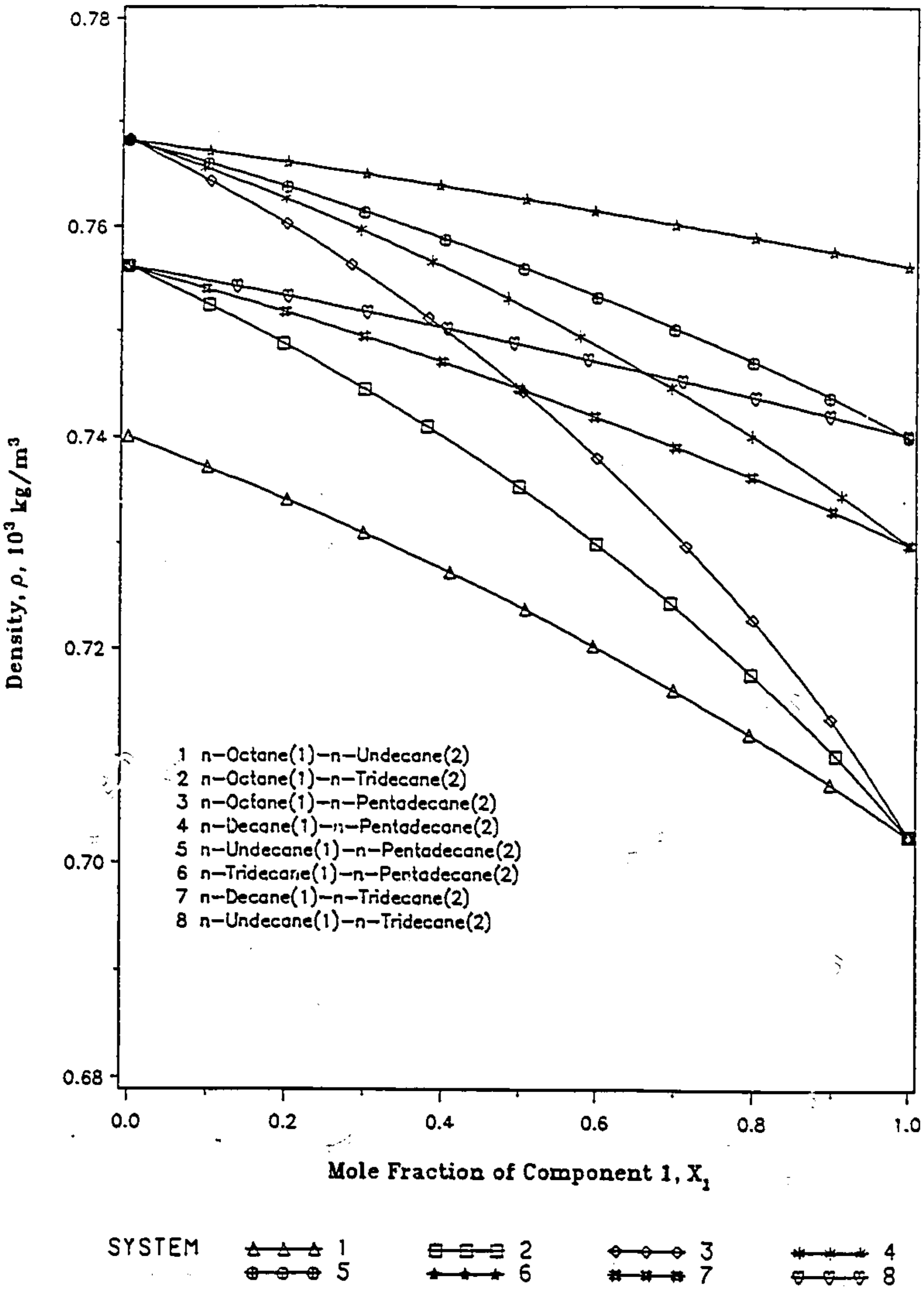


Figure 4.1. Density vs. Composition for Binary n-Alkane Systems at 293.15 K

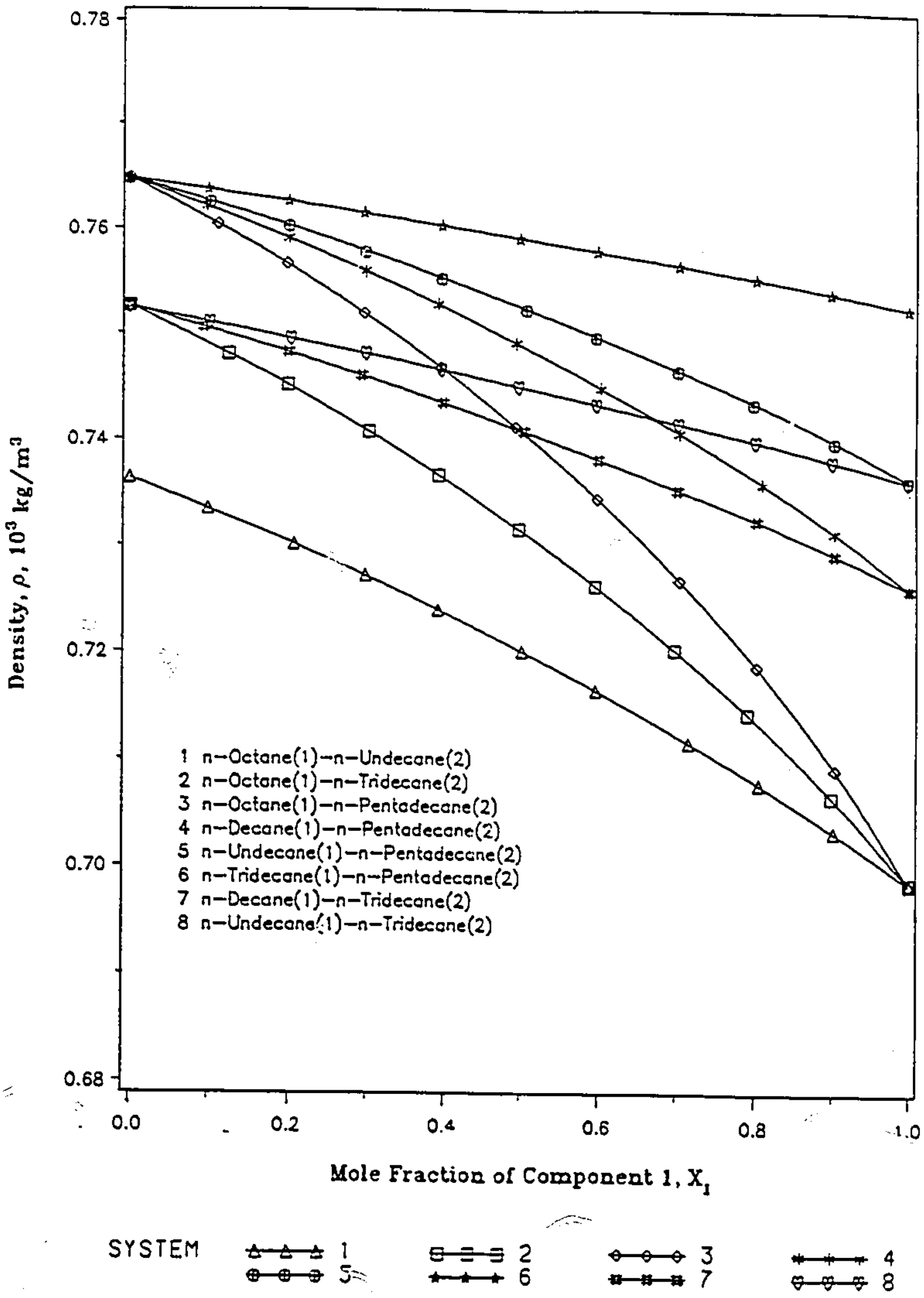


Figure 4.2. Density vs. Composition for Binary n-Alkane Systems at 298.15 K

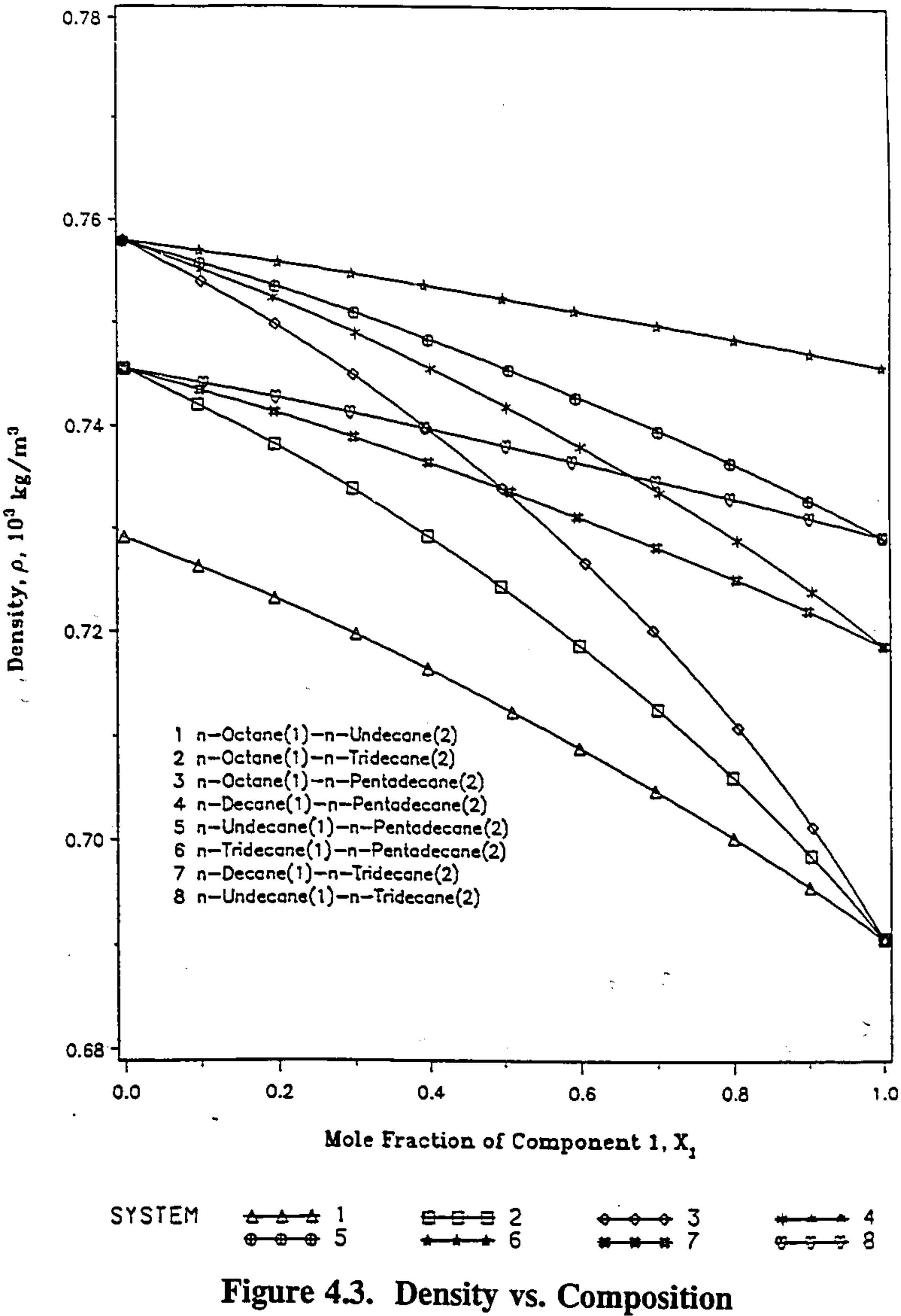


Figure 4.3. Density vs. Composition for Binary n-Alkane Systems at 308.15 K

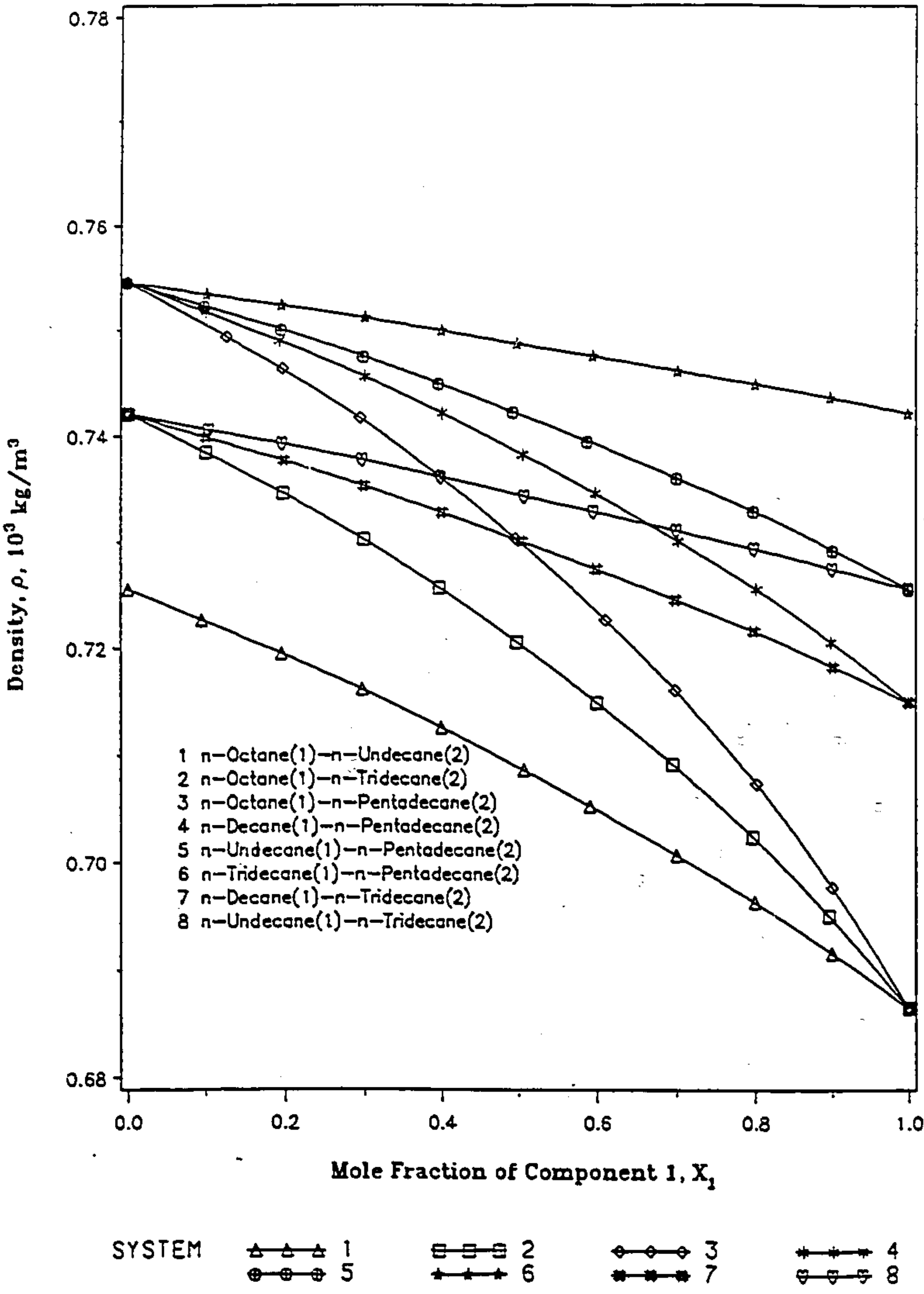


Figure 4.4. Density vs. Composition
for Binary n-Alkane Systems at 313.15 K

Table 4.4

Densities and Excess Volumes of the System n-Octane(1)-n-Undecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7401	0.0000	0.0000	0.7363	0.0000
0.1007	0.7371	-0.1060	0.0992	0.7335	-0.0252
0.2018	0.7341	-0.0384	0.2059	0.7302	-0.0423
0.2995	0.7309	-0.0406	0.2978	0.7272	-0.0561
0.4108	0.7272	-0.0568	0.3916	0.7240	-0.0602
0.5076	0.7237	-0.0597	0.5019	0.7200	-0.0681
0.5964	0.7203	-0.0523	0.5959	0.7164	-0.0681
0.6995	0.7161	-0.0413	0.7154	0.7116	-0.0591
0.7954	0.7120	-0.0274	0.8041	0.7077	-0.0477
0.8978	0.7074	-0.0088	0.9011	0.7033	-0.0321
1.0000	0.7025	0.0000	1.0000	0.6984	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7291	0.0000	0.0000	0.7255	0.0000
0.0969	0.7263	-0.0312	0.0953	0.7227	-0.0232
0.1954	0.7232	-0.0463	0.1962	0.7195	-0.0454
0.3001	0.7197	-0.0651	0.2979	0.7161	-0.0694
0.3970	0.7164	-0.0787	0.3995	0.7125	-0.0730
0.5094	0.7122	-0.0793	0.5061	0.7086	-0.0829
0.5981	0.7088	-0.0825	0.5914	0.7052	-0.0813
0.6968	0.7047	-0.0743	0.7010	0.7006	-0.0734
0.7992	0.7002	-0.0611	0.3001	0.6963	-0.0639
0.8998	0.6955	-0.0392	0.8999	0.6915	-0.0391
1.0000	0.6905	0.0000	1.0000	0.6865	0.0000

Table 4.5

Densities and Excess Volumes of the System n-Octane(1)-n-Tridecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7562	0.0000	0.0000	0.7526	0.0000
0.1023	0.7525	-0.0355	0.1246	0.7481	-0.0432
0.1966	0.7488	-0.0582	0.1988	0.7452	-0.0697
0.3011	0.7445	-0.0807	0.3028	0.7408	-0.0906
0.3813	0.7409	-0.1075	0.3936	0.7366	-0.1094
0.4993	0.7352	-0.1373	0.4971	0.7315	-0.1171
0.6002	0.7298	-0.1408	0.5956	0.7262	-0.1229
0.6949	0.7243	-0.1345	0.6970	0.7202	-0.1169
0.7976	0.7176	-0.0807	0.7898	0.7143	-0.0978
0.9047	0.7100	-0.0486	0.8988	0.7065	-0.0612
1.0000	0.7025	0.0000	1.0000	0.6985	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7456	0.0000	0.0000	0.7421	0.0000
0.0968	0.7420	-0.0380	0.0995	0.7385	-0.0553
0.1961	0.7381	-0.0803	0.1969	0.7346	-0.1024
0.2969	0.7338	-0.1104	0.2992	0.7302	-0.1357
0.3966	0.7292	-0.1276	0.3967	0.7256	-0.1568
0.4942	0.7243	-0.1376	0.4967	0.7206	-0.1710
0.5983	0.7186	-0.1426	0.5993	0.7149	-0.1646
0.7006	0.7125	-0.1361	0.6949	0.7091	-0.1683
0.7985	0.7060	-0.1110	0.7971	0.7023	-0.1411
0.9019	0.6985	-0.0644	0.8964	0.6950	-0.0831
1.0000	0.6905	0.0000	1.0000	0.6865	0.0000

Table 4.6

Densities and Excess Volumes of the System n-Octane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7684	0.0000	0.0000	0.7648	0.0000
0.1032	0.7643	-0.0494	0.1114	0.7604	-0.0629
0.2005	0.7602	-0.1000	0.1978	0.7567	-0.0966
0.2833	0.7563	-0.1208	0.2962	0.7520	-0.1273
0.3826	0.7512	-0.1351	0.3951	0.7469	-0.1571
0.5038	0.7442	-0.1471	0.4927	0.7413	-0.1810
0.6008	0.7380	-0.1605	0.5978	0.7345	-0.1993
0.7150	0.7296	-0.1493	0.7035	0.7268	-0.1875
0.7994	0.7227	-0.1292	0.8008	0.7187	-0.1601
0.8985	0.7134	-0.0793	0.9026	0.7091	-0.1030
1.0000	0.7025	0.0000	1.0000	0.6984	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7580	0.0000	0.0000	0.7545	0.0000
0.1015	0.7539	-0.0661	0.1274	0.7494	-0.0953
0.1973	0.7500	-0.1180	0.1958	0.7463	-0.1285
0.2986	0.7449	-0.1655	0.2936	0.7417	-0.1891
0.3975	0.7397	-0.1959	0.3949	0.7363	-0.2192
0.4973	0.7338	-0.2210	0.4956	0.7304	-0.2537
0.6065	0.7266	-0.2277	0.6113	0.7226	-0.2529
0.6944	0.7201	-0.2182	0.6990	0.7161	-0.2489
0.8053	0.7108	-0.1763	0.8018	0.7073	-0.1974
0.9040	0.7012	-0.1133	0.9006	0.6977	-0.1203
1.0000	0.6905	0.0000	1.0000	0.6865	0.0000

Table 4.7

Densities and Excess Volumes of the System n-Decane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7683	0.0000	0.0000	0.7648	0.0000
0.0958	0.7656	-0.0005	0.0973	0.7621	-0.0305
0.1989	0.7626	-0.0179	0.1999	0.7591	-0.0443
0.2947	0.7596	-0.0238	0.2983	0.7560	-0.0506
0.3865	0.7566	-0.0340	0.3925	0.7529	-0.0650
0.4839	0.7531	-0.0369	0.4933	0.7492	-0.0619
0.5780	0.7495	-0.0356	0.6025	0.7450	-0.0641
0.6958	0.7446	-0.0303	0.7033	0.7407	-0.0652
0.7980	0.7400	-0.0219	0.8079	0.7360	-0.0516
0.9126	0.7344	-0.0114	0.9014	0.7313	-0.0396
1.0000	0.7298	0.0000	1.0000	0.7260	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7580	0.0000	0.0000	0.7546	0.0000
0.1015	0.7551	-0.0225	0.0999	0.7518	-0.0181
0.1942	0.7523	-0.0390	0.1932	0.7490	-0.0421
0.3018	0.7489	-0.0589	0.3013	0.7456	-0.0697
0.4028	0.7454	-0.0612	0.4013	0.7421	-0.0842
0.5033	0.7418	-0.0728	0.5059	0.7382	-0.0869
0.6010	0.7379	-0.0731	0.5986	0.7345	-0.0903
0.7040	0.7335	-0.0736	0.7028	0.7301	-0.0774
0.8051	0.7289	-0.0597	0.8009	0.7255	-0.0632
0.9044	0.7239	-0.0381	0.8988	0.7205	-0.0338
1.0000	0.7186	0.0000	1.0000	0.7150	0.0000

Table 4.8

Densities and Excess Volumes of the System n-Undecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7683	0.0000	0.0000	0.7648	0.0000
0.1013	0.7660	-0.0143	0.1026	0.7626	-0.0154
0.2015	0.7637	-0.0159	0.2004	0.7603	-0.0231
0.3004	0.7613	-0.0240	0.2994	0.7579	-0.0267
0.4036	0.7587	-0.0271	0.3983	0.7554	-0.0348
0.5046	0.7560	-0.0258	0.5079	0.7524	-0.0365
0.6010	0.7532	-0.0334	0.5973	0.7498	-0.0325
0.7004	0.7502	-0.0277	0.7022	0.7466	-0.0334
0.8000	0.7470	-0.0207	0.7975	0.7436	-0.0281
0.8974	0.7437	-0.0156	0.9019	0.7400	-0.0161
1.0000	0.7400	0.0000	1.0000	0.7364	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7580	0.0000	0.0000	0.7546	0.0000
0.1005	0.7557	-0.0125	0.0989	0.7523	-0.0054
0.1976	0.7535	-0.0278	0.1959	0.7501	-0.0220
0.3007	0.7509	-0.0394	0.2996	0.7475	-0.0299
0.4013	0.7483	-0.0366	0.3974	0.7450	-0.0458
0.5074	0.7454	-0.0459	0.4945	0.7423	-0.0455
0.5964	0.7428	-0.0381	0.5890	0.7395	-0.0518
0.7036	0.7395	-0.0504	0.7019	0.7360	-0.0372
0.7972	0.7364	-0.0301	0.7983	0.7328	-0.0357
0.9014	0.7328	-0.0230	0.9013	0.7292	-0.0109
1.0000	0.7291	0.0000	1.0000	0.7255	0.0000

Table 4.9

Densities and Excess Volumes of the System n-Tridecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7683	0.0000	0.0000	0.7648	0.0000
0.1030	0.7672	-0.0057	0.1000	0.7637	-0.0002
0.2020	0.7660	-0.0051	0.2001	0.7626	-0.0049
0.3023	0.7649	-0.0076	0.2975	0.7615	-0.0018
0.3967	0.7638	-0.0069	0.3984	0.7603	-0.0061
0.5078	0.7625	-0.0056	0.5000	0.7591	-0.0048
0.5972	0.7614	-0.0073	0.5985	0.7579	-0.0011
0.7003	0.7601	-0.0092	0.7037	0.7566	-0.0107
0.8019	0.7588	-0.0053	0.8004	0.7553	-0.0031
0.9018	0.7575	-0.0023	0.8983	0.7540	0.0079
1.0000	0.7561	0.0000	1.0000	0.7526	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7580	0.0000	0.0000	0.7546	0.0000
0.1004	0.7569	0.0042	0.1024	0.7534	-0.0076
0.2017	0.7557	-0.0045	0.1963	0.7523	-0.0018
0.2979	0.7546	-0.0067	0.3028	0.7511	-0.0060
0.3944	0.7535	-0.0036	0.4019	0.7499	-0.0108
0.4994	0.7522	-0.0116	0.4997	0.7487	-0.0002
0.5935	0.7511	-0.0074	0.5970	0.7475	-0.0101
0.7015	0.7497	-0.0132	0.7040	0.7462	-0.0121
0.8030	0.7483	-0.0072	0.8017	0.7448	-0.0002
0.9036	0.7470	0.0018	0.8985	0.7435	-0.0072
1.0000	0.7456	0.0000	1.0000	0.7421	0.0000

Table 4.10

Densities and Excess Volumes of the System n-Decane(1)-n-Tridecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7561	0.0000	0.0000	0.7526	0.0000
0.0987	0.7540	-0.0126	0.0939	0.7506	-0.0078
0.1990	0.7518	-0.0185	0.1985	0.7483	-0.0197
0.2995	0.7495	-0.0254	0.2931	0.7461	-0.0211
0.3997	0.7470	-0.0283	0.3988	0.7435	-0.0273
0.4986	0.7445	-0.0237	0.5031	0.7408	-0.0226
0.5974	0.7419	-0.0296	0.5995	0.7382	-0.0254
0.7015	0.7390	-0.0287	0.7013	0.7353	-0.0238
0.7973	0.7362	-0.0173	0.8001	0.7324	-0.0275
0.9014	0.7330	-0.0115	0.9011	0.7292	-0.0131
1.0000	0.7297	0.0000	1.0000	0.7260	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7456	0.0000	0.0000	0.7421	0.0000
0.0991	0.7435	-0.0206	0.1001	0.7399	-0.0108
0.1956	0.7413	-0.0331	0.1979	0.7377	-0.0226
0.2991	0.7388	-0.0373	0.2988	0.7353	-0.0368
0.3993	0.7363	-0.0366	0.3996	0.7328	-0.0444
0.5073	0.7335	-0.0396	0.5025	0.7301	-0.0444
0.5962	0.7311	-0.0400	0.5994	0.7274	-0.0449
0.6991	0.7282	-0.0443	0.6995	0.7245	-0.0441
0.8037	0.7250	-0.0359	0.7980	0.7215	-0.0316
0.8994	0.7220	-0.0232	0.9013	0.7182	-0.0227
1.0000	0.7186	0.0000	1.0000	0.7148	0.0000

Table 4.11

Densities and Excess Volumes of the System n-Undecane(1)-n-Tridecane(2)

Mole Fraction x_1	Density kg/L	Excess Volume L/kmol	Mole Fraction x_1	Density kg/L	Excess Volume L/kmol
Temperature = 293.15 K			Temperature = 298.15 K		
0.0000	0.7562	0.0000	0.0000	0.7525	0.0000
0.1389	0.7542	-0.0053	0.1023	0.7511	-0.0074
0.2035	0.7533	-0.0061	0.2031	0.7496	-0.0068
0.3044	0.7518	-0.0076	0.2991	0.7482	-0.0104
0.4076	0.7502	-0.0063	0.3981	0.7466	-0.0103
0.4924	0.7488	-0.0034	0.4981	0.7450	-0.0080
0.5897	0.7473	-0.0094	0.5976	0.7434	-0.0072
0.7108	0.7452	-0.0079	0.7028	0.7416	-0.0075
0.8027	0.7436	-0.0007	0.8000	0.7399	-0.0092
0.8980	0.7419	0.0043	0.9004	0.7381	-0.0003
1.0000	0.7401	0.0000	1.0000	0.7362	0.0000
Temperature = 308.15 K			Temperature = 313.15 K		
0.0000	0.7456	0.0000	0.0000	0.7421	0.0000
0.1051	0.7441	-0.0006	0.1040	0.7406	-0.0027
0.1986	0.7428	-0.0118	0.1964	0.7393	-0.0127
0.2953	0.7413	-0.0116	0.2972	0.7377	-0.0119
0.3954	0.7397	-0.0107	0.3982	0.7361	-0.0157
0.5039	0.7379	-0.0071	0.5078	0.7343	-0.0129
0.5908	0.7365	-0.0111	0.5968	0.7328	-0.0153
0.6994	0.7346	-0.0140	0.7018	0.7310	-0.0209
0.7972	0.7329	-0.0159	0.7985	0.7293	-0.0080
0.9027	0.7310	-0.0099	0.9009	0.7274	-0.0096
1.0000	0.7291	0.0000	1.0000	0.7255	0.0000

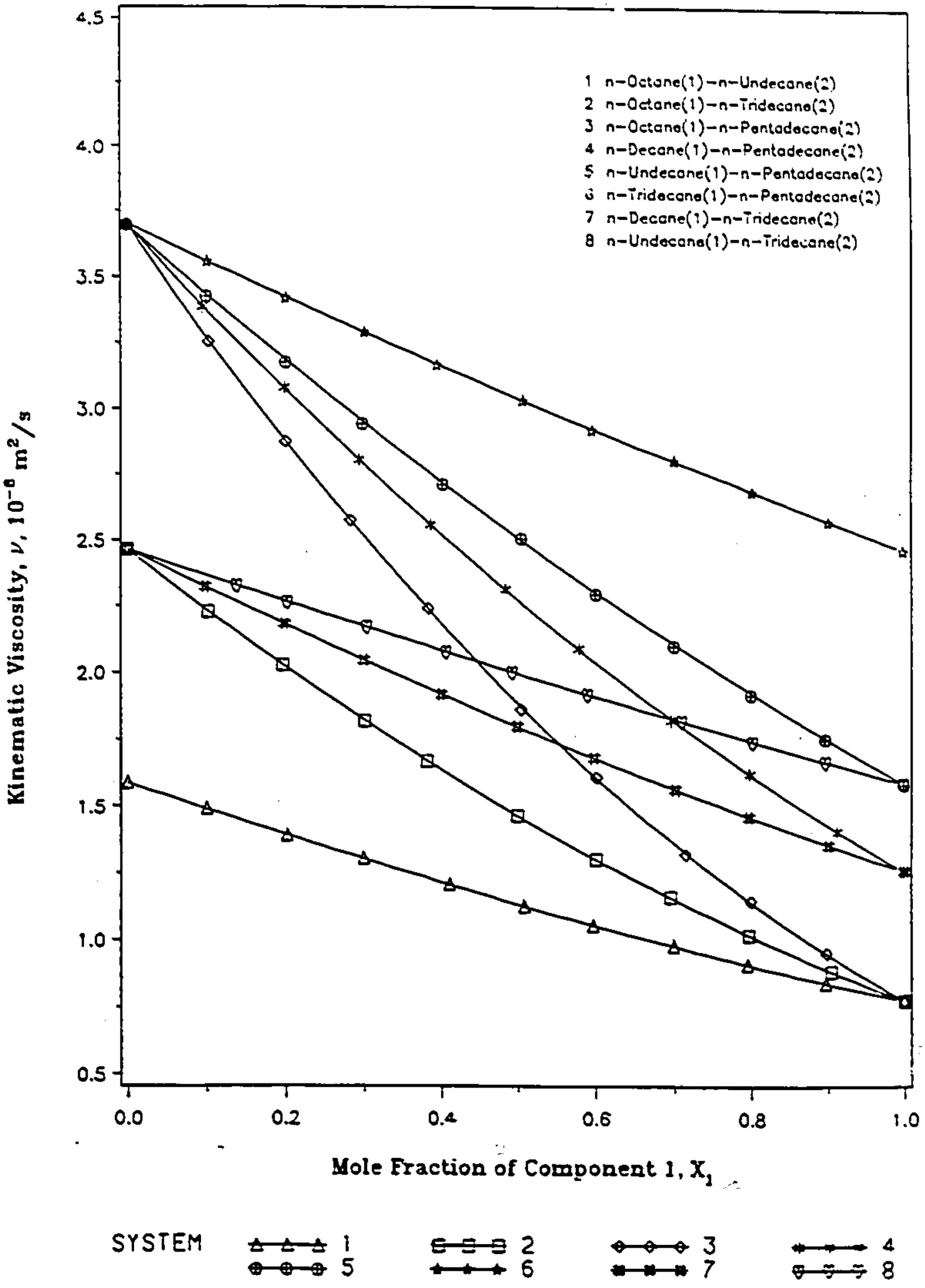


Figure 4.5. Viscosity vs. Composition for Binary n-Alkane Systems at 293.15 K

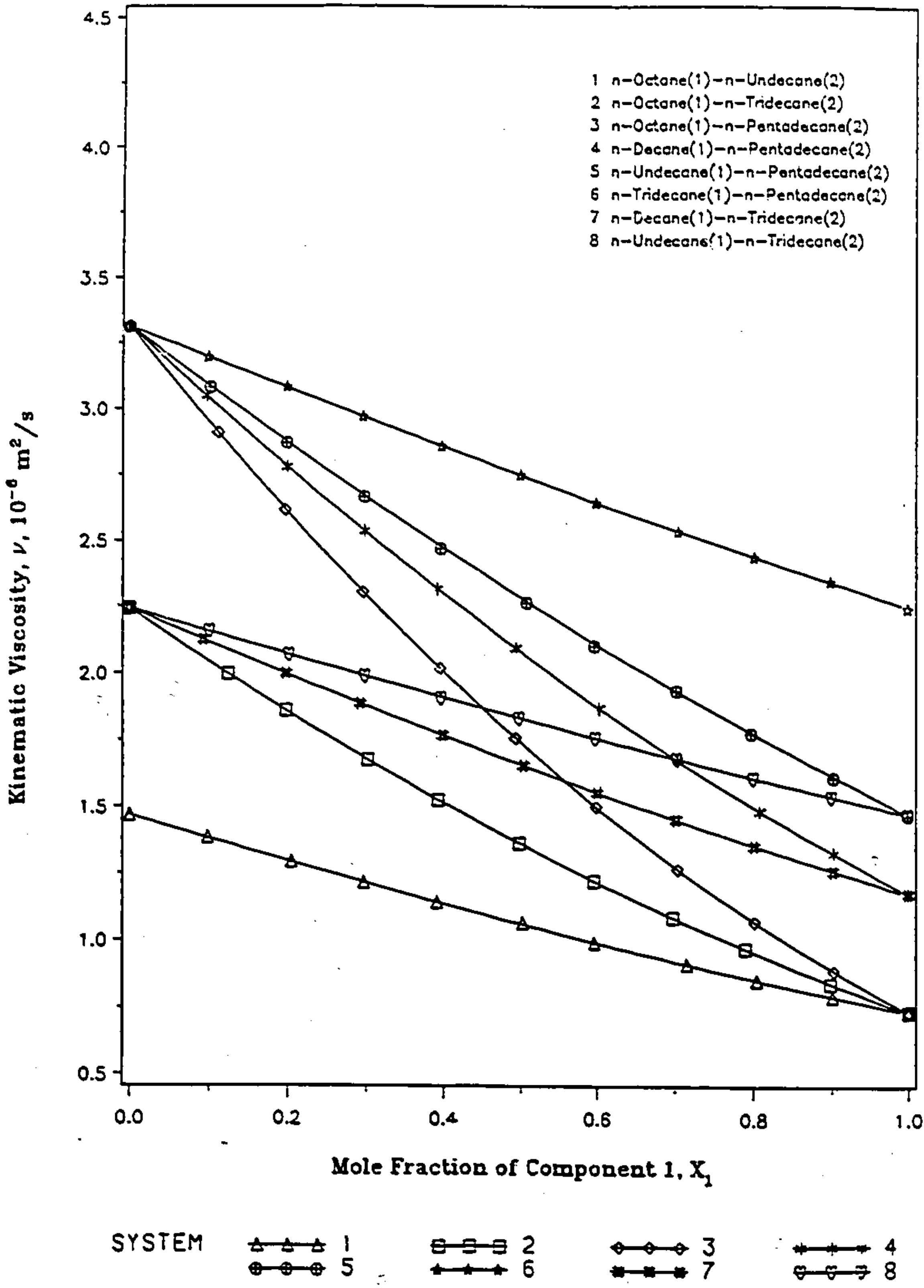


Figure 4.6. Viscosity vs. Composition
for Binary n-Alkane Systems at 298.15 K

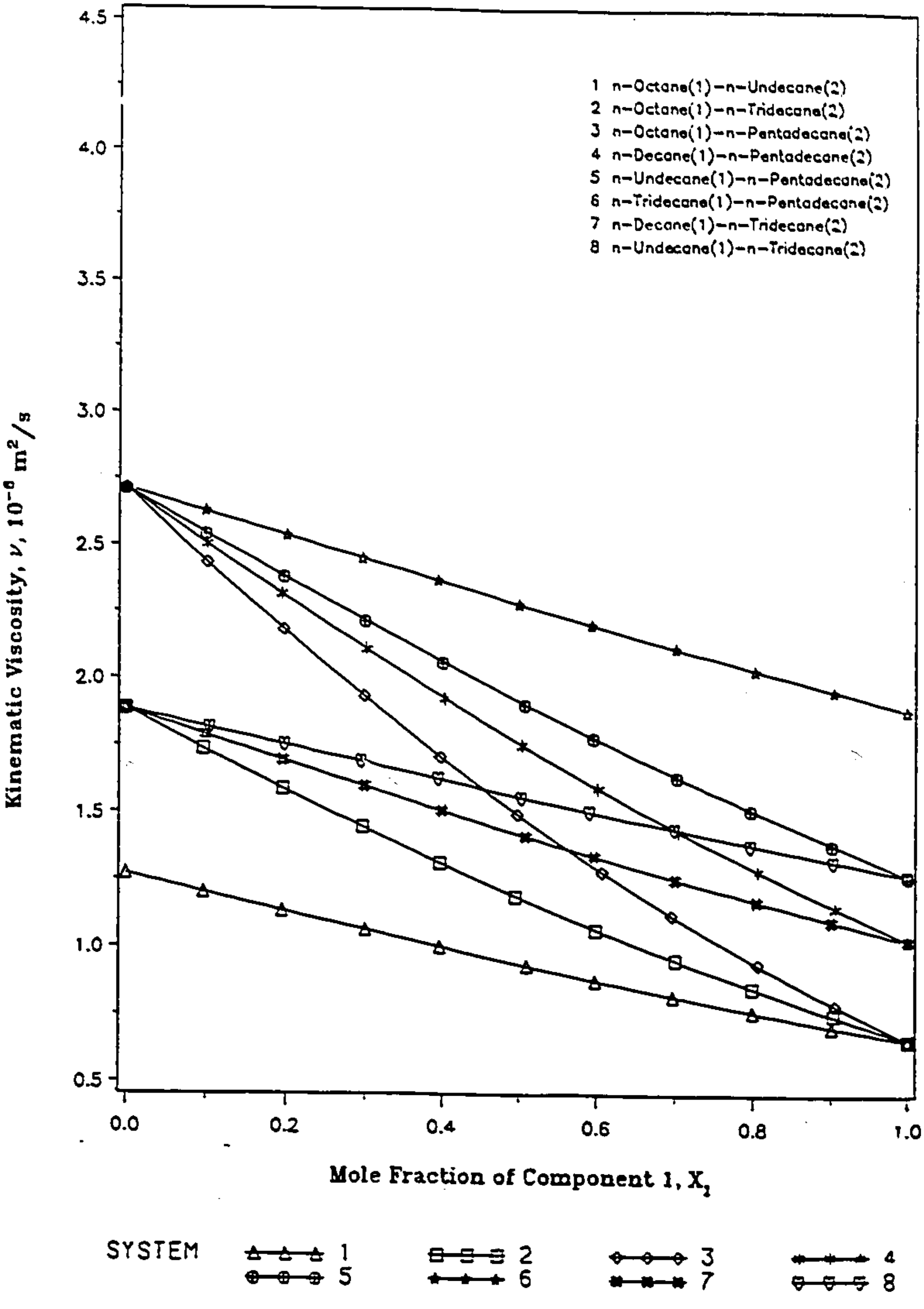


Figure 4.7. Viscosity vs. Composition for Binary n-Alkane Systems at 308.15 K

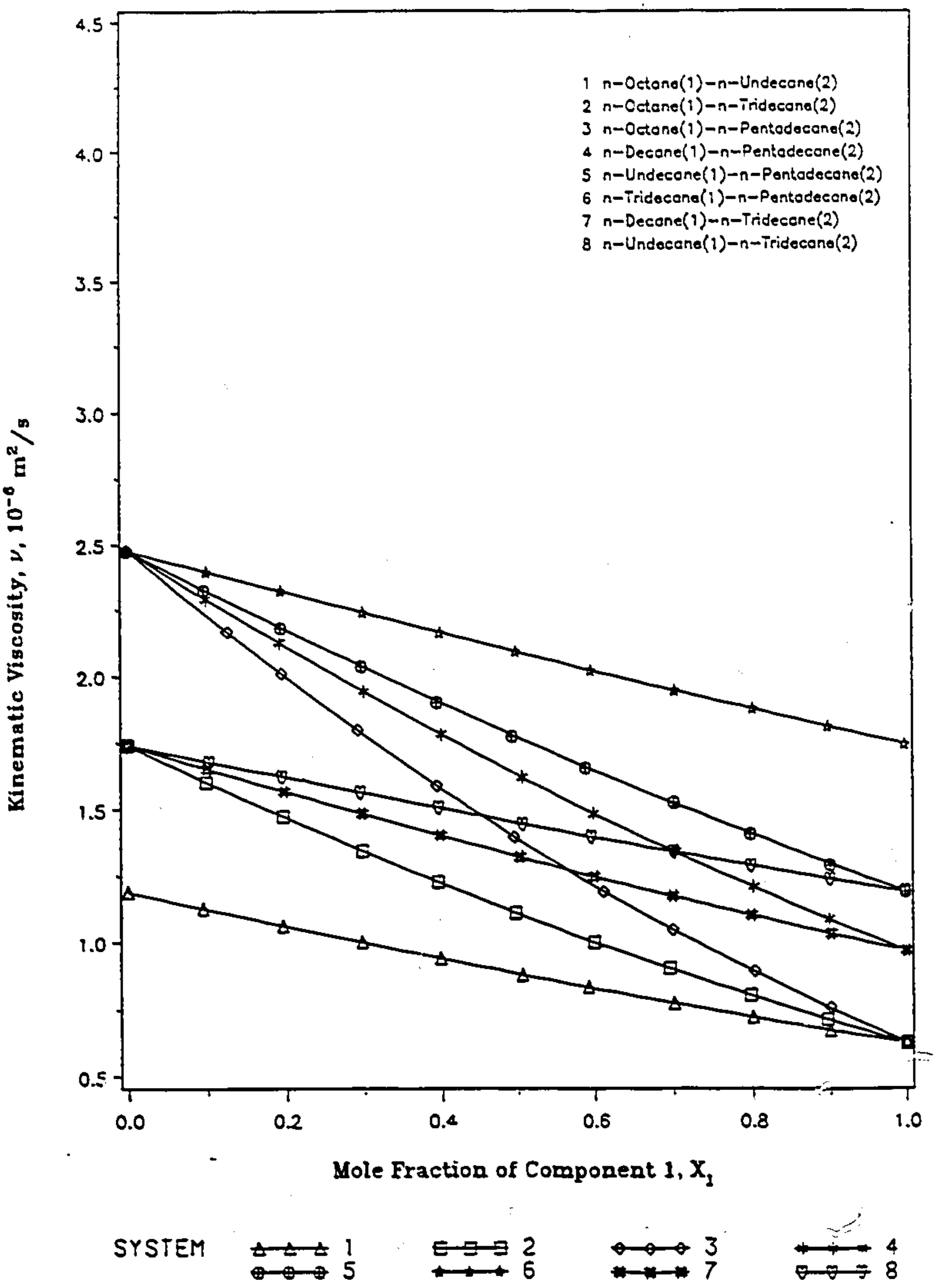


Figure 4.8. Viscosity vs. Composition for Binary n-Alkane Systems at 313.15 K

Table 4.12

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Octane(1)-n-Undecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
Temperature = 293.15 K				
0.0000	1.5869	1.1744	0.0000	0.0000
0.1007	1.4910	1.0991	-0.0118	34.4653
0.2018	1.3901	1.0204	-0.0266	48.9088
0.2995	1.3044	0.9534	-0.0320	70.9063
0.4108	1.2073	0.8779	-0.0373	81.6587
0.5076	1.1255	0.8145	-0.0396	81.7471
0.5964	1.0550	0.7599	-0.0381	79.0857
0.6995	0.9777	0.7001	-0.0328	71.0946
0.7954	0.9090	0.6472	-0.0252	56.0375
0.8978	0.8387	0.5933	-0.0145	30.4807
1.0000	0.7734	0.5433	0.0000	0.0000
Temperature = 298.15 K				
0.0000	1.4679	1.0809	0.0000	0.0000
0.0992	1.3803	1.0124	-0.0119	28.9612
0.2059	1.2890	0.9412	-0.0223	52.4521
0.2978	1.2141	0.8829	-0.0281	68.5674
0.3916	1.1402	0.8255	-0.0320	78.9547
0.5019	1.0616	0.7644	-0.0302	94.6327
0.5959	0.9901	0.7093	-0.0317	83.7418
0.7154	0.9098	0.6474	-0.0255	76.6877
0.8041	0.8511	0.6023	-0.0199	59.2225
0.9011	0.7905	0.5559	-0.0110	35.1805
1.0000	0.7309	0.5105	0.0000	0.0000

Table 4.12 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Octane(1)-n-Undecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
		Temperature = 308.15 K		
0.0000	1.2719	0.9273	0.0000	0.0000
0.0969	1.2019	0.8729	-0.0086	28.6132
0.1954	1.1339	0.8200	-0.0149	54.0281
0.3001	1.0643	0.7660	-0.0194	75.1502
0.3970	1.0022	0.7179	-0.0217	88.7324
0.5094	0.9337	0.6650	-0.0215	99.0137
0.5981	0.8782	0.6224	-0.0221	91.0535
0.6968	0.8212	0.5787	-0.0192	82.5191
0.7992	0.7639	0.5349	-0.0146	63.8305
0.8998	0.7101	0.4939	-0.0080	37.3399
1.0000	0.6583	0.4545	0.0000	0.0000
		Temperature = 313.15 K		
0.0000	1.1899	0.8633	0.0000	0.0000
0.0953	1.1269	0.8144	-0.0076	27.9552
0.1962	1.0635	0.7652	-0.0130	54.7831
0.2979	1.0019	0.7175	-0.0166	76.2628
0.3995	0.9428	0.6718	-0.0183	92.3124
0.5061	0.8810	0.6243	-0.0195	96.0964
0.5914	0.8349	0.5888	-0.0180	98.3707
0.7010	0.7742	0.5424	-0.0168	81.7004
0.8001	0.7236	0.5038	-0.0125	65.3852
0.8999	0.6735	0.4658	-0.0073	36.3218
1.0000	0.6258	0.4296	0.0000	0.0000

Table 4.13

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Octane(1)-n-Tridecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
		Temperature = 293.15 K		
0.0000	2.4638	1.8631	0.0000	0.0000
0.1023	2.2282	1.6767	-0.0513	66.5319
0.1966	2.0280	1.5187	-0.0850	120.6914
0.3011	1.8193	1.3544	-0.1113	166.0044
0.3813	1.6692	1.2367	-0.1231	190.7308
0.4993	1.4935	1.0760	-0.1281	209.6532
0.6002	1.3010	0.9495	-0.1214	207.1383
0.6949	1.1597	0.8400	-0.1060	188.3204
0.7976	1.0178	0.7304	-0.0800	147.0541
0.9047	0.8829	0.6269	-0.0422	81.0164
1.0000	0.7734	0.5433	0.0000	0.0000
		Temperature = 298.15 K		
0.0000	2.2427	1.6879	0.0000	0.0000
0.1246	1.9946	1.4921	-0.0491	83.1579
0.1988	1.8564	1.3833	-0.0705	124.8681
0.3028	1.6727	1.2391	-0.0923	170.5899
0.3936	1.5229	1.1218	-0.1026	199.4045
0.4971	1.3619	0.9963	-0.1064	215.3638
0.5956	1.2186	0.8850	-0.1016	213.1251
0.6970	1.0845	0.7811	-0.0861	199.4471
0.7898	0.9689	0.6921	-0.0659	166.0345
0.8988	0.8392	0.5929	-0.0367	90.5990
1.0000	0.7309	0.5105	0.0000	0.0000

Table 4.13 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Octane(1)-n-Tridecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^{*}G^{E}$ J/mol
Temperature = 308.15 K				
0.0000	1.8856	1.4059	0.0000	0.0000
0.0968	1.7350	1.2874	-0.0264	70.3199
0.1961	1.5884	1.1724	-0.0469	131.1525
0.2969	1.4459	1.0610	-0.0624	177.3084
0.3966	1.3133	0.9577	-0.0709	209.9969
0.4942	1.1903	0.8621	-0.0736	226.3498
0.5983	1.0672	0.7669	-0.0698	226.9019
0.7006	0.9553	0.6806	-0.0588	211.9393
0.7985	0.8532	0.6024	-0.0439	172.8641
0.9019	0.7556	0.5278	-0.0202	118.2215
1.0000	0.6583	0.4546	0.0000	0.0000
Temperature = 313.15 K				
0.0000	1.7410	1.2920	0.0000	0.0000
0.0995	1.6033	1.1840	-0.0222	74.0878
0.1969	1.4739	1.0827	-0.0395	133.6083
0.2992	1.3443	0.9816	-0.0524	182.1096
0.3967	1.2273	0.8906	-0.0593	214.9710
0.4967	1.1134	0.8023	-0.0614	233.0765
0.5993	1.0028	0.7169	-0.0583	233.4713
0.6949	0.9069	0.6431	-0.0496	219.9543
0.7971	0.8058	0.5659	-0.0386	170.4821
0.8964	0.7143	0.4964	-0.0225	99.8355
1.0000	0.6258	0.4296	0.0000	0.0000

Table 4.14

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Octane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
		Temperature = 293.15 K		
0.0000	3.6979	2.8413	0.0000	0.0000
0.1032	3.2552	2.4880	-0.1161	119.6919
0.2005	2.8738	2.1846	-0.1959	216.2265
0.2833	2.5750	1.9475	-0.2428	284.5688
0.3826	2.2409	1.6833	-0.2787	342.4358
0.5038	1.8610	1.3850	-0.2986	363.3479
0.6008	1.6085	1.1871	-0.2736	377.7588
0.7150	1.3199	0.9631	-0.2352	319.0088
0.7994	1.1456	0.8279	-0.1764	276.9753
0.8985	0.9507	0.6782	-0.0983	166.3823
1.0000	0.7734	0.5433	0.0000	0.0000
		Temperature = 298.15 K		
0.0000	3.3136	2.5343	0.0000	0.0000
0.1114	2.9072	2.2107	-0.0981	133.3051
0.1978	2.6134	1.9775	-0.1565	219.1627
0.2962	2.3036	1.7324	-0.2025	299.0771
0.3951	2.0160	1.5057	-0.2290	356.2383
0.4927	1.7543	1.3004	-0.2367	386.5989
0.5978	1.4971	1.0996	-0.2248	387.9487
0.7035	1.2638	0.9185	-0.1920	353.3140
0.8008	1.0694	0.7686	-0.1450	282.4937
0.9026	0.8872	0.6291	-0.0784	164.9750
1.0000	0.7309	0.5104	0.0000	0.0000

Table 4.14 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Octane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m²/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	Δ^*G^E J/mol
		Temperature = 308.15 K		
0.0000	2.7124	2.0559	0.0000	0.0000
0.1015	2.4322	1.8337	-0.0597	127.0478
0.1973	2.1811	1.6353	-0.1047	225.7658
0.2986	1.9332	1.4401	-0.1377	309.6018
0.3975	1.7077	1.2631	-0.1562	368.1792
0.4973	1.4956	1.0975	-0.1621	400.0127
0.6065	1.2817	0.9313	-0.1534	400.6040
0.6944	1.1233	0.8089	-0.1351	372.3059
0.8053	0.9424	0.6698	-0.0965	299.8799
0.9040	0.7907	0.5545	-0.0539	171.8228
1.0000	0.6583	0.4546	0.0000	0.0000
		Temperature = 313.15 K		
0.0000	2.4757	1.8680	0.0000	0.0000
0.1274	2.1672	1.6241	-0.0606	157.4021
0.1958	2.0098	1.5000	-0.0863	227.4998
0.2936	1.7974	1.3331	-0.1126	312.0928
0.3949	1.5896	1.1704	-0.1296	373.5203
0.4956	1.3967	1.0201	-0.1350	407.2629
0.6113	1.1921	0.8614	-0.1273	408.8123
0.6990	1.0493	0.7514	-0.1112	380.8423
0.8018	0.8949	0.6329	-0.0817	311.0525
0.9006	0.7599	0.5301	-0.0424	202.5082
1.0000	0.6258	0.4296	0.0000	0.0000

Table 4.15

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Decane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
Temperature = 293.15 K				
0.0000	3.6979	2.8411	0.0000	0.0000
0.0958	3.3867	2.5930	-0.0639	52.6719
0.1989	3.0790	2.3481	-0.1106	104.8836
0.2947	2.8056	2.1312	-0.1433	139.5744
0.3865	2.5567	1.9343	-0.1636	160.7630
0.4839	2.3122	1.7413	-0.1694	175.2693
0.5780	2.0907	1.5670	-0.1628	177.1496
0.6958	1.8228	1.3573	-0.1459	147.4353
0.7980	1.6222	1.2005	-0.1062	122.4323
0.9126	1.4071	1.0334	-0.0530	60.2131
1.0000	1.2583	0.9183	0.0000	0.0000
Temperature = 298.15 K				
0.0000	3.3136	2.5343	0.0000	0.0000
0.0973	3.0466	2.3220	-0.0485	58.2415
0.1999	2.7764	2.1076	-0.0901	106.0720
0.2983	2.5343	1.9159	-0.1161	143.5033
0.3925	2.3145	1.7425	-0.1309	167.9470
0.4933	2.0913	1.5668	-0.1369	179.8614
0.6025	1.8664	1.3904	-0.1294	178.6227
0.7033	1.6720	1.2385	-0.1116	160.7179
0.8079	1.4831	1.0915	-0.0825	122.9532
0.9014	1.3258	0.9696	-0.0469	72.2716
1.0000	1.1715	0.8505	0.0000	0.0000

Table 4.15 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Decane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	Δ^*G^E J/mol
		Temperature = 308.15 K		
0.0000	2.7124	2.0559	0.0000	0.0000
0.1015	2.5017	1.8891	-0.0333	61.3433
0.1942	2.3139	1.7409	-0.0596	103.9613
0.3018	2.1093	1.5797	-0.0792	144.8617
0.4028	1.9252	1.4351	-0.0910	168.2878
0.5033	1.7522	1.2997	-0.0942	179.4218
0.6010	1.5943	1.1764	-0.0890	178.9185
0.7040	1.4343	1.0521	-0.0778	158.0065
0.8051	1.2880	0.9388	-0.0582	122.9040
0.9044	1.1524	0.8342	-0.0322	68.9955
1.0000	1.0306	0.7406	0.0000	0.0000
		Temperature = 313.15 K		
0.0000	2.4757	1.8682	0.0000	0.0000
0.0999	2.2918	1.7229	-0.0278	60.0494
0.1932	2.1265	1.5927	-0.0483	106.1795
0.3013	1.9424	1.4482	-0.0658	146.1495
0.4013	1.7807	1.3215	-0.0749	171.2866
0.5059	1.6200	1.1959	-0.0774	183.9942
0.5986	1.4840	1.0901	-0.0743	181.4641
0.7028	1.3387	0.9773	-0.0645	162.4201
0.8009	1.2102	0.8780	-0.0485	129.5562
0.8988	1.0876	0.7836	-0.0278	75.8249
1.0000	0.9685	0.6924	0.0000	0.0000

Table 4.16

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Undecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^{*}G^{E}$ J/mol
		Temperature = 293.15 K		
0.0000	3.6979	2.8409	0.0000	0.0000
0.1013	3.4282	2.6262	-0.0459	33.9191
0.2015	3.1790	2.4279	-0.0772	64.2913
0.3004	2.9445	2.2418	-0.0985	87.1884
0.4036	2.7150	2.0599	-0.1084	106.0899
0.5046	2.5083	1.8962	-0.1038	122.9530
0.6010	2.2975	1.7305	-0.1087	107.1521
0.7004	2.1041	1.5785	-0.0951	94.8145
0.8000	1.9213	1.4352	-0.0724	73.1493
0.8974	1.7536	1.3042	-0.0411	43.3568
1.0000	1.5869	1.1743	0.0000	0.0000
		Temperature = 298.15 K		
0.0000	3.3136	2.5343	0.0000	0.0000
0.1026	3.0828	2.3509	-0.0343	38.0129
0.2004	2.8711	2.1829	-0.0601	66.6975
0.2994	2.6667	2.0211	-0.0781	89.2972
0.3983	2.4696	1.8655	-0.0900	102.4267
0.5079	2.2661	1.7050	-0.0911	112.2657
0.5973	2.1047	1.5781	-0.0881	108.9702
0.7022	1.9357	1.4452	-0.0685	110.0293
0.7975	1.7752	1.3200	-0.0553	82.4534
0.9019	1.6103	1.1916	-0.0319	42.7202
1.0000	1.4679	1.0809	0.0000	0.0000

Table 4.16 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Undecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	Δ^*G^E J/mol
Temperature = 308.15 K				
0.0000	2.7124	2.0559	0.0000	0.0000
0.1005	2.5412	1.9204	-0.0220	38.0027
0.1976	2.3797	1.7930	-0.0399	66.0710
0.3007	2.2167	1.6646	-0.0519	90.6552
0.4013	2.0612	1.5424	-0.0606	103.5191
0.5074	1.9066	1.4211	-0.0621	111.4035
0.5964	1.7809	1.3228	-0.0600	108.7745
0.7036	1.6362	1.2100	-0.0518	96.3065
0.7972	1.5154	1.1159	-0.0402	75.9663
0.9014	1.3864	1.0159	-0.0226	41.1635
1.0000	1.2719	0.9273	0.0000	0.0000
Temperature = 313.15 K				
0.0000	2.4757	1.8681	0.0000	0.0000
0.0989	2.3266	1.7504	-0.0183	36.9855
0.1959	2.1843	1.6384	-0.0329	65.7502
0.2996	2.0378	1.5232	-0.0438	89.3006
0.3974	1.9035	1.4180	-0.0508	102.3707
0.4945	1.7769	1.3189	-0.0523	110.2465
0.5890	1.6573	1.2256	-0.0507	108.7046
0.7019	1.5275	1.1242	-0.0386	108.2701
0.7983	1.4083	1.0320	-0.0339	74.9146
0.9013	1.2931	0.9429	-0.0196	40.1046
1.0000	1.1899	0.8633	0.0000	0.0000

Table 4.17

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Tridecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	Δ^*G^E J/mol
Temperature = 293.15 K				
0.0000	3.6979	2.8409	0.0000	0.0000
0.1030	3.5558	2.7278	-0.0123	8.6070
0.2020	3.4179	2.6183	-0.0251	11.8402
0.3023	3.2910	2.5173	-0.0279	20.1172
0.3967	3.1662	2.4184	-0.0345	20.0521
0.5078	3.0301	2.3104	-0.0339	23.2388
0.5972	2.9200	2.2233	-0.0336	21.3281
0.7003	2.8039	2.1313	-0.0247	23.7757
0.8019	2.6852	2.0375	-0.0192	17.6673
0.9018	2.5723	1.9484	-0.0105	10.0964
1.0000	2.4638	1.8629	0.0000	0.0000
Temperature = 298.15 K				
0.0000	3.3136	2.5343	0.0000	0.0000
0.1000	3.1939	2.4393	-0.0104	7.7160
0.2001	3.0794	2.3485	-0.0165	15.7891
0.2975	2.9679	2.2601	-0.0224	19.8456
0.3984	2.8562	2.1717	-0.0254	23.1860
0.5000	2.7464	2.0849	-0.0262	24.6351
0.5985	2.6430	2.0031	-0.0246	24.6327
0.7037	2.5331	1.9166	-0.0221	20.4059
0.8004	2.4362	1.8401	-0.0167	16.0940
0.8983	2.3454	1.7684	-0.0055	14.9426
1.0000	2.2427	1.6879	0.0000	0.0000

Table 4.17 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Tridecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^{*}G^{E}$ J/mol
		Temperature = 298.15 K		
0.0000	2.7124	2.0560	0.0000	0.0000
0.1004	2.6242	1.9862	-0.0046	11.0631
0.2017	2.5339	1.9150	-0.0099	17.5036
0.2979	2.4497	1.8486	-0.0137	21.7793
0.3944	2.3687	1.7848	-0.0149	26.3287
0.4994	2.2800	1.7151	-0.0163	26.7050
0.5935	2.2037	1.6551	-0.0151	26.9859
0.7015	2.1172	1.5872	-0.0128	24.2299
0.8030	2.0367	1.5241	-0.0099	18.1657
0.9036	1.9595	1.4637	-0.0050	11.0208
1.0000	1.8856	1.4060	0.0000	0.0000
		Temperature = 313.15 K		
0.0000	2.4757	1.8680	0.0000	0.0000
0.1024	2.3948	1.8043	-0.0047	9.6734
0.1963	2.3204	1.7458	-0.0092	15.2675
0.3028	2.2383	1.6812	-0.0124	20.5039
0.4019	2.1637	1.6227	-0.0138	23.8956
0.4997	2.0916	1.5660	-0.0142	25.6138
0.5970	2.0198	1.5098	-0.0143	23.6647
0.7040	1.9446	1.4510	-0.0115	22.1700
0.8017	1.8756	1.3970	-0.0092	16.3897
0.8985	1.8095	1.3454	-0.0050	9.9090
1.0000	1.7410	1.2920	0.0000	0.0000

Table 4.18

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Decane(1)-n-Tridecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m²/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
		Temperature = 293.15K		
0.0000	2.4638	1.8629	0.0000	0.0000
0.0987	2.3210	1.7501	-0.0196	22.9110
0.1990	2.1819	1.6404	-0.0345	42.1194
0.2995	2.0445	1.5323	-0.0477	52.3604
0.3997	1.9178	1.4327	-0.0526	63.2314
0.4986	1.7965	1.3375	-0.0544	67.1014
0.5974	1.6808	1.2470	-0.0515	66.2251
0.7015	1.5626	1.1548	-0.0454	56.7302
0.7973	1.4602	1.0749	-0.0347	44.6320
0.9014	1.3537	0.9922	-0.0191	24.4301
1.0000	1.2583	0.9182	0.0000	0.0000
		Temperature = 298.15K		
0.0000	2.2427	1.6879	0.0000	0.0000
0.0939	2.1236	1.5940	-0.0152	22.4933
0.1985	1.9948	1.4927	-0.0290	41.7243
0.2931	1.8837	1.4054	-0.0371	56.0028
0.3988	1.7638	1.3114	-0.0426	66.0757
0.5031	1.6496	1.2220	-0.0446	69.2435
0.5995	1.5500	1.1442	-0.0416	69.5650
0.7013	1.4507	1.0668	-0.0339	67.0544
0.8001	1.3517	0.9901	-0.0278	46.8778
0.9011	1.2587	0.9179	-0.0154	26.7111
1.0000	1.1715	0.8505	0.0000	0.0000

Table 4.18 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow
of the System n-Decane(1)-n-Tridecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^* G^E$ J/mol
Temperature = 308.15 K				
0.0000	1.8856	1.4059	0.0000	0.0000
0.0991	1.7880	1.3293	-0.0106	24.3741
0.1956	1.6936	1.2555	-0.0203	40.4240
0.2991	1.5979	1.1806	-0.0263	56.1464
0.3993	1.5082	1.1105	-0.0297	66.0539
0.5073	1.4138	1.0371	-0.0313	68.8514
0.5962	1.3397	0.9795	-0.0298	68.0500
0.6991	1.2561	0.9147	-0.0261	59.9164
0.8037	1.1744	0.8515	-0.0197	45.1053
0.8994	1.1029	0.7963	-0.0112	26.3457
1.0000	1.0306	0.7406	0.0000	0.0000
Temperature = 313.15 K				
0.0000	1.7410	1.2920	0.0000	0.0000
0.1001	1.6520	1.2223	-0.0097	23.5728
0.1979	1.5678	1.1565	-0.0168	42.5076
0.2988	1.4837	1.0909	-0.0219	57.5242
0.3996	1.4017	1.0271	-0.0253	66.3163
0.5025	1.3212	0.9645	-0.0261	70.6887
0.5994	1.2480	0.9078	-0.0248	69.7427
0.6995	1.1743	0.8508	-0.0217	61.7899
0.7980	1.1048	0.7971	-0.0163	49.1940
0.9013	1.0341	0.7427	-0.0088	28.2601
1.0000	0.9685	0.6923	0.0000	0.0000

Table 4.19

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Undecane(1)-n-Tridecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^{*}G^{E}$ J/mol
Temperature = 298.15 K				
0.0000	2.4638	1.8631	0.0000	0.0000
0.1389	2.3260	1.7543	-0.0131	12.4724
0.2035	2.2639	1.7054	-0.0176	17.1745
0.3044	2.1703	1.6316	-0.0219	24.1174
0.4076	2.0762	1.5575	-0.0249	27.7875
0.4924	2.0008	1.4983	-0.0257	28.9078
0.5897	1.9178	1.4331	-0.0239	29.7911
0.7108	1.8180	1.3548	-0.0187	28.2593
0.8027	1.7403	1.2941	-0.0161	18.7900
0.8980	1.6663	1.2362	-0.0084	12.8103
1.0000	1.5869	1.1744	0.0000	0.0000
Temperature = 298.15 K				
0.0000	2.2427	1.6877	0.0000	0.0000
0.1023	2.1552	1.6187	-0.0069	11.8034
0.2031	2.0666	1.5491	-0.0153	15.9711
0.2991	1.9875	1.4870	-0.0192	21.7227
0.3981	1.9064	1.4234	-0.0227	23.5406
0.4981	1.8277	1.3617	-0.0237	24.5405
0.5976	1.7540	1.3039	-0.0211	26.8218
0.7028	1.6771	1.2437	-0.0174	25.2261
0.8000	1.6066	1.1888	-0.0133	19.2641
0.9004	1.5359	1.1336	-0.0075	10.7730
1.0000	1.4679	1.0807	0.0000	0.0000

Table 4.19 (cont'd)

Viscosity, Excess Viscosity and Excess Energy of Viscous Flow of the System n-Undecane(1)-n-Tridecane(2)

Mole Fraction x_1	Kinematic Viscosity 10^{-6} m²/s	Absolute Viscosity 10^{-3} Pa.s	Excess Viscosity 10^{-3} Pa.s	$\Delta^{*}G^{E}$ J/mol
		Temperature = 308.15 K		
0.0000	1.8856	1.4060	0.0000	0.0000
0.1051	1.8138	1.3497	-0.0060	9.7022
0.1986	1.7519	1.3012	-0.0097	17.2886
0.2953	1.6892	1.2522	-0.0125	23.1867
0.3954	1.6267	1.2033	-0.0135	28.7158
0.5039	1.5587	1.1502	-0.0146	29.2435
0.5908	1.5059	1.1091	-0.0142	28.4079
0.6994	1.4420	1.0594	-0.0119	25.8523
0.7972	1.3841	1.0144	-0.0100	17.8481
0.9027	1.3244	0.9681	-0.0058	8.6831
1.0000	1.2719	0.9274	0.0000	0.0000
		Temperature = 313.15 K		
0.0000	1.7410	1.2921	0.0000	0.0000
0.1040	1.6777	1.2425	-0.0050	9.7926
0.1964	1.6220	1.1991	-0.0088	15.6945
0.2972	1.5636	1.1535	-0.0112	21.9366
0.3982	1.5069	1.1093	-0.0121	27.0067
0.5078	1.4455	1.0615	-0.0129	27.7690
0.5968	1.3965	1.0234	-0.0128	25.9241
0.7018	1.3404	0.9799	-0.0113	22.1849
0.7985	1.2900	0.9408	-0.0089	16.5313
0.9009	1.2378	0.9004	-0.0054	7.8660
1.0000	1.1899	0.8633	0.0000	0.0000

Table 4.20

Densities and Viscosities of Ternary System n-Octane(1)-n-Undecane(2)-n-Tridecane(3)

Mole Fraction x_1	Mole Fraction , x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.7734	0.5433	0.7024	1.0000	0.0000	0.7309	0.5104	0.6983
0.0000	1.0000	1.5869	1.1744	0.7400	0.0000	1.0000	1.4679	1.0807	0.7362
0.0000	0.0000	2.4638	1.8631	0.7562	0.0000	0.0000	2.2427	1.6877	0.7525
0.1043	0.1018	2.1314	1.6005	0.7509	0.1053	0.1002	1.9533	1.4594	0.7472
0.1087	0.7938	1.5495	1.1448	0.7388	0.1052	0.7940	1.4407	1.0592	0.7352
0.2058	0.2046	1.8339	1.3664	0.7451	0.2045	0.2002	1.6939	1.2560	0.7415
0.2096	0.3973	1.6725	1.2402	0.7415	0.2062	0.3907	1.5556	1.1480	0.7380
0.2055	0.5981	1.5269	1.1267	0.7379	0.1979	0.6038	1.4234	1.0453	0.7343
0.3124	0.2921	1.5684	1.1585	0.7386	0.2998	0.2950	1.4715	1.0821	0.7354
0.3067	0.3977	1.5008	1.1059	0.7369	0.3058	0.3990	1.3928	1.0210	0.7331
0.4072	0.1998	1.4735	1.0841	0.7358	0.3981	0.1976	1.3828	1.0128	0.7325
0.3990	0.3049	1.4124	1.0368	0.7340	0.4031	0.3008	1.3095	0.9561	0.7301
0.4007	0.4024	1.3426	0.9826	0.7319	0.4020	0.4011	1.2479	0.9084	0.7280
0.6048	0.2009	1.1648	0.8443	0.7248	0.5925	0.2097	1.1013	0.7945	0.7215
0.7999	0.1002	0.9586	0.6851	0.7147	0.7978	0.1025	0.9046	0.6430	0.7108

Table 4.20 (cont'd)

Densities and Viscosities of Ternary System n-Octane(1)-n-Undecane(2)-n-Tridecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.6583	0.4545	0.6905	1.0000	0.0000	0.6258	0.4295	0.6864
0.0000	1.0000	1.2719	0.9273	0.7291	0.0000	1.0000	1.1899	0.8632	0.7254
0.0000	0.0000	1.8856	1.4058	0.7455	0.0000	0.0000	1.7410	1.2919	0.7420
0.1062	0.0985	1.6564	1.2259	0.7401	0.0964	0.0975	1.5502	1.1425	0.7370
0.1048	0.7952	1.2489	0.9091	0.7279	0.1026	0.8003	1.1688	0.8466	0.7243
0.2060	0.1951	1.4543	1.0679	0.7343	0.1981	0.1988	1.3634	0.9968	0.7311
0.2060	0.3903	1.3422	0.9809	0.7308	0.1993	0.3896	1.2619	0.9181	0.7275
0.2033	0.6018	1.2294	0.8937	0.7269	0.2063	0.5973	1.1509	0.8323	0.7232
0.3072	0.2969	1.2636	0.9196	0.7278	0.2961	0.2976	1.1955	0.8663	0.7246
0.3030	0.3960	1.2155	0.8824	0.7260	0.3028	0.4014	1.1366	0.8209	0.7222
0.3973	0.1979	1.2077	0.8759	0.7252	0.4045	0.1996	1.1204	0.8080	0.7212
0.3922	0.3072	1.1533	0.8341	0.7232	0.4032	0.2947	1.0774	0.7749	0.7192
0.4007	0.4006	1.0962	0.7901	0.7207	0.4016	0.4002	1.0297	0.7382	0.7169
0.6069	0.1954	0.9648	0.6883	0.7135	0.6117	0.1972	0.9056	0.6424	0.7093
0.7962	0.1023	0.8132	0.5719	0.7033	0.8004	0.1002	0.7619	0.5327	0.6992

Table 4.21

Densities and Viscosities of Ternary System n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.7734	0.5433	0.7025	1.0000	0.0000	0.7309	0.5104	0.6983
0.0000	1.0000	1.5869	1.1745	0.7401	0.0000	1.0000	1.4679	1.0807	0.7362
0.0000	0.0000	3.6979	2.8414	0.7684	0.0000	0.0000	3.3136	2.5339	0.7647
0.1038	0.1022	3.0058	2.2900	0.7619	0.1108	0.0995	2.7039	2.0494	0.7580
0.1091	0.7909	1.6329	1.2096	0.7408	0.1096	0.7909	1.5097	1.1124	0.7369
0.2082	0.2010	2.4104	1.8183	0.7544	0.2131	0.2016	2.1856	1.6402	0.7504
0.2019	0.3999	2.0444	1.5306	0.7487	0.2068	0.3952	1.8717	1.3939	0.7447
0.2111	0.5953	1.6758	1.2424	0.7414	0.2087	0.5964	1.5557	1.1475	0.7376
0.3068	0.2995	1.9143	1.4279	0.7459	0.3025	0.2971	1.7775	1.3196	0.7424
0.3029	0.3988	1.7509	1.3003	0.7426	0.3083	0.3947	1.6122	1.1908	0.7386
0.4042	0.1993	1.8103	1.3459	0.7435	0.4039	0.2016	1.6674	1.2331	0.7395
0.4007	0.3008	1.6485	1.2198	0.7400	0.4023	0.2988	1.5272	1.1241	0.7361
0.4051	0.3987	1.4877	1.0948	0.7359	0.4027	0.4017	1.3826	1.0121	0.7320
0.6019	0.2054	1.3013	0.9492	0.7294	0.5903	0.2059	1.2367	0.8983	0.7264
0.8005	0.1016	1.0172	0.7297	0.7174	0.8034	0.1012	0.9554	0.6813	0.7131

Table 4.21 (cont'd)

Densities and Viscosities of Ternary System n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.6583	0.4545	0.6905	1.0000	0.0000	0.6258	0.4296	0.6864
0.0000	1.0000	1.2719	0.9273	0.7291	0.0000	1.0000	1.1899	0.8632	0.7254
0.0000	0.0000	2.7124	2.0557	0.7579	0.0000	0.0000	2.4757	1.8679	0.7545
0.1208	0.0986	2.2240	1.6693	0.7506	0.1018	0.0951	2.0918	1.5649	0.7481
0.1086	0.7899	1.3078	0.9545	0.7298	0.1030	0.7965	1.2251	0.8898	0.7263
0.2077	0.2035	1.8547	1.3791	0.7436	0.2004	0.1964	1.7406	1.2892	0.7406
0.2102	0.3937	1.5907	1.1732	0.7375	0.1992	0.3971	1.4970	1.0995	0.7345
0.2025	0.6004	1.3505	0.9869	0.7308	0.2014	0.5952	1.2688	0.9229	0.7274
0.3060	0.3009	1.5088	1.1089	0.7350	0.2998	0.2988	1.4198	1.0391	0.7319
0.3043	0.4040	1.3862	1.0139	0.7314	0.2939	0.4029	1.3138	0.9572	0.7285
0.3956	0.2061	1.4455	1.0593	0.7328	0.4021	0.1961	1.3473	0.9824	0.7291
0.3983	0.3050	1.3218	0.9635	0.7290	0.4011	0.2953	1.2411	0.9004	0.7255
0.4032	0.3977	1.2068	0.8748	0.7249	0.3947	0.4066	1.1374	0.8206	0.7215
0.6003	0.2049	1.0700	0.7685	0.7183	0.6050	0.1965	1.0072	0.7197	0.7145
0.7969	0.1060	0.8564	0.6045	0.7059	0.8015	0.0977	0.8070	0.5665	0.7020

Table 4.22

Densities and Viscosities of Ternary System n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	1.5869	1.1743	0.7400	1.0000	0.0000	1.4679	1.0807	0.7362
0.0000	1.0000	2.4638	1.8631	0.7562	0.0000	1.0000	2.2427	1.6877	0.7525
0.0000	0.0000	3.6979	2.8409	0.7682	0.0000	0.0000	3.3136	2.5339	0.7647
0.1007	0.1011	3.2812	2.5100	0.7650	0.1023	0.0994	2.9716	2.2622	0.7613
0.1031	0.7990	2.4643	1.8633	0.7561	0.0985	0.8009	2.2530	1.6954	0.7525
0.2021	0.2018	2.9155	2.2196	0.7613	0.2025	0.2008	2.6506	2.0083	0.7577
0.2019	0.3946	2.7002	2.0491	0.7589	0.2038	0.3949	2.4489	1.8492	0.7551
0.1997	0.6040	2.4707	1.8682	0.7561	0.1998	0.6008	2.2579	1.6990	0.7525
0.3002	0.2943	2.5981	1.9683	0.7576	0.3023	0.2957	2.3609	1.7797	0.7538
0.3019	0.4009	2.4803	1.8753	0.7561	0.3033	0.3999	2.2593	1.6998	0.7524
0.4029	0.2007	2.4780	1.8735	0.7560	0.3984	0.2034	2.2712	1.7090	0.7525
0.4006	0.3009	2.3802	1.7964	0.7547	0.3991	0.3013	2.1783	1.6360	0.7511
0.4001	0.4047	2.2746	1.7133	0.7532	0.4024	0.4038	2.0786	1.5578	0.7495
0.6052	0.1973	2.0862	1.5650	0.7502	0.6028	0.1982	1.9195	1.4329	0.7465
0.7995	0.0998	1.8296	1.3638	0.7454	0.8027	0.0970	1.6843	1.2490	0.7416

Table 4.22 (cont'd)

Densities and Viscosities of Ternary System n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	1.2719	0.9274	0.7291	1.0000	0.0000	1.1899	0.8632	0.7254
0.0000	1.0000	1.8856	1.4058	0.7456	0.0000	1.0000	1.7410	1.2920	0.7421
0.0000	0.0000	2.7124	2.0559	0.7580	0.0000	0.0000	2.4757	1.8680	0.7545
0.1030	0.1021	2.4487	1.8474	0.7545	0.0957	0.0937	2.2586	1.6969	0.7513
0.0996	0.8060	1.8887	1.4081	0.7455	0.1027	0.7995	1.7425	1.2930	0.7420
0.2019	0.1981	2.2101	1.6596	0.7509	0.2012	0.2007	2.0273	1.5153	0.7474
0.2021	0.3938	2.0534	1.5367	0.7484	0.1956	0.3991	1.8950	1.4117	0.7449
0.2005	0.6019	1.8958	1.4135	0.7456	0.1959	0.6024	1.7563	1.3034	0.7421
0.3030	0.2983	1.9764	1.4762	0.7469	0.2922	0.2978	1.8366	1.3659	0.7437
0.2996	0.4028	1.9021	1.4182	0.7456	0.2987	0.3959	1.7620	1.3077	0.7422
0.3968	0.2000	1.9147	1.4277	0.7457	0.3950	0.2005	1.7696	1.3134	0.7422
0.3732	0.3142	1.8636	1.3879	0.7447	0.3973	0.2983	1.7017	1.2605	0.7407
0.4028	0.3998	1.7602	1.3071	0.7426	0.3945	0.4067	1.6345	1.2082	0.7392
0.6018	0.1983	1.6337	1.2082	0.7396	0.6018	0.1966	1.5163	1.1160	0.7360
0.8003	0.1013	1.4444	1.0610	0.7345	0.7985	0.1008	1.3478	0.9853	0.7310

Table 4.23

Densities and Viscosities of Ternary System n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.7734	0.5432	0.7024	1.0000	0.0000	0.7309	0.5104	0.6983
0.0000	1.0000	2.4638	1.8630	0.7561	0.0000	1.0000	2.2427	1.6877	0.7525
0.0000	0.0000	3.6979	2.8410	0.7683	0.0000	0.0000	3.3136	2.5339	0.7647
0.1051	0.0982	3.1046	2.3689	0.7630	0.1047	0.0988	2.8230	2.1438	0.7594
0.1085	0.7960	2.3131	1.7433	0.7537	0.1049	0.7983	2.1213	1.5913	0.7501
0.2089	0.2018	2.6031	1.9706	0.7570	0.2045	0.1972	2.3941	1.8043	0.7537
0.2045	0.3899	2.4192	1.8256	0.7547	0.2084	0.3920	2.1950	1.6479	0.7507
0.2074	0.5969	2.1921	1.6472	0.7514	0.2074	0.5969	2.0119	1.5043	0.7477
0.3024	0.2956	2.1955	1.6488	0.7510	0.2986	0.2974	2.0221	1.5114	0.7474
0.3061	0.4020	2.0779	1.5566	0.7491	0.3028	0.4005	1.9194	1.4310	0.7456
0.4009	0.1998	1.9797	1.4788	0.7470	0.4034	0.1996	1.8217	1.3537	0.7431
0.4030	0.2986	1.8904	1.4087	0.7452	0.4092	0.2977	1.7251	1.2784	0.7411
0.4037	0.4025	1.7943	1.3338	0.7433	0.3998	0.4065	1.6673	1.2334	0.7397
0.6185	0.1931	1.4105	1.0336	0.7328	0.6052	0.1990	1.3402	0.9781	0.7298
0.8009	0.1005	1.0752	0.7741	0.7200	0.8034	0.0973	1.0072	0.7210	0.7159

Table 4.23 (cont'd)

Densities and Viscosities of Ternary System n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.6583	0.4545	0.6904	1.0000	0.0000	0.6258	0.4296	0.6865
0.0000	1.0000	1.8856	1.4058	0.7456	0.0000	1.0000	1.7410	1.2919	0.7421
0.0000	0.0000	2.7124	2.0557	0.7579	0.0000	0.0000	2.4757	1.8680	0.7545
0.1061	0.1009	2.3343	1.7565	0.7525	0.0998	0.1140	2.1460	1.6076	0.7491
0.1086	0.7972	1.7849	1.3261	0.7430	0.1048	0.7955	1.6602	1.2280	0.7397
0.2076	0.1961	2.0020	1.4947	0.7466	0.2059	0.2011	1.8452	1.3712	0.7431
0.2048	0.3941	1.8589	1.3829	0.7439	0.2003	0.3951	1.7269	1.2790	0.7407
0.2113	0.5947	1.6999	1.2589	0.7406	0.2037	0.5988	1.5881	1.1710	0.7374
0.3057	0.2932	1.7030	1.2604	0.7401	0.3116	0.3083	1.5574	1.1462	0.7360
0.3113	0.3989	1.6182	1.1943	0.7381	0.2998	0.4051	1.5212	1.1182	0.7351
0.4065	0.1978	1.5530	1.1428	0.7359	0.4000	0.1977	1.4555	1.0664	0.7327
0.4081	0.2967	1.4838	1.0892	0.7341	0.3978	0.3018	1.3980	1.0220	0.7310
0.4017	0.4051	1.4273	1.0456	0.7326	0.3959	0.4064	1.3403	0.9775	0.7293
0.6080	0.1958	1.1654	0.8419	0.7224	0.6015	0.1995	1.1024	0.7927	0.7191
0.8052	0.0976	0.8940	0.6332	0.7082	0.8047	0.0986	0.8457	0.5957	0.7044

Table 4.24

Densities and Viscosities of Ternary System n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m^2/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	1.2583	0.9183	0.7298	1.0000	0.0000	1.1715	0.8504	0.7259
0.0000	1.0000	2.4638	1.8631	0.7562	0.0000	1.0000	2.2427	1.6877	0.7525
0.0000	0.0000	3.6979	2.8410	0.7683	0.0000	0.0000	3.3136	2.5339	0.7647
0.1002	0.1025	3.2260	2.4658	0.7644	0.0814	0.1024	2.9727	2.2630	0.7613
0.0993	0.8003	2.4266	1.8333	0.7555	0.0836	0.8163	2.2293	1.6767	0.7521
0.1979	0.2021	2.8208	2.1442	0.7602	0.1700	0.2119	2.6252	1.9880	0.7573
0.1999	0.3961	2.5995	1.9694	0.7576	0.1652	0.4175	2.4286	1.8330	0.7548
0.2022	0.6023	2.3810	1.7969	0.7547	0.1696	0.6245	2.2190	1.6682	0.7518
0.2970	0.3010	2.4600	1.8587	0.7556	0.2514	0.3155	2.3287	1.7540	0.7532
0.2981	0.4012	2.3488	1.7712	0.7541	0.2547	0.4316	2.2119	1.6622	0.7515
0.3953	0.1983	2.3147	1.7442	0.7535	0.3479	0.2183	2.2060	1.6571	0.7512
0.4052	0.2985	2.1905	1.6465	0.7516	0.3490	0.3270	2.1033	1.5766	0.7496
0.3976	0.4006	2.1145	1.5867	0.7504	0.3494	0.4377	2.0022	1.4974	0.7479
0.6004	0.2004	1.8504	1.3792	0.7454	0.5485	0.2266	1.7824	1.3249	0.7433
0.8006	0.1023	1.5322	1.1308	0.7380	0.7675	0.1177	1.4660	1.0782	0.7355

Table 4.24 (cont'd)

Densities and Viscosities of Ternary System n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m²/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L	Mole Fraction x_1	Mole Fraction x_2	Kinematic Viscosity 10^{-6} m²/s	Absolute Viscosity 10^{-3} Pa.s	Density kg/L
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	1.0306	0.7406	0.7186	1.0000	0.0000	0.9685	0.6923	0.7148
0.0000	1.0000	1.8856	1.4058	0.7455	0.0000	1.0000	1.7410	1.2919	0.7421
0.0000	0.0000	2.7124	2.0558	0.7579	0.0000	0.0000	2.4757	1.8680	0.7545
0.1003	0.1008	2.4165	1.8218	0.7539	0.1006	0.1034	2.2124	1.6604	0.7505
0.0996	0.7999	1.8601	1.3855	0.7448	0.0998	0.8028	1.7166	1.2726	0.7413
0.2013	0.1990	2.1399	1.6040	0.7496	0.1981	0.2033	1.9703	1.4702	0.7462
0.1997	0.4042	1.9808	1.4794	0.7469	0.1987	0.3985	1.8330	1.3629	0.7435
0.2000	0.6013	1.8336	1.3644	0.7441	0.2005	0.6017	1.6934	1.2541	0.7406
0.3010	0.3019	1.8780	1.3986	0.7447	0.2941	0.2992	1.7479	1.2961	0.7415
0.2998	0.4028	1.8068	1.3431	0.7433	0.3010	0.4011	1.6715	1.2366	0.7398
0.4008	0.2012	1.7816	1.3230	0.7426	0.3971	0.1948	1.6588	1.2264	0.7393
0.4006	0.3003	1.7105	1.2676	0.7411	0.3995	0.3033	1.5849	1.1690	0.7376
0.4008	0.3996	1.6417	1.2141	0.7395	0.4001	0.4007	1.5229	1.1209	0.7360
0.6014	0.2009	1.4595	1.0720	0.7345	0.6018	0.1979	1.3605	0.9944	0.7309
0.8038	0.0978	1.2314	0.8952	0.7270	0.8038	0.1000	1.1517	0.8330	0.7233

CHAPTER 5

DISCUSSION OF RESULTS

5.1 Density Data

5.1.1 Accuracy and precision of the density measurements

The DMA 60 density meter, in combination with DMA 602 remote cells, provides a system for fluid density measurement with a very high accuracy. Since the samples are kept in the closed glass sample tube during the measurement, the evaporation of the liquid can be avoided.

There are two error sources in density measurements, one is owing to the temperature fluctuation in the thermostat control of the instrument and the other, the thermal expansion of the sample itself. Usually, a possible maximum error of ± 1 in the fifth decimal place of the density meter readings appeared and such an error would result in an average error of $\pm 1.1 \times 10^{-4}$ kg/L in density (refer to Appendix C for error analysis of density measurements). In order to check the accuracy of experiments in this work, the experimental values of densities of the pure components were compared with their corresponding literature values. The comparison results are listed in Table 5.1. It is obvious that there is close agreement between the experimental density values of the pure components obtained in this study and those reported in the literature.

For n-tridecane and n-pentadecane at 298.15 and 308.15 K, the density values were

Table 5.1

. Comparison of Experimental Values with Literature Values of the Pure Component Properties

Compound	Density, kg/L		Kinematic Viscosity, 10^{-6} m^2/s		Absolute Viscosity, 10^{-3} Pa.s	
	Experimental Value	Literature Value*	Experimental Value	Literature Value*	Experimental Value	Literature Value*
			Temperature = 293.15K			
n-Octane	0.7025	0.70267	0.7734	0.7758	0.5433	0.5450
n-Decane	0.7298	0.73012	1.2583	1.268	0.9183	0.9253
n-Undecane	0.7401	0.7402	1.5869	1.601	1.1744	1.185
n-Tridecane	0.7562	0.7561	2.4638	2.486	1.8631	1.880
n-Pentadecane	0.7684	0.76838	3.6679	3.726	2.8413	2.863
			Temperature = 298.15K			
n-Octane	0.6985	0.69862	0.7309	0.7352	0.5105	0.5136
n-Decane	0.7260	0.72635	1.1715	1.182	0.8505	0.8588
n-Undecane	0.7364	0.7365	1.4679	1.483	1.0809	1.092
n-Tridecane	0.7526	0.75271	2.2427	2.266	1.6879	1.706
n-Pentadecane	0.7648	0.76490	3.3136	3.347	2.5343	2.560

* TRC Tables, 1988

Table 5.1 (cont'd)

. Comparison of Experimental Values with Literature Values of the Pure Component Properties

Compound	Density, kg/L		Kinematic Viscosity, 10^{-6} m^2/s		Absolute Viscosity, 10^{-3} Pa.s	
	Experimental Value	Literature Value	Experimental Value	Literature Value	Experimental Value	Literature Value
			Temperature = 308.15K			
n-Octane	0.6905	0.69042 [1]	0.6583	0.6649 [3]	0.4545	0.4591 [3]
n-Decane	0.7186	0.71915 [2]	1.0306	1.039 [3]	0.7406	0.7465 [3]
n-Undecane	0.7291	0.72951 [1]	1.2719	1.283 [3]	0.9274	0.9357 [3]
n-Tridecane	0.7456	0.7459* [3]	1.8856	1.909 [3]	1.4060	1.424 [3]
n-Pentadecane	0.7580	0.7581* [3]	2.7124	2.749 [3]	2.0560	2.084 [3]
			Temperature = 313.15K			
n-Octane	0.6865	0.6863 [3]	0.6258	0.6343 [3]	0.4296	0.4355 [3]
n-Decane	0.7149	0.7150 [3]	0.9685	0.9777 [3]	0.6924	0.6989 [3]
n-Undecane	0.7255	0.7255 [3]	1.1899	1.200 [3]	0.8633	0.8707 [3]
n-Tridecane	0.7421	0.7424 [3]	1.7410	1.766 [3]	1.2921	1.310 [3]
n-Pentadecane	0.7546	0.7546 [3]	2.4757	2.510 [3]	1.8682	1.894 [3]

[1] Inglese *et al.*, 1983

[2] Garcia *et al.*, 1986.

[3] TRC Tables, 1988.

* These values are not directly available from the literature. Therefore, they are calculated from absolute viscosity and kinematic viscosity values listed in [3].

not available from the literature directly. Therefore, their listed literature values were calculated from the corresponding kinematic viscosity and absolute viscosity values listed in the TRC Tables (1988).

5.1.2 The density - composition correlations

The experimental density data of each binary mixture were fitted to a polynomial of its compositions as follows

$$\rho = \sum_{i=0}^{n} A_i \, x_1^i \tag{5.1}$$

Table 5.2 lists the least squares constants of the polynomial for each binary system at each temperature level. From these constants, the density at any composition for these mixtures can be calculated in the 293 - 313 K temperature range.

5.2 Viscosity Data

5.2.1 Accuracy and precision of the viscosity measurements

By using the Cannon-Ubbelohde viscometers, the reproducibility of the viscosities were within ± 0.1%.

The accuracy of the viscosity measurements in this study has been checked by comparing the experimental values of viscosities of the pure components with their

Table 5.2

Least-Squares Constants for the Equation

$$\rho = \sum_{i=0}^{n} A_i \, x_1^i \ , \ kg \, / \, L$$

System	A_0	A_1	A_2	A_3	A_4	std dev (kg/L)
			Temperature = 293.15 K			
n-Octane(1)-n-Undecane(2)	0.7401	-0.0285	-0.0052	-0.0052	0.0014	1.36×10^{-5}
n-Octane(1)-n-Tridecane(2)	0.7562	-0.0357	-0.0072	-0.0108		3.32×10^{-5}
n-Octane(1)-n-Pentadecane(2)	0.7683	-0.0363	-0.0231	0.0074	-0.0138	2.05×10^{-5}
n-Decane(1)-n-Pentadecane(2)	0.7683	-0.0272	-0.0064	-0.0049		8.47×10^{-6}
n-Undecane(1)-n-Pentadecane(2)	0.7683	-0.0215	-0.0045	-0.0023		1.10×10^{-5}
n-Tridecane(1)-n-Pentadecane(2)	0.7683	-0.0106	0.0013	-0.0002		5.60×10^{-6}
n-Decane(1)-n-Tridecane(2)	0.7561	-0.0208	-0.0041	-0.0014		1.10×10^{-5}
n-Undecane(1)-n-Tridecane(2)	0.7562	-0.0139	-0.0020	-0.0003		1.05×10^{-5}
			Temperature = 298.15 K			
n-Octane(1)-n-Undecane(2)	0.7363	-0.0286	-0.0062	-0.0031		1.41×10^{-5}
n-Octane(1)-n-Tridecane(2)	0.7527	-0.0360	-0.0079	-0.0103		4.30×10^{-5}
n-Octane(1)-n-Pentadecane(2)	0.7648	-0.0369	-0.0219	0.0064	-0.0141	1.46×10^{-5}
n-Decane(1)-n-Pentadecane(2)	0.7648	-0.0263	-0.0112	0.0027	-0.0040	1.19×10^{-5}
n-Undecane(1)-n-Pentadecane(2)	0.7648	-0.0216	-0.0046	-0.0023		1.06×10^{-5}
n-Tridecane(1)-n-Pentadecane(2)	0.7648	-0.0106	-0.0016			1.43×10^{-5}
n-Decane(1)-n-Tridecane(2)	0.7526	-0.0211	-0.0039	-0.0015		1.52×10^{-5}
n-Undecane(1)-n-Tridecane(2)	0.7525	-0.0140	-0.0020	-0.0003		7.60×10^{-6}

Table 5.2 (cont'd)

System	A_0	A_1	A_2	A_3	A_4	std dev (kg/L)
			Temperature = 308.15 K			
n-Octane(1)-n-Undecane(2)	0.7291	-0.0291	-0.0063	-0.0033		1.67×10^{-5}
n-Octane(1)-n-Tridecane(2)	0.7456	-0.0365	-0.0081	-0.0105		4.18×10^{-5}
n-Octane(1)-n-Pentadecane(2)	0.7580	-0.0373	-0.0219	0.0051	-0.0134	2.03×10^{-5}
n-Decane(1)-n-Pentadecane(2)	0.7580	-0.0277	-0.0060	-0.0056		2.52×10^{-5}
n-Undecane(1)-n-Pentadecane(2)	0.7580	-0.0219	-0.0046	-0.0024		1.93×10^{-5}
n-Tridecane(1)-n-Pentadecane(2)	0.7580	-0.0109	-0.0011	-0.0004		1.08×10^{-5}
n-Decane(1)-n-Tridecane(2)	0.7456	-0.0207	-0.0071	0.0032	-0.0024	6.30×10^{-6}
n-Undecane(1)-n-Tridecane(2)	0.7456	-0.0139	-0.0033	0.0020	-0.0012	1.03×10^{-5}
			Temperature = 313.15 K			
n-Octane(1)-n-Undecane(2)	0.7255	-0.0295	-0.0062	-0.0033		1.68×10^{-5}
n-Octane(1)-n-Tridecane(2)	0.7421	-0.0350	-0.0167	0.0029	-0.0069	1.84×10^{-5}
n-Octane(1)-n-Pentadecane(2)	0.7545	-0.0376	-0.0212	0.0036	-0.0128	1.91×10^{-5}
n-Decane(1)-n-Pentadecane(2)	0.7546	-0.0275	-0.0075	-0.0039	-0.0008	9.36×10^{-6}
n-Undecane(1)-n-Pentadecane(2)	0.7546	-0.0220	-0.0046	-0.0025		1.32×10^{-5}
n-Tridecane(1)-n-Pentadecane(2)	0.7546	-0.0110	-0.0012	-0.0003		1.42×10^{-5}
n-Decane(1)-n-Tridecane(2)	0.7421	-0.0215	-0.0040	-0.0017		8.32×10^{-6}
n-Undecane(1)-n-Tridecane(2)	0.7421	-0.0143	-0.0019	-0.0004		1.16×10^{-5}

corresponding literature values. The results are also listed in Table 5.1. As can be seen from Table 5.1, for the viscosity, the experimental values are close to their corresponding literature values and the experimental values are always lower than the values reported by TRC Tables (1988).

5.2.2 The viscosity - composition correlations

The experimental viscosity data of each binary system were also fitted to a viscosity-composition polynomial for the purpose of viscosity prediction over the complete composition range. The polynomial is as follows

$$\nu = \sum_{i=0}^{n} B_i \, x_1^i \qquad (5.2)$$

The least squares constants are presented in Table 5.3.

5.3 The Excess Properties of Mixing

5.3.1 Excess volume of mixing

Excess volumes on mixing liquid mixtures can be attributed to any one of the following factors: (1) difference in size of the component molecules, (2) difference in shape of the component molecules, (3) structural changes such as changes in the

Table 5.3

Least-Squares Constants for the Equation

$$\nu = \sum_{i=0}^{n} B_i x_1^i \; , \quad 10^{-6} m^2/s$$

System	B_0	B_1	B_2	B_3	B_4	std dev (10^{-6} m^2/s)
			Temperature = 293.15 K			
n-Octane(1)-n-Undecane(2)	1.5880	-1.0078	0.1932			1.40 x 10^{-3}
n-Octane(1)-n-Tridecane(2)	2.4635	-2.3757	0.8463	-0.2360	0.0754	4.71 x 10^{-4}
n-Octane(1)-n-Pentadecane(2)	3.6974	-4.4392	1.6986	-0.1815		4.82 x 10^{-3}
n-Decane(1)-n-Pentadecane(2)	3.6946	-3.2566	0.8218			2.93 x 10^{-3}
n-Undecane(1)-n-Pentadecane(2)	3.6932	-2.6391	0.5326			4.99 x 10^{-3}
n-Tridecane(1)-n-Pentadecane(2)	3.6987	-1.4283	0.2482	-0.0544		2.11 x 10^{-3}
n-Decane(1)-n-Tridecane(2)	2.4634	-1.4711	0.2663			9.57 x 10^{-4}
n-Undecane(1)-n-Tridecane(2)	2.4637	-1.0127	0.1637	-0.0279		8.64 x 10^{-4}
			Temperature = 298.15 K			
n-Octane(1)-n-Undecane(2)	1.4679	-0.9062	0.1934	-0.0244		1.30 x 10^{-3}
n-Octane(1)-n-Tridecane(2)	2.2429	-2.0713	0.6470	-0.0878		1.00 x 10^{-3}
n-Octane(1)-n-Pentadecane(2)	3.3136	-3.8014	1.3631	-0.1443		3.45 x 10^{-4}
n-Decane(1)-n-Pentadecane(2)	3.3140	-2.8313	0.7450	-0.0563		6.42 x 10^{-4}
n-Undecane(1)-n-Pentadecane(2)	3.3148	-2.3318	0.5763	-0.0921		3.05 x 10^{-3}
n-Tridecane(1)-n-Pentadecane(2)	3.3132	-1.1984	0.1299			1.70 x 10^{-3}
n-Decane(1)-n-Tridecane(2)	2.2429	-1.2999	0.2599	-0.0319		1.20 x 10^{-3}
n-Undecane(1)-n-Tridecane(2)	2.2430	-0.8872	0.1133			1.30 x 10^{-3}

Table 5.3 (Cont'd)

System	B_0	B_1	B_2	B_3	B_4	std dev (10^{-6} m^2/s)
Temperature = 308.15 K						
n-Octane(1)-n-Undecane(2)	1.2718	-0.7383	0.1884	-0.1209	0.0576	6.65×10^{-4}
n-Octane(1)-n-Tridecane(2)	1.8851	-1.5625	0.1935	0.3476	-0.2044	1.43×10^{-3}
n-Octane(1)-n-Pentadecane(2)	2.7125	-2.8422	0.7394	0.1699	-0.1215	9.34×10^{-4}
n-Decane(1)-n-Pentadecane(2)	2.7125	-2.1355	0.4540			8.33×10^{-4}
n-Undecane(1)-n-Pentadecane(2)	2.7126	-1.7418	0.3008			5.48×10^{-4}
n-Tridecane(1)-n-Pentadecane(2)	2.7130	-0.9041	0.0772			6.81×10^{-4}
n-Decane(1)-n-Tridecane(2)	1.8856	-1.0066	0.1517			4.49×10^{-4}
n-Undecane(1)-n-Tridecane(2)	1.8856	-0.6982	0.1450	-0.1209	0.0602	4.17×10^{-4}
Temperature = 313.15 K						
n-Octane(1)-n-Undecane(2)	1.1898	-0.6748	0.1845	-0.1442	0.0687	5.57×10^{-4}
n-Octane(1)-n-Tridecane(2)	1.7413	-1.4266	0.3461	-0.0354		8.11×10^{-4}
n-Octane(1)-n-Pentadecane(2)	2.4753	-2.4806	0.4922	0.3424	-0.2026	1.47×10^{-3}
n-Decane(1)-n-Pentadecane(2)	2.4758	-1.8774	0.3436	0.0630	-0.0365	2.97×10^{-4}
n-Undecane(1)-n-Pentadecane(2)	2.4752	-1.5327	0.2474			2.44×10^{-3}
n-Tridecane(1)-n-Pentadecane(2)	2.4759	-0.8048	0.0701			4.19×10^{-4}
n-Decane(1)-n-Tridecane(2)	1.7410	-0.9025	0.1378	-0.0076		1.88×10^{-4}
n-Undecane(1)-n-Tridecane(2)	1.7411	-0.6255	0.1239	-0.1039	0.0542	3.28×10^{-4}

correlation of molecular orientations, (4) difference in the intermolecular interaction energy between like and unlike molecules, and (5) formation of new chemical species. Normally volume changes occur because of a combination of these various factors. Consequently the complexity associated with the origin of excess volume, coupled with ease with which the latter can be obtained experimentally with good precision, makes it a sensitive tool for testing the theories of liquid mixtures. Apart from this, excess volume (V^E) data are useful in the conversion of excess thermodynamic functions determined at constant pressure to the condition of mixing at constant volume, in determining composition from density measurements on mixtures, and in industrial technology. These measurements of excess volumes continue to be an area of active interest (Handa and Benson, 1979).

Figures 5.1 to 5.4 represent plots of the excess molar volumes of the binary mixtures versus composition at the four designated temperatures. It can be observed from these figures that almost all the values of excess volumes are negative and for systems n-undecane-n-tridecane and n-tridecane-n-pentadecane the values of V^E are very close to zero. It can also be seen from Figures 5.1 to 5.4 that the excess molar volumes of mixing increase with the increasing differences between the numbers of carbon atoms of the two components of a system and with the temperature.

From the binary and ternary experimental excess volume data presented in Tables 4.4 to 4.11 and Tables B.1 to B.5, respectively, it is found that the values of all the excess volumes are negative except for four data points in binary systems (they are: C_{11} - C_{13} at $x_1 = 0.8980$, 293.15 K; C_{13} - C_{15} at $x_1 = 0.8983$, 298.15 K, at $x_1 = 0.1004$, 308.15

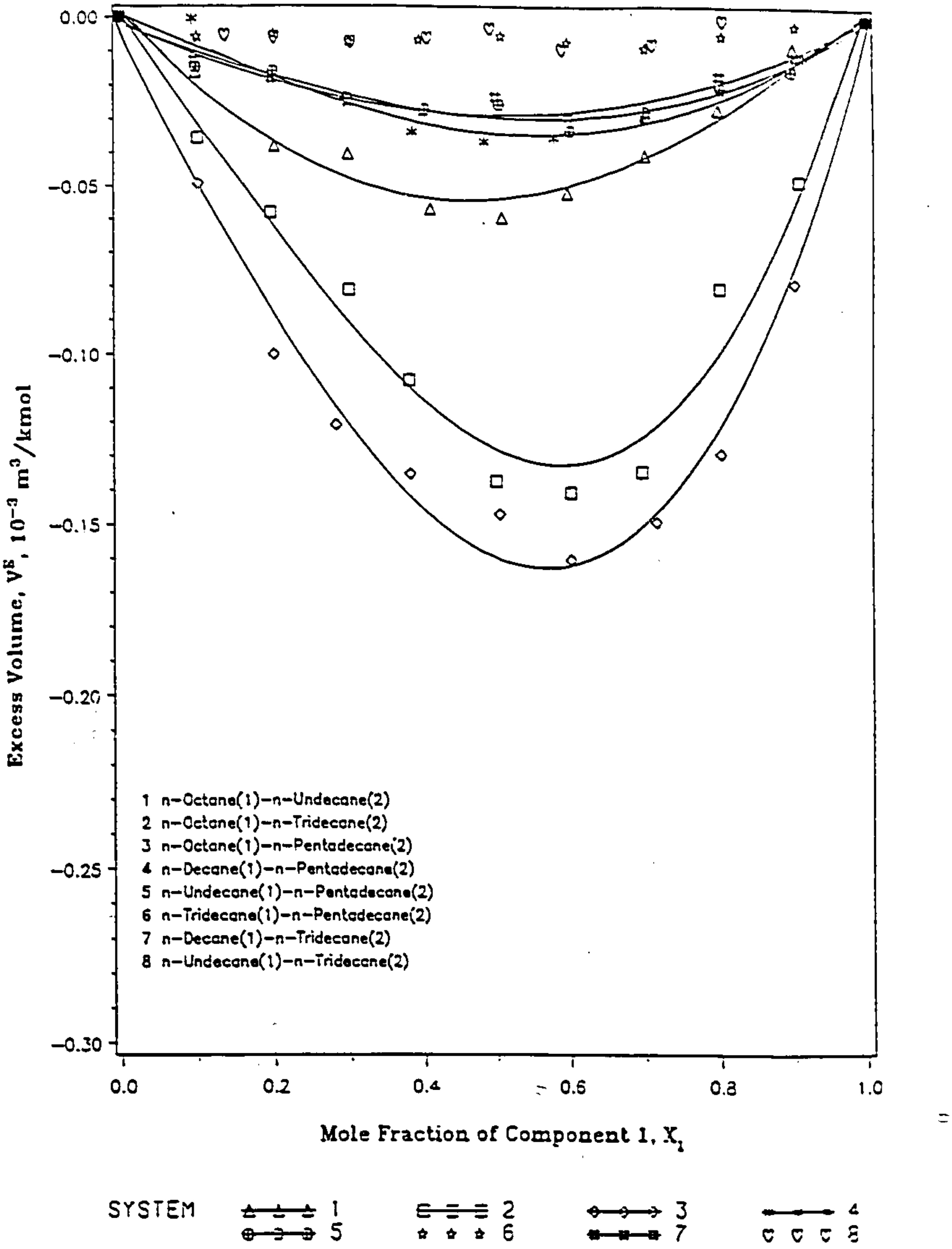


Figure 5.1. Excess Volume vs. Composition for Binary n-Alkane Systems at 293.15 K

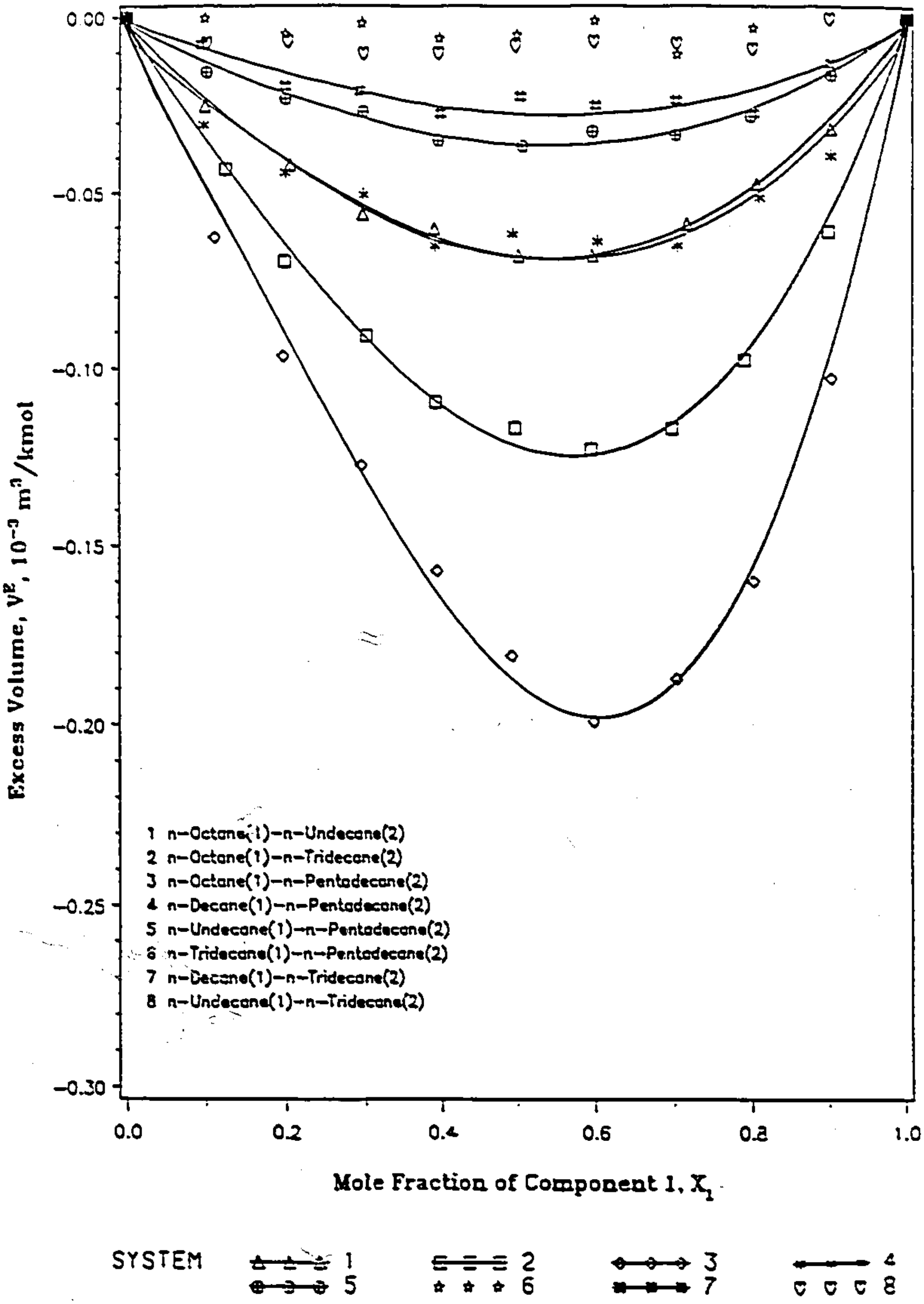


Figure 5.2. Excess Volume vs. Composition for Binary n-Alkane Systems at 298.15 K

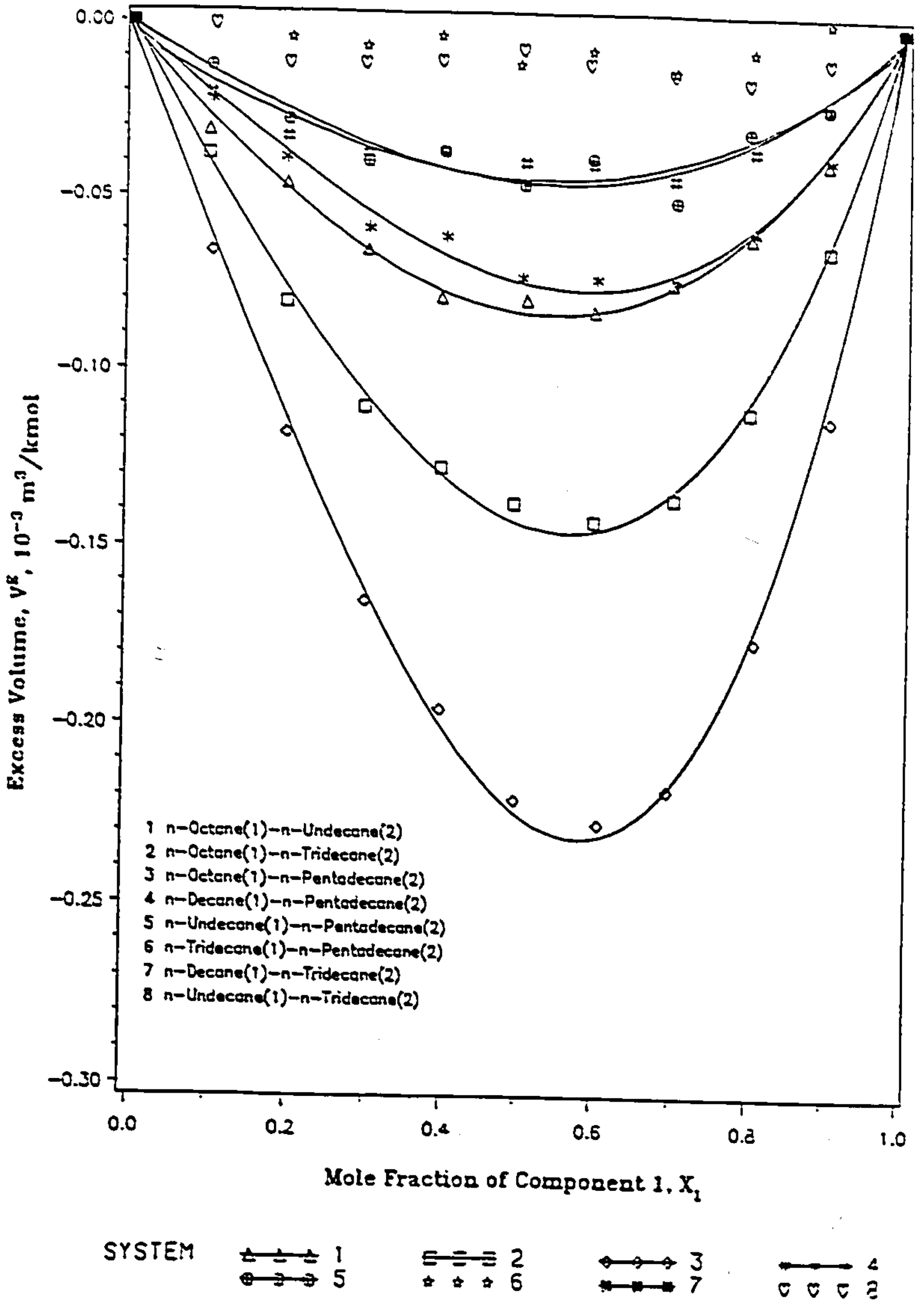


Figure 5.3. Excess Volume vs. Composition for Binary n-Alkane Systems at 308.15 K

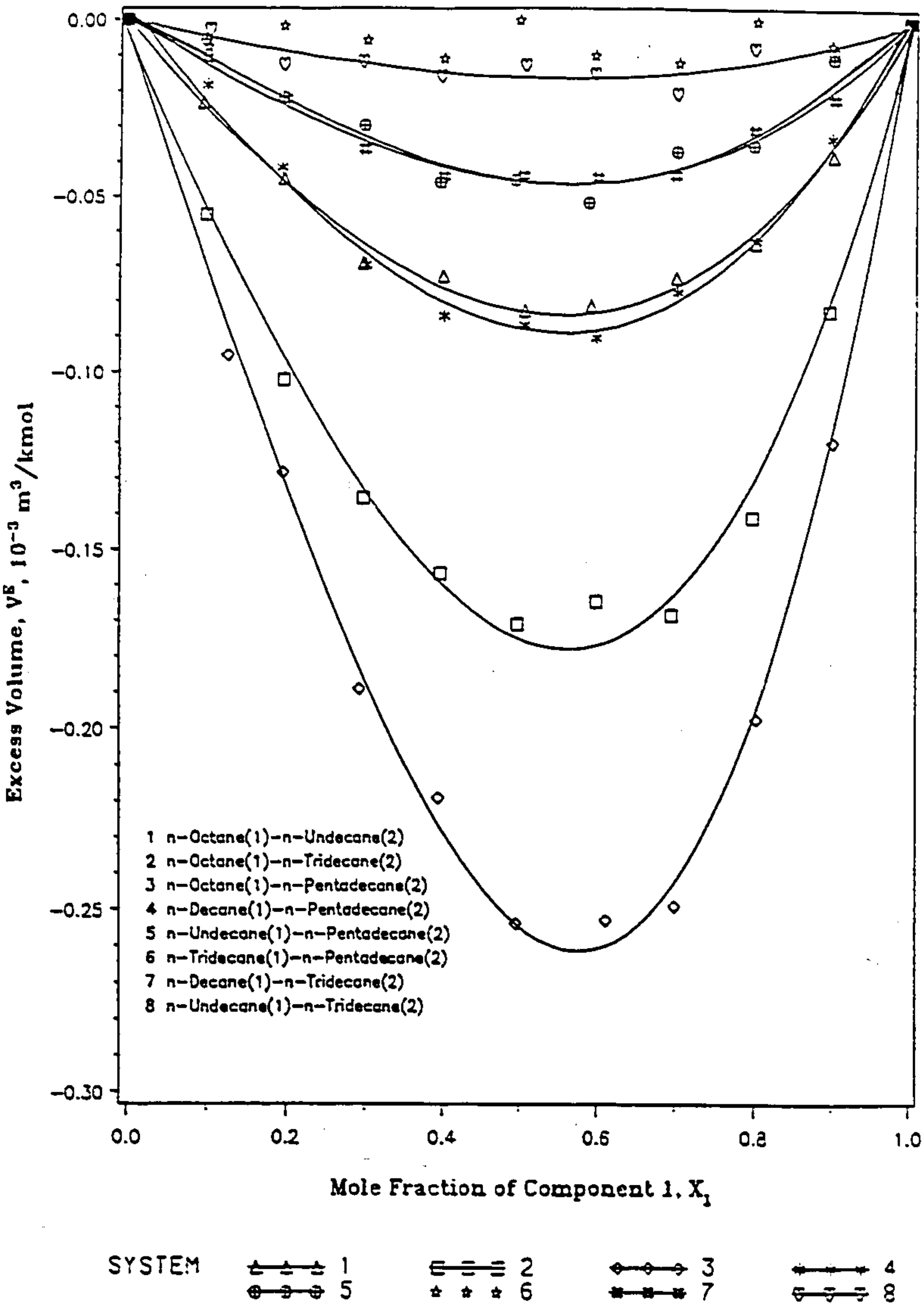


Figure 5.4. Excess Volume vs. Composition for Binary n-Alkane Systems at 313.15 K

K and at x_1 = 0.9036, 308.15 K) and one data point in ternary systems (it is: C_{10} - C_{11} - C_{15} at x_1 = 0.0836, x_2 = 0.8163, 298.15 K). These five data points show positive values for V^E and their values are very close to zero. The slightly positive values of V^E can be attributed to experimental errors. The negative values of the excess volumes of both binary and ternary n-alkane mixtures are consistent with the predictions of the Congruence Principle (Brønsted and Koefoed, 1946; Desmyter and van der Waals, 1958; Lim *et al.*, 1980).

The maximum error in determining the excess volume data was estimated to be ± 0.0002 L/kmol (Wu and Asfour, 1991a). Appendix C gives the details of error analysis for excess volumes.

If the additivity of volumes on mixing is used as a criterion of ideal solution behavior (Asfour and Dullien, 1981), then the binary systems n-undecane-n-tridecane and n-tridecane-n-pentadecane, and the ternary system n-undecane-n-tridecane-n-pentadecane at all four temperature levels are quite close to ideal solution behavior because their absolute values of V^E are very close to zero (refer to Tables 4.4 to 4.11 and B.1 to B.5, respectively).

Garcia *et al.* (1986) reported excess volume data on the system n-octane-n-undecane between 288.15 and 308.15 K. V^E values obtained in this study for this system are in very good agreement with the data reported by Garcia *et al.* in two respects: firstly, V^E values for this system are negative over the entire composition range at 293.15, 298.15 and 308.15 K. Secondly, at 293.15, 298.15 and 308.15 K, the composition range which corresponds to the minimum V^E values and the minimum values of V^E are very close.

Comparison of the two sets of data at these three temperature levels is given in Table 5.4.

No comparison can be made at 313.15 K since Garcia *et al.* did not report data at that temperature.

On the basis of the experimental data of excess volume of both binary and ternary systems (section 4.4 and Apendice B) one can conclude that the trends of the excess volume - composition relationship are similar at different temperatures. Therefore, data at only one temperature level were arbitrarily selected to test some literature models as follows. The chosen temperature level is 293.15 K.

For the eight binary systems at 293.15 K, the excess volume data have been reported in Chapter 4 (Tables 4.4 to 4.11). These data were correlated to the composition by the following expression which was reported by Heric and Brewer (1969)

$$V^E = x_1 x_2 [A_{12} + B_{12}(x_1 - x_2) + C_{12}(x_1 - x_2)^2 + \cdots] \tag{5.3}$$

and

$$V^E = x_1 x_2 [A'_{12} + B'_{12} x_1 + C'_{12} x_1^2 + \cdots] \tag{5.4}$$

Eq. (5.3) is called symmetric because it reduces to a single term in A_{ij} at $x_i = x_j = 0.5$ whereas eq. (5.4) is called asymmetric. The orders of eqs. (5.3) and (5.4) were selected for a system beyond which additional terms did not give significantly improved results. The constants of the above equations were calculated by the least squares technique and are reported in Table 5.5.

The following equations reported by Heric and Brewer (1969) for ternary systems were fitted to the ternary system data obtained in this study:

Table 5.4

Comparison of the Minimum Values of Fxcess Volume of This Study with Literature Data for System n-Octane(1)-n-Undecane(2)

Garcia *et al.*		This Study	
composition x_1	minimum V^E L/kmol	composition x_1	minimum V^E L/kmol
Temperature = 293.15 K			
0.5259	-0.0598	0.5076	-0.0597
Temperature = 298.15 K			
0.5134	-0.0632	0.5019	-0.0681
0.5902	-0.0629	0.5959	-0.0681
Temperature = 308.15 K			
0.4920	-0.0744	0.5094	-0.0793
0.5403	-0.0744	0.5981	-0.0825

Table 5.5

Least-Square Constants for Heric's Binary Excess Volume Model at 293.15 K

Asymmetric Model

System	A_{12}	B_{12}	C_{12}	D_{12}	std dev L/kmol
n-octane(1)-n-undecane(2)	-0.2306	0.0374	0.1297		0.0031
n-octane(1)-n-tridecane(2)	-0.5366	-0.3206	0.1886	0.4033	0.0065
n-octane(1)-n-pentadecane(2)	-0.6100	-0.1754	-0.2305		0.0045
n-decane(1)-n-pentadecane(2)	-0.1487	-0.0354	0.0947		0.0023
n-undecane(1)-n-pentadecane(2)	-0.0093	-0.0523			0.0058
n-tridecane(1)-n-pentadecane(2)	0.0417	-0.0617	0.2277		0.0064
n-decane(1)-n-tridecane(2)	-0.0322	-0.0719	0.1829		0.0031
n-undecane(1)-n-tridecane(2)	-0.0268	0.0200			0.0029

Symmetric Model

System	A'_{12}	B'_{12}	C'_{12}	D'_{12}	std dev L/kmol
n-octane(1)-n-undecane(2)	-0.1383	-0.4442	0.5190		0.0031
n-octane(1)-n-tridecane(2)	-0.4307	1.0241	-4.0849	3.2262	0.0065
n-octane(1)-n-pentadecane(2)	-0.6652	0.5715	-0.9222		0.0045
n-decane(1)-n-pentadecane(2)	0.0528	-1.0183	1.6615	-0.8576	0.0018
n-undecane(1)-n-pentadecane(2)	0.0430	-0.1045			0.0058
n-tridecane(1)-n-pentadecane(2)	0.4915	-2.3116	3.7884	-1.9214	0.0057
n-decane(1)-n-tridecane(2)	0.2225	-0.8752	0.7315		0.0031
n-undecane(1)-n-tridecane(2)	-0.1382	0.8210	-1.8526	1.2865	0.0021

$$V^E = \sum_{\substack{i=1 \\ i<j}}^{n} x_i x_j [A_{ij} + B_{ij}(x_i - x_j) + (x_i - x_j)^2 + \cdots] \\ + x_1 x_2 x_3 A_{123} \tag{5.5}$$

and by the asymmetric form of eq. (5.5)

$$V^E = \sum_{\substack{i=1 \\ i<j}}^{n} x_i x_j [A'_{ij} + B'_{ij} x_i + C'_{ij} x_i^2 + \cdots] + x_1 x_2 x_3 A'_{ij} \tag{5.6}$$

The constants in eqs. (5.5) and (5.6) were determined by the least squares methods from the ternary excess volume data at 293.15 K and are listed in Table 5.6.

5.3.2 Excess activation energy of viscous flow

The experimental viscosity data were used to calculate the excess activation energy of viscous flow by eq. (2.113). The values of excess activation energy of viscous flow of binary and ternary mixtures have already been reported in Chapter 4 (section 4.4) and in Appendix B. All the values of excess energy are positive and increase with temperature. The positive values of $\Delta^* G^E$ indicate that viscosity of the mixtures are greater than that of the ideal mixtures.

The plot of $\Delta^* G^E$ versus mole fraction of component 1 of the eight binary systems at four temperatures are shown in Figures 5.5 to 5.8. All the curves seem to be symmetrical with their maxima at $x_1 = 0.5$.

According to Figures 5.5 to 5.8, the changes of the excess free energy of viscous

Table 5.6

Least-Squares Constants for Heric's Ternary Excess Volume Model at 293.15 K

	Asymmetric Model		Symmetric Model	
System	A_{123}	std dev L/kmol	A^*_{123}	std dev L/kmol
n-octane(1)-n-undecane(2)-n-tridecane(3)	0.0773	0.0073	-0.2071	0.0066
n-octane(1)-n-undecane(2)-n-pentadecane(3)	-0.6843	0.0089	-0.8422	0.0092
n-undecane(1)-n-tridecane(2)-n-pentadecane(3)	-0.9079	0.0119	-1.0740	0.0142
n-octane(1)-n-tridecane(2)-n-pentadecane(3)	-0.1288	0.0104	-0.6344	0.0096
n-decane(1)-n-tridecane(2)-n-pentadecane(3)	-0.9040	0.0088	-1.2160	0.0112

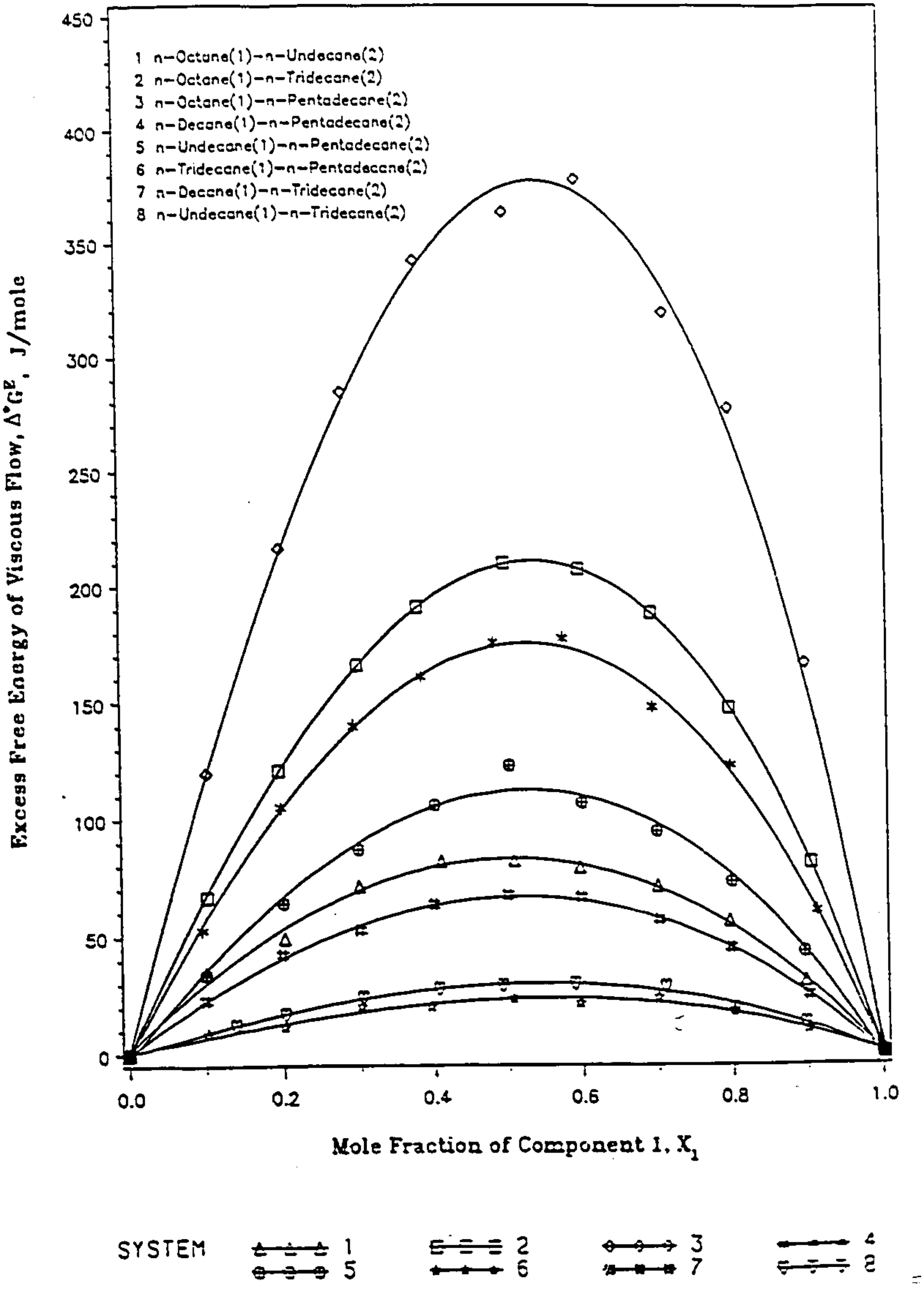


Figure 5.5. Excess Free Energy of Viscous Flow vs. Composition for Binary n-Alkane Systems at 293.15 K

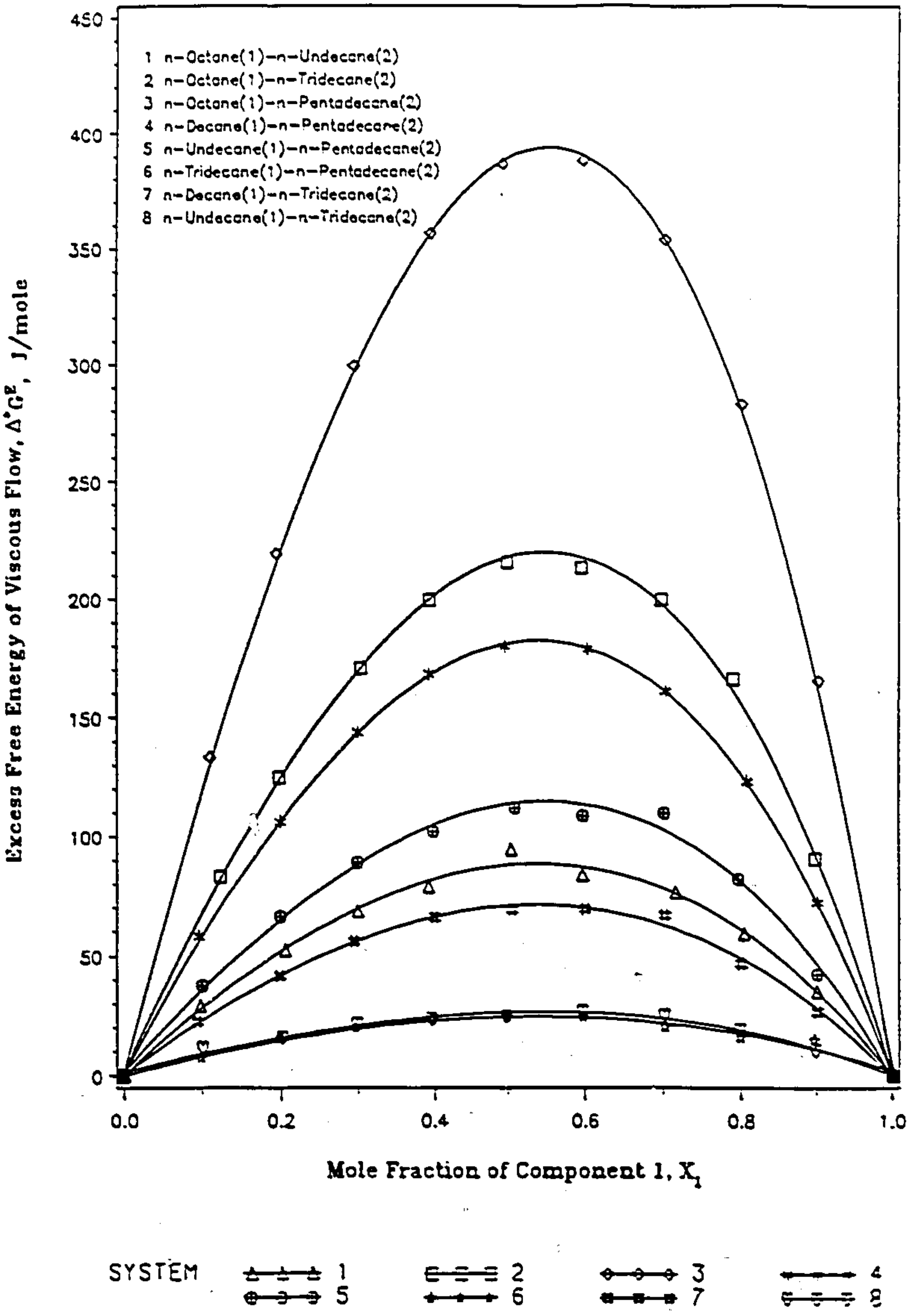


Figure 5.6. Excess Free Energy of Viscous Flow vs. Composition for Binary n-Alkane Systems at 298.15 K

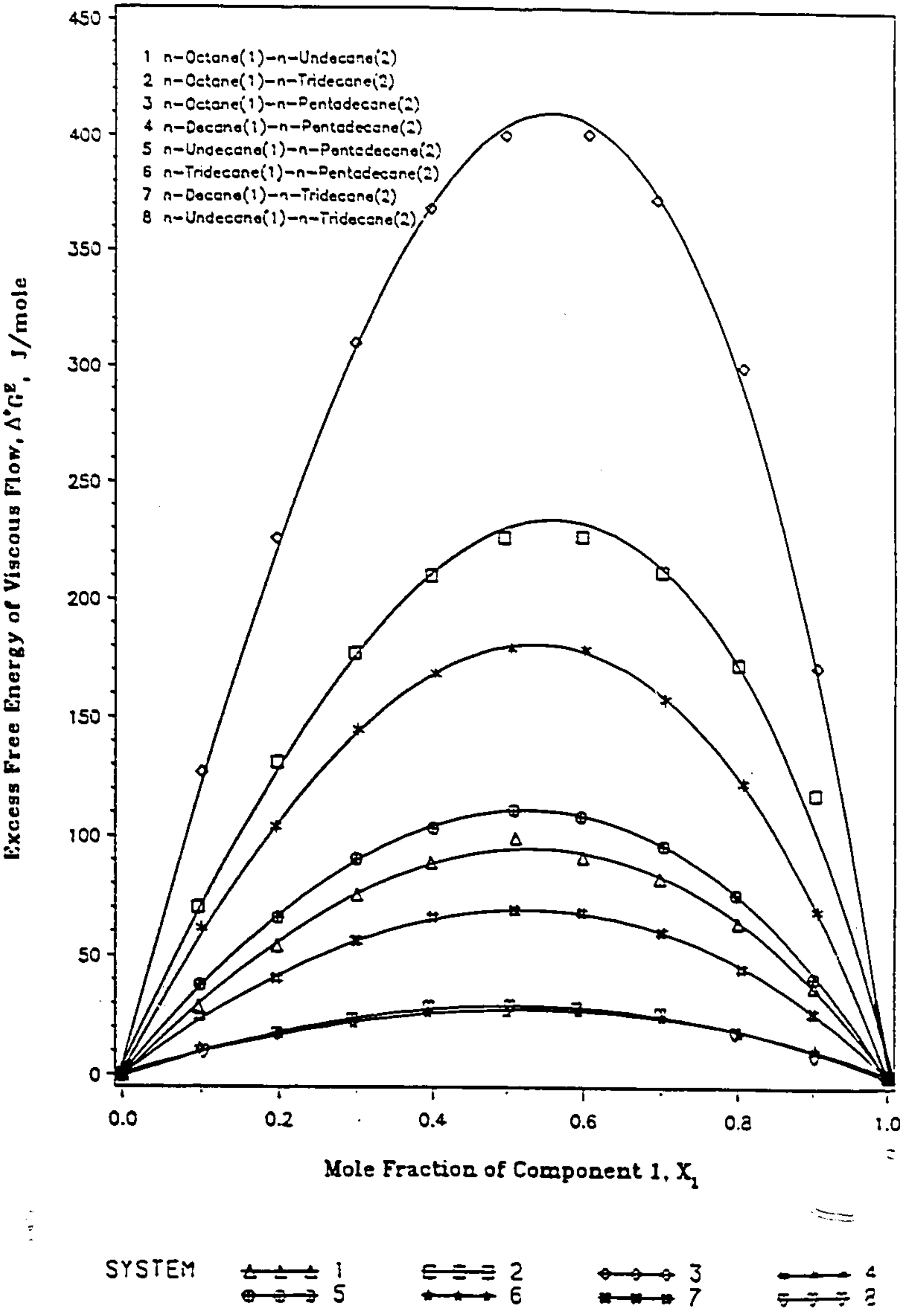


Figure 5.7. Excess Free Energy of Viscous Flow vs. Composition for Binary n-Alkane Systems at 308.15 K

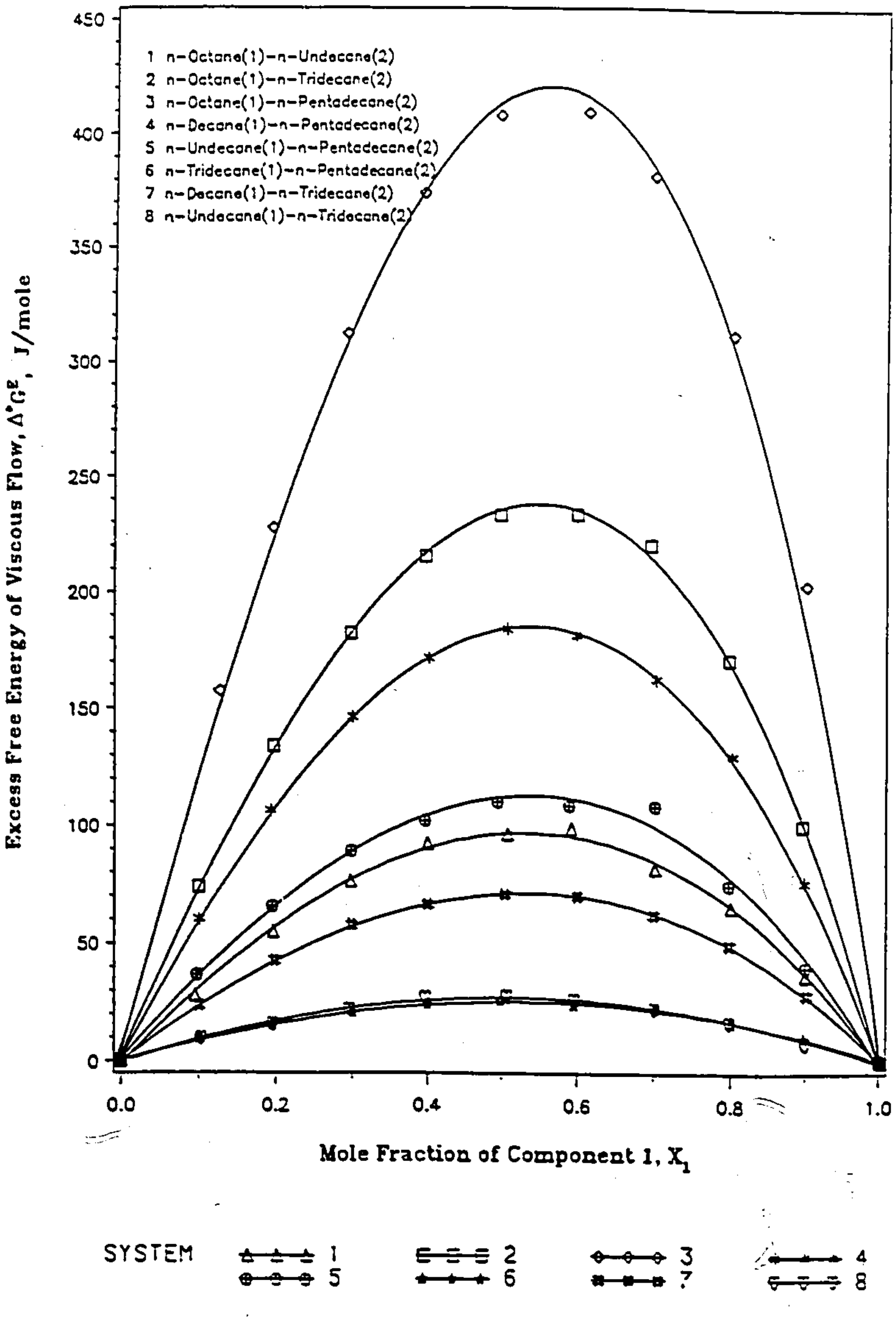


Figure 5.8. Excess Free Energy of Viscous Flow vs. Composition for Binary n-Alkane Systems at 313.15 K

flow with composition at the four temperature levels investigated are similar. Therefore, the values of $\Delta^{*}G^{E}$ vs composition at 293.15 K were used to test the following literature model proposed by Heric and Brewer (1969):

$$\Delta^{*}G^{E} = x_1x_2[A_{12} + B_{12}(x_1 - x_2) + C_{12}(x_1 - x_2)^2 + \cdots] \qquad (5.7)$$

or alternatively

$$\Delta^{*}G^{E} = x_1x_2[A'_{12} + B'_{12}x_1 + C'_{12}x_1^2 + \cdots] \qquad (5.8)$$

The least squares constants of eqs. (5.7) and (5.8) are presented in Table 5.7.

The ternary excess activation energy data at 293.15 K were used to test the following extensions of eqs. (5.7) and (5.8):

$$\Delta^{*}G^{E} = \sum_{\substack{i=1\\ i<j}}^{3} x_ix_j[A_{ij} + B_{ij}(x_i - x_j) + C_{ij}(x_i - x_j^2 + \cdots] + x_1x_2x_3A_{123} \qquad (5.9)$$

and

$$\Delta^{*}G^{E} = \sum_{\substack{i=1\\ i<j}}^{3} x_ix_j[A'_{ij} + B'_{ij}x_i + C'_{ij}x_i^2 + \cdots] + x_1x_2x_3A'_{123} \qquad (5.10)$$

Also, eq. (5.9) is called symmetric and eq. (5.10) asymmetric. The least squares constants of eqs. (5.9) and (5.10) are listed in Table 5.8.

Table 5.7

Least-Squares Constants for Heric's Binary Model of Excess Energy of Viscous Flow at 293.15 K

Asymmetric Model

System	A_{12}	B_{12}	C_{12}	D_{12}	std dev 10^2 J/mol
n-octane(1)-n-undecane(2)	3.3057	0.1489	0.1674	-0.3077	0.0256
n-octane(1)-n-tridecane(2)	8.3919	1.2183	-0.0697	0.1343	0.0040
n-octane(1)-n-pentadecane(2)	14.8286	2.0369	1.2168	2.2212	0.0646
n-decane(1)-n-pentadecane(2)	6.9897	0.7280			0.0292
n-undecane(1)-n-pentadecane(2)	4.3953	0.5858			0.0304
n-tridecane(1)-n-pentadecane(2)	1.0214	0.1447	-0.2143		0.0100
n-decane(1)-n-tridecane(2)	2.7004	0.2634	0.3140		0.0180
n-undecane(1)-n-tridecane(2)	1.1920	0.2016			0.0102

Symmetric Model

System	A'_{12}	B'_{12}	C'_{12}	D'_{12}	std dev 10^2 J/mol
n-octane(1)-n-undecane(2)	3.4260	-0.5725	0.6665		0.0244
n-octane(1)-n-tridecane(2)	6.9695	3.5215	-1.8910	1.0748	0.0040
n-octane(1)-n-pentadecane(2)	11.7873	12.5339	-21.7876	-17.7699	0.0646
n-decane(1)-n-pentadecane(2)	6.2617	1.4560			0.0292
n-undecane(1)-n-pentadecane(2)	3.0741	7.7890	-16.2447	11.5728	0.0230
n-tridecane(1)-n-pentadecane(2)	0.6623	1.1466	-0.8571		0.0100
n-decane(1)-n-tridecane(2)	2.2890	2.9753	-7.1037	5.5936	0.0158
n-undecane(1)-n-tridecane(2)	0.9904	0.4033			0.0102

5.4 Application of the McAllister Viscosity Model to n-Alkane Liquid Mixtures

5.4.1 Test of the McAllister model using binary mixture data

The semi-empirical McAllister model is based on Eyring's theory of absolute reaction rates and the assumption that for liquid mixtures the free energies of activation for viscosity are additive on a mole fraction basis (McAllister, 1960). By considering the three- and four-body interactions in one plane, respectively, McAllister derived two equations: one is cubic which is given by eq. (2.25), the other is quantic which is given by eq. (2.26). Since the less complicated three-body model works very well for our n-alkane mixtures, it was chosen to fit the experimentally obtained binary viscosity data. The values of the two adjustable parameters of the model, ν_{12} and ν_{21}, were calculated from the experimental binary viscosity data by the least squares technique at each designated temperature, and are listed in Table 5.9. Also listed in Table 5.9 are the standard deviations of the fit.

Asfour *et al.* (1991) developed a new technique for predicting the McAllister parameters for binary n-alkane liquid systems from pure component properties. This technique has already been covered in Chapter 2.

The experimental kinematic viscosity - composition data obtained in this study for the binary systems: n-octane-n-undecane, n-tridecane-n-pentadecane, n-decane-n-tridecane and n-undecane-n-tridecane in the temperature range 293.15 to 313.15 K were used in predicting the McAllister three-body model parameters. Table 5.10 shows a comparison

Table 5.8

Least-Squares Constants for Heric's Ternary Model of Excess Energy of Viscous Flow at 293.15 K

	Asymmetric Model		Symmetric Model	
System	A_{123}	std dev 10^2 J/mol	A'_{123}	std dev 10^2J/mol
n-octan(1)e-n-undecane(2)-n-tridecane(3)	-1.2108	0.015	0.2544	0.016
n-octane(1)-n-undecane(2)-n-pentadecane(3)	0.5110	0.031	3.1294	0.039
n-undecane(1)-n-tridecane(2)-n-pentadecane(3)	-2.2438	0.041	-1.3709	0.036
n-octane(1)-n-tridecane(2)-n-pentadecane(3)	-3.4659	0.047	0.1985	0.045
n-decane(1)-n-tridecane(2)-n-pentadecane(3)	-1.6942	0.051	-0.6529	0.049

Table 5.9

Values of Parameters in McAllister Binary Mixture Model for n-Alkane Mixtures

	ν_{12}	ν_{21}	std dev 10^{-6} m^2/s	ν_{12}	ν_{21}	std dev 10^{-6} m^2/s
System	Temperature = 293.15 K			Temperature = 298.15 K		
C_8(1)-C_{11}(2)	1.0177	1.2925	9.68×10^{-4}	0.9624	1.1992	1.07×10^{-3}
C_8(1)-C_{13}(2)	1.2643	1.8020	1.88×10^{-4}	1.1892	1.6566	1.07×10^{-3}
C_8(1)-C_{15}(2)	1.5883	2.4949	3.10×10^{-3}	1.4896	2.2742	1.06×10^{-3}
C_{10}(1)-C_{15}(2)	1.9662	2.7651	1.20×10^{-3}	1.8180	2.5052	3.14×10^{-4}
C_{11}(1)-C_{15}(2)	2.2269	2.9159	1.82×10^{-3}	2.0481	2.6288	1.42×10^{-3}
C_{13}(1)-C_{15}(2)	2.8599	3.2542	6.24×10^{-4}	2.5879	2.9367	7.19×10^{-4}
C_{10}(1)-C_{13}(2)	1.6240	2.0232	4.05×10^{-4}	1.5071	1.8539	7.37×10^{-4}
C_{11}(1)-C_{13}(2)	1.8671	2.1504	4.17×10^{-4}	1.7135	1.9656	5.96×10^{-4}
	Temperature = 308.15 K			Temperature = 313.15 K		
C_8(1)-C_{11}(2)	0.8564	1.0554	6.91×10^{-4}	0.8099	0.9927	7.42×10^{-4}
C_8(1)-C_{13}(2)	1.0580	1.4207	3.06×10^{-3}	0.9899	1.3322	8.92×10^{-4}
C_8(1)-C_{15}(2)	1.3073	1.9141	2.05×10^{-3}	1.2359	1.7634	3.73×10^{-3}
C_{10}(1)-C_{15}(2)	1.5529	2.0965	4.14×10^{-4}	1.4481	1.9301	4.09×10^{-4}
C_{11}(1)-C_{15}(2)	1.7247	2.1988	2.73×10^{-4}	1.6038	2.0176	1.46×10^{-3}
C_{13}(1)-C_{15}(2)	2.1566	2.4301	2.97×10^{-4}	1.9802	2.2237	1.93×10^{-4}
C_{10}(1)-C_{13}(2)	1.3001	1.5826	2.52×10^{-4}	1.2167	1.4685	1.74×10^{-4}
C_{11}(1)-C_{13}(2)	1.4685	1.6730	4.00×10^{-4}	1.3649	1.5505	3.56×10^{-4}

between the values of ν_{12} and ν_{21} determined from fitting experimental data to eq. (2.25) using the method of least squares and those calculated from eqs. (2.27) and (2.28). The close agreement is very obvious. Moreover, the values of ν_{12} and ν_{21} calculated from eqs. (2.27) and (2.28) were substituted into eq. (2.25) and the kinematic viscosities of the mixture were calculated over the entire composition range. Then the calculated and the experimental kinematic viscosity were compared. The average absolute deviation (AAD) is defined by

$$AAD = \frac{1}{m}\{\sum_{i}^{m} |\frac{\eta_i^{calcd} - \eta_i^{exptl}}{\eta_i^{exptl}}| \times 100\} \qquad (5.11)$$

where AAD is a percentage and m is the number of the experimental points. The values of AAD are also listed in Table 5.10. The very good agreement between the calculated and the experimental kinematic viscosity is quite clear.

To test the equations of predicting the McAllister four-body binary parameters, eqs. (2.29) and (2.30), proposed also by Asfour *et al.* (1991), the viscosity data of the following binary systems were used: n-octane-n-tridecane, n-octane-n-pentadecane, n-decane-n-pentadecane and n-undecane-n-pentadecane. The comparison of the values of ν_{1112}, ν_{1122} and ν_{2221} calculated from eqs. (2.29) and (2.30) with those determined from experimental data is shown in Table 5.11. The corresponding AAD values are also listed in Table 5.11. Again, there is very close agreement between the predicted and the experimental values.

Table 5.10

Comparison of the Method of Asfour *et al.* with Experimental Data
for Binary McAllister Three-body Model

System	Temp. K	Calculated from Experimental Data		Calculated from eqs. (2.27) & (2.28)		
		ν_{12}	ν_{21}	ν_{12}	ν_{21}	AAD
n-Octane(1)	293.15	1.027	1.293	1.026	1.304	0.4
-n-Undecane(2)	298.15	0.963	1.199	0.963	1.215	0.3
	308.15	0.857	1.055	0.856	1.066	0.2
	313.15	0.811	0.992	0.810	1.003	0.2
n-Tridecane(1)	293.15	2.859	3.260	2.857	3.271	0.07
-n-Pentadecane(2)	298.15	2.587	2.937	2.587	2.947	0.1
	308.15	2.173	2.426	2.156	2.434	0.15
	313.15	2.980	2.224	1.983	2.230	0.1
n-Decane(1)	293.15	1.627	2.024	1.631	2.040	0.02
-n-Tridecane(2)	298.15	1.507	1.854	1.507	1.871	0.02
	308.15	1.300	1.583	1.306	1.597	0.03
	313.15	1.216	1.469	1.220	1.484	0.03
n-Undecane(1)	293.15	1.867	2.150	1.865	2.160	0.1
-n-Tridecane(2)	298.15	1.718	1.967	1.716	1.977	0.1
	308.15	1.469	1.673	1.472	1.679	0.15
	313.15	1.365	1.550	1.371	1.557	0.2

5.4.2 Extension of the Asfour *et al.* technique to n-alkane ternary systems

The extended McAllister expression for ternary systems, eq. (2.31), has already been given in Chapter 2. The values of the ternary parameter, ν_{123}, determined from the experimental ternary viscosity data in this study are listed in Table 5.12.

The ternary parameter ν_{123} involves the interactions among three molecules each of type 1, 2 and 3 whereas ν_1, ν_2 and ν_3, the kinematic viscosities of pure components 1, 2 and 3, involve the interactions among molecules of type 1, type 2 and type 3, respectively (Kalidas and Laddha, 1964). Therefore, based on the technique of Asfour *et al.*(1991), it is assumed that

$$\nu_{123} \propto (\nu_1 \nu_2 \nu_3)^{1/3} \tag{5.12}$$

in this way, the temperature dependent parameter ν_{123} is converted into a temperature independent lumped parameter $\nu_{123}/(\nu_1\nu_2\nu_3)^{1/3}$ as shown in Table 5.13.

Plotting $\nu_{123}/(\nu_1\nu_2\nu_3)^{1/3}$ versus $(N_3 - N_1)^2/N_2$ gives a straight line as shown in Figure 5.9. The equation of the straight line is

$$\frac{\nu_{123}}{(\nu_1 \nu_2 \nu_3)^{1/3}} = 0.9942 + 0.03167 \frac{(N_3 - N_1)^2}{N_2} \tag{5.13}$$

By using eq. (5.13), the ternary McAllister parameter ν_{123} can be predicted from the viscosity and the numbers of carbon atoms of the pure components of the n-alkane mixtures. Eq. (5.13) was tested by using the viscosity data of n-hexane-n-tetradecane-n-hexdecane mixture at 298.15 K taken from literature (Heric and Brewer, 1969). First, the

Table 5.11

Comparison of the Method of Asfour *et al.* with Experimental Data for McAllister Binary Four-body Model

System	Temp. K	Calculated from Experimental Data ν_{1112}	Calculated from Experimental Data ν_{1122}	Calculated from Experimental Data ν_{2221}	Calculated from eqs. (2.29) & (2.30) ν_{1112}	Calculated from eqs. (2.29) & (2.30) ν_{1122}	Calculated from eqs. (2.29) & (2.30) ν_{2221}	AAD
n-Octane(1)	293.15	1.196	1.435	1.972	1.109	1.481	1.972	0.3
-n-Tridecane(2)	298.15	1.055	1.388	1.792	1.038	1.374	1.818	0.4
	308.15	0.948	1.195	1.542	0.919	1.196	1.555	0.5
	313.15	0.884	1.136	1.428	0.867	1.120	1.447	0.5
n-Octane(1)	293.15	1.324	1.963	2.761	1.269	1.917	2.834	0.7
-n-Pentadecane(2)	298.15	1.248	1.811	2.507	1.209	1.764	2.574	0.8
	308.15	1.120	1.531	2.111	1.063	1.515	2.158	0.9
	313.15	1.052	1.428	1.946	1.000	1.411	1.990	1.0
n-Decane(1)	293.15	1.750	2.338	2.962	1.748	2.288	2.996	0.3
-n-Pentadecane(2)	298.15	1.628	2.122	2.688	1.612	2.090	2.711	0.4
	308.15	1.402	1.793	2.238	1.393	1.774	2.259	0.2
	313.15	1.310	1.661	2.057	1.299	1.643	2.077	0.3
n-Undecane(1)	293.15	2.046	2.539	3.090	2.033	2.512	3.104	0.3
-n-Pentadecane(2)	298.15	1.891	2.299	2.795	1.866	2.287	2.804	0.3
	308.15	1.597	1.944	2.316	1.594	1.926	2.328	0.15
	313.15	1.490	1.791	2.125	1.482	1.780	2.138	0.15

Table 5.12

Values of Parameter in McAllister Ternary Model for n-Alkane Systems

System	ν_{123}	std dev 10^{-10} m^2/s	ν_{123}	std dev 10^{-10}m^2/s
	T=293.15 K		T=298.15 K	
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	1.5364	0.56	1.4277	1.00
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.8726	1.33	1.7306	1.57
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	2.4902	1.80	2.3025	0.60
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	2.1372	1.71	1.9757	0.96
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	2.3710	2.05	2.1751	0.27
	T=308.15 K		T=313.15 K	
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	1.2394	1.42	1.1667	0.78
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.4885	1.76	1.3885	1.03
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	1.9321	0.29	1.7832	0.38
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.6719	1.43	1.5517	1.59
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	1.8382	0.30	1.7025	0.37

Table 5.13

Values of $v_{123} / (v_1 v_2 v_3)^{1/3}$ of Ternary n-Alkane Systems

System	$v_{123} / (v_1 v_2 v_3)^{1/3}$			
	293.15 K	298.15 K	308.15 K	313.15 K
C_8(1)-C_{11}(2)-C_{13}(3)	1.0625	1.0654	1.0644	1.0700
C_8(1)-C_{11}(2)-C_{15}(3)	1.1310	1.1339	1.1324	1.1324
C_{11}(1)-C_{13}(2)-C_{15}(3)	1.0222	1.0382	1.0350	1.0340
C_8(1)-C_{13}(2)-C_{15}(3)	1.1148	1.1239	1.1155	1.1147
C_{10}(1)-C_{13}(2)-C_{15}(3)	1.0515	1.0573	1.0562	1.0574

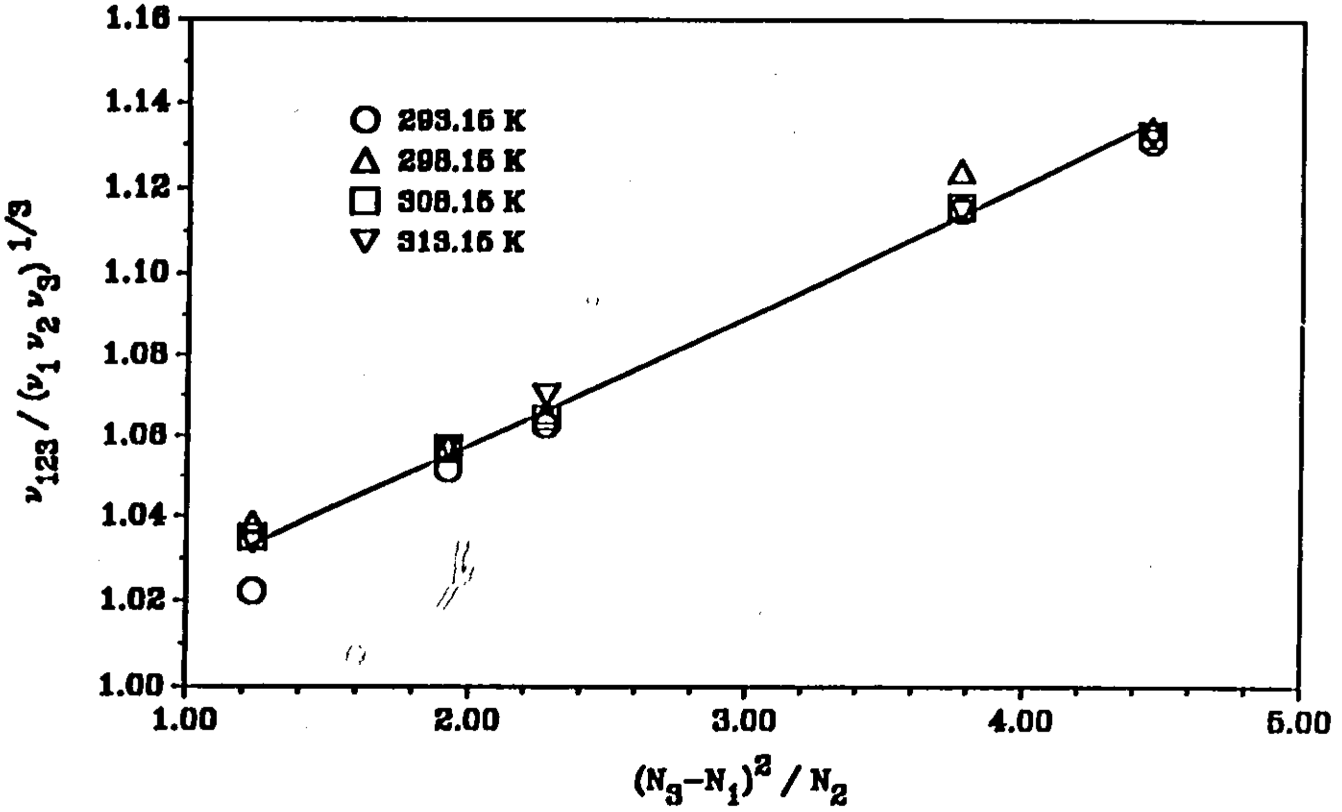


Figure 5.9. Plot of Eq. (5.13)

value of v_{123} for this system was calculated by eq. (5.13), then the calculated value of v_{123} was substituted into McAllister ternary model, eq. (2.31), to get the predicted viscosity. Figure 5.10 shows that the predicted and experimental viscosity are in very good agreement. Actually, the average absolute deviation between these values is only 0.36%.

The fact that eq. (5.13) can accurately predict v_{123} of n-hexane-n-tetradecane-n-hexadecane mixture which is beyond the carbon atom number range for which the equation was derived lends more support to the validity of the technique reported by Asfour *et al.*(1991).

5.4.3 Further extension of the McAllister model

The McAllister model would be very complicated and would possess more undetermined adjustable parameters if it is extended to multicomponent mixtures (n>3). This is because of the higher order interactions among the components. Therefore, for mixtures having more than three components, it is not practical to employ the McAllister model.

Consequently, for multicomponent mixtures, a model having reasonable accuracy and easy to use is required.

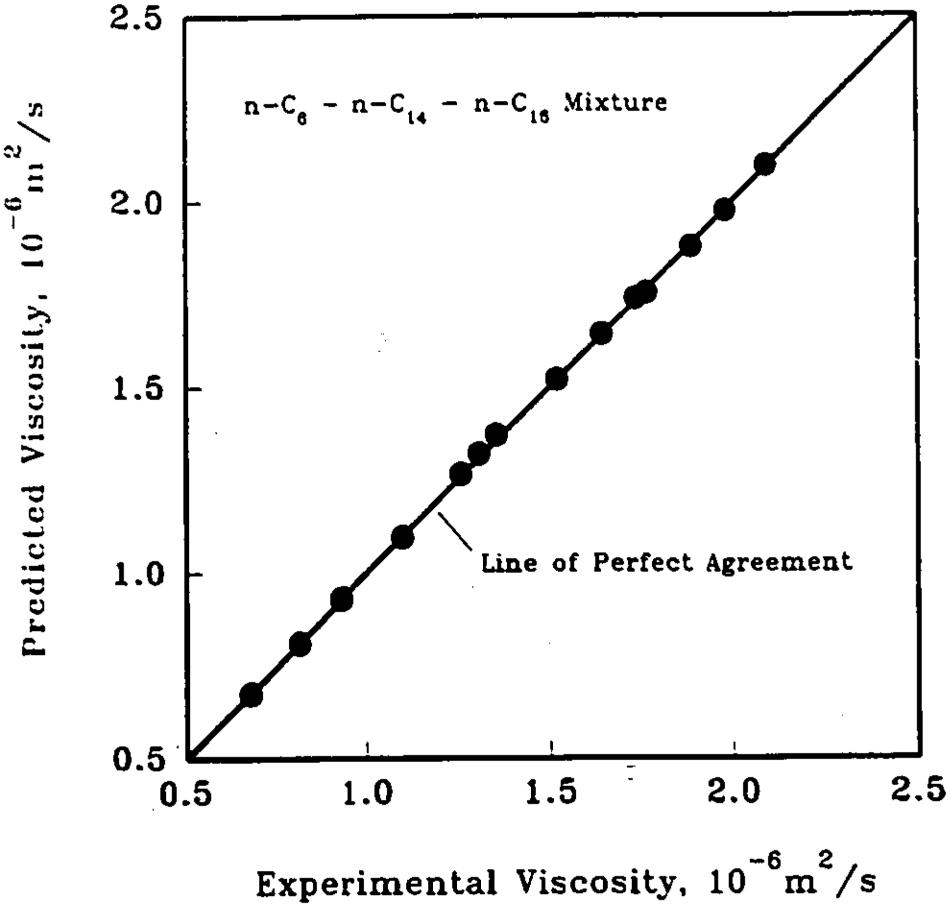


Figure 5.10. Results of Testing Eq. (5.13)

5.5 Application of the Generalized Corresponding States Principle (GCSP) to the Viscosity of n-Alkane Liquid Mixtures

The GCSP method is based on the Corresponding States Principle and the properties of two reference fluids: (i) the acentric factor, (ii) the viscosity and, (iii) the critical properties. As we pointed earlier, in Chapter 2, it gave better results than the TRAPP method in many cases. Therefore, it is explored for the specific use with n-alkane mixtures in this study. When applying the GCSP method to mixtures, appropriate mixing rules should be employed to find the pseudocritical properties of the mixture. The mixing rules proposed by Wong *et al.*(1983) which are especially useful for a large range of mixtures are used in this study. These were reported in Chapter 2, by eqs. (2.97) to (2.103).

The binary interaction coefficient, ξ_{ij}, is given by eq. (2.100). It can be calculated from experimental mixture data. The GCSP method is flexible since if some mixture viscosity data are available, ξ_{ij} can be included in the calculation, in such a case, the method becomes one of the correlation rather than prediction and if the mixture viscosity data are not available, ξ_{ij} can be set equal to unity, thus, the method becomes predictive (Teja and Rice, 1981). The predictive capability of the model is very important, because the costly and time consuming experimental work could be avoided if a reliable predictive model is available.

5.5.1 Test of GCSP using experimental binary n-alkane viscosity data

The Generalized Corresponding States Principle (GCSP) method was tested by using the viscosity data of the eight n-alkane binary mixtures at the four temperature levels. The results are reported in Table 5.14. The term MAX in the table is the maximum deviation, defined as

$$MAX = \max\{\frac{\eta_i^{calcd} - \eta_i^{expd}}{\eta_i^{expd}}\} \times 100 \qquad (5.14)$$

where MAX is a percentage.

As shown in Table 5.14, when the binary interaction coefficient ξ_{12} is set to unity, the AAD is in the range 0.15 - 3.61%, and for each system AAD increases with temperature. Whereas when ξ_{12} is included as an adjustable parameter, the range of AAD is 0.01 - 0.38%. Table 5.14 also shows that ξ_{12} is almost independent of temperature.

According to the data given in Table 5.14, when ξ_{12} is included in the GCSP method as an adjustable parameter, the AAD is remarkably reduced. Consequently, being able to predict a *priori* value of ξ_{12} would represent a significant achievement.

It was noticed from Table 5.14 that ξ_{12} increases with the difference between the carbon atom numbers of the two pure components of the mixture. A plot of ξ_{12} versus $(N_2 - N_1)^2$ shows a reasonably good linear relationship as depicted in Figure 5.11. The equation of the straight line is given by

Table 5.14

Results of Testing GCSP Method by Binary n-Alkane Viscosity Data

	$\xi_{12} = 1$		$\xi_{12} \neq 1$		
System	AAD	MAX	AAD	MAX	ξ_{12}
Temperature = 293.15 K					
$C_8(1)$-$C_{11}(2)$	0.38	-0.59	0.13	-0.29	1.0209
$C_8(1)$-$C_{13}(2)$	1.16	-1.63	0.11	0.24	1.0668
$C_8(1)$-$C_{15}(2)$	2.42	-3.46	0.38	-0.97	1.1439
$C_{10}(1)$-$C_{15}(2)$	1.34	-1.98	0.16	0.23	1.0781
$C_{11}(1)$-$C_{15}(2)$	1.00	-1.86	0.16	0.21	1.0575
$C_{13}(1)$-$C_{15}(2)$	0.21	-0.38	0.07	-0.15	1.0106
$C_{10}(1)$-$C_{13}(2)$	0.43	-0.61	0.05	-0.09	1.0238
$C_{11}(1)$-$C_{13}(2)$	0.24	-0.40	0.05	-0.12	1.0129
Temperature = 298.15 K					
$C_8(1)$-$C_{11}(2)$	0.51	-0.97	0.07	-0.28	1.0283
$C_8(1)$-$C_{13}(2)$	1.44	-1.87	0.07	0.16	1.0801
$C_8(1)$-$C_{15}(2)$	2.88	-3.87	0.28	0.54	1.1712
$C_{10}(1)$-$C_{15}(2)$	1.56	-2.11	0.09	0.16	1.0892
$C_{11}(1)$-$C_{15}(2)$	1.10	-1.72	0.10	-0.37	1.0609
$C_{13}(1)$-$C_{15}(2)$	0.26	-0.34	0.04	-0.21	1.0131
$C_{10}(1)$-$C_{13}(2)$	0.55	-0.88	0.05	-0.20	1.0307
$C_{11}(1)$-$C_{13}(2)$	0.16	-0.27	0.05	-0.10	1.0083

Table 5.14 (cont'd)

Results of Testing GCSP Method by Binary n-Alkane Viscosity Data

	$\xi_{12} = 1$		$\xi_{12} \neq 1$		
System	AAD	MAX	AAD	MAX	ξ_{12}
		Temperature = 308.15 K			
$C_8(1)$-$C_{11}(2)$	0.68	-1.12	0.06	-0.18	1.0386
$C_8(1)$-$C_{13}(2)$	1.87	-2.35	0.15	-0.76	1.1021
$C_8(1)$-$C_{15}(2)$	3.28	-4.33	0.22	0.37	1.1939
$C_{10}(1)$-$C_{15}(2)$	1.53	-2.10	0.11	0.17	1.0872
$C_{11}(1)$-$C_{15}(2)$	1.00	-1.39	0.06	0.14	1.0565
$C_{13}(1)$-$C_{15}(2)$	0.32	-0.42	0.02	-0.06	1.0172
$C_{10}(1)$-$C_{13}(2)$	0.50	-0.69	0.04	-0.07	1.0280
$C_{11}(1)$-$C_{13}(2)$	0.21	-0.33	0.04	0.08	1.0118
		Temperature = 313.15 K			
$C_8(1)$-$C_{11}(2)$	0.71	-1.12	0.07	0.14	1.0404
$C_8(1)$-$C_{13}(2)$	1.89	-2.60	0.09	0.22	1.1081
$C_8(1)$-$C_{15}(2)$	3.61	-4.62	0.21	0.31	1.2085
$C_{10}(1)$-$C_{15}(2)$	1.64	-2.21	0.09	0.14	1.0927
$C_{11}(1)$-$C_{15}(2)$	1.01	-1.62	0.11	-0.34	1.0574
$C_{13}(1)$-$C_{15}(2)$	0.24	-0.34	0.01	0.04	1.0134
$C_{10}(1)$-$C_{13}(2)$	0.56	-0.75	0.01	-0.02	1.0306
$C_{11}(1)$-$C_{13}(2)$	0.15	-0.26	0.04	0.09	1.0085

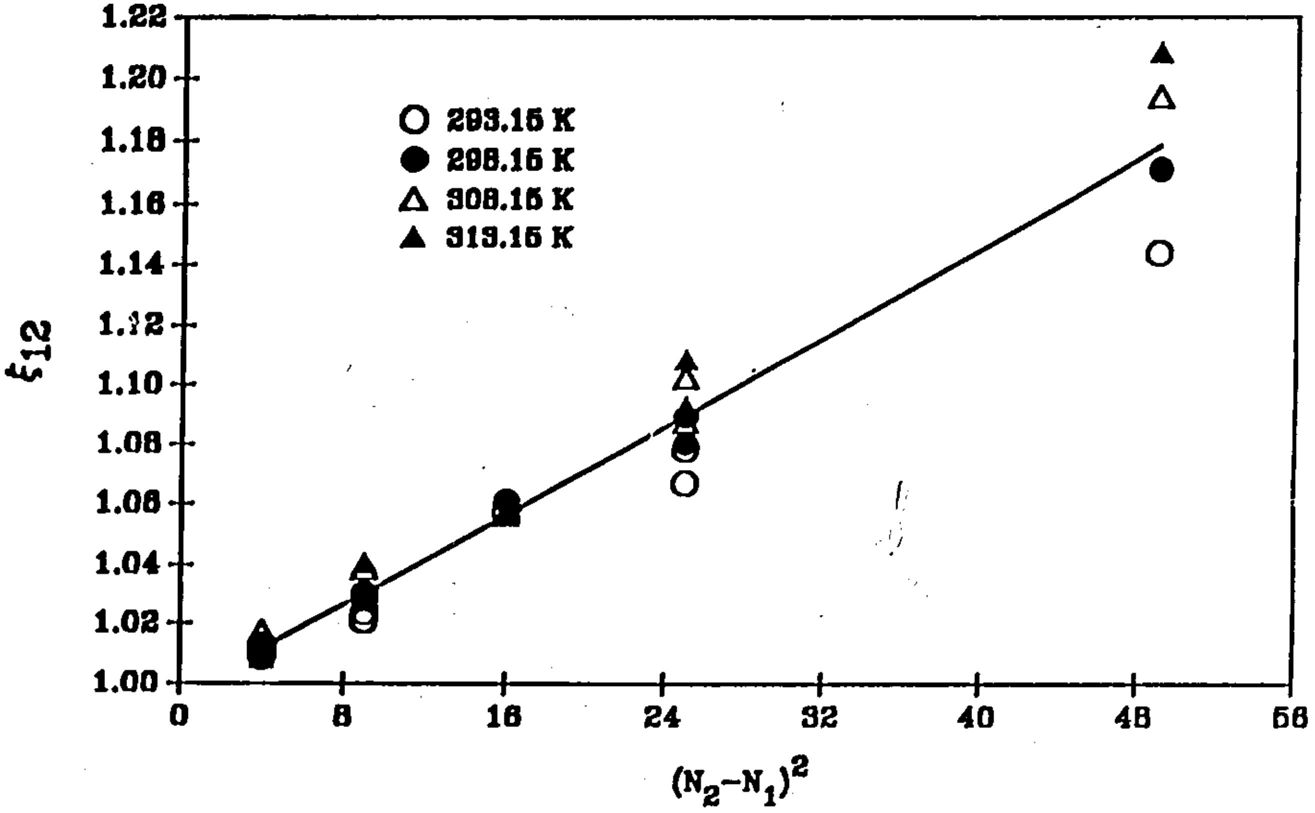


Figure 5.11. Plot of Eq. (5.15)

$$\xi_{12} = A_o + A_1(N_2 - N_1)^2 \tag{5.15}$$

where A_o and A_1 were calculated from the ξ_{12} values of eight systems at four temperatures (32 points) listed in Table 5.14. A_o is 0.9969 and A_1 is 0.0037, the standard deviation of the fit is 0.012. Eq. (2.100) hence has the form for binary n-alkane liquid mixtures

$$T_{cij} = [0.9969 + 0.0037(N_2 - N_1)^2](T_{cii} + T_{cii})^{1/2} \tag{5.16}$$

The GCSP method generalized by eq. (5.15) was tested by using literature data (Chevalier *et al.*, 1990; Heric *et al.*, 1967; Wakefield, 1987, 1988; Wakefield *et al.*, 1988). These data are over the entire composition range of each system. The values of AAD are given in Table 5.15 for the cases when $\xi_{12} = 1$ and $\xi_{12} \neq 1$. It is obvious that the value of ξ_{12} calculated from eq. (5.15) gave much smaller AAD compared with the case when ξ_{12} is set equal to 1. Therefore, it is possible to utilize the approach proposed here for improving the performance of the GCSP method for binary n-alkane systems without need for experimental data.

5.5.2 Test GCSP using experimental ternary n-alkane viscosity data

Application of GCSP to mixtures of more than two components requires appropriate choice of the reference fluids, r1 and r2. Since the number of combinations of 2 elements from 3 elements is 3, the number of combinations of 2 elements from 4 elements is 6 and so on.

The results of applying GCSP to the five ternary n-alkane liquid mixtures at four

Table 5.15

Testing Eq. (5.15) with Literature Data of Some n-Alkane Systems

System	No. of Points	Temp. K	Pressure atm	GCSP AAD (ξ_{12}=1)	GCSP AAD (ξ_{12}≠1)*
$C_6(1)$-$C_{10}(2)$ [1]	9	298	1	1.78	0.76
$C_6(1)$-$C_{14}(2)$ [2]	10	298	1	3.92	0.63
$C_6(1)$-$C_{16}(2)$ [2]	20	298	1	6.91	1.08
$C_8(1)$-$C_{16}(2)$ [3]	7	318	1	3.55	1.23
$C_8(1)$-$C_{16}(2)$ [3]	7	328	1	4.06	1.06
$C_8(1)$-$C_{16}(2)$ [3]	7	338	1	3.65	0.96

* ξ_{12} calculated from eq. (5.15)

[1] Chevalier *et al.*, 1990

[2] Heric and Brewer, 1967

[3] Wakefield and Marsh, 1987

temperatures with different combinations of reference fluids are reported in Table 5.16. The binary interaction coefficient, ξ_{ij}, was set equal to unity. Because in the case of ternary mixtures, there are three binary coefficients, ξ_{12}, ξ_{13} and ξ_{23}, it would be difficult to determine all of them if only limited experimental data were available. The data in Table 5.16 indicate that the accuracy of prediction differs with the different combinations of r1 and r2, but since the value of MAX and that of AAD do not show large differences, the effect of composition of the ternary mixture on the prediction accuracy is not as significant as the selection of the reference fluids. In order to see the trend clearer, the data in Table 5.16 were plotted in Figures 5.12, 5.13, 5.14 and 5.15. The three different combinations of r1 and r2 are represented by different boxes. As can be seen from these figures, the average absolute percent deviation (AAD) of prediction is a minimum when r1 and r2 are chosen according to

$$N^{r2} - N^{r1} = \min \{N_j - N_i\} \qquad (5.17)$$

$$i<j, \quad i=1,2,...n, \quad j=1,2,...n$$

where N^{r1} is the number of carbon atoms of reference fluid 1, N^{r2} is that of reference fluid 2 and n is the total number of components in the system. Eq. (5.17) indicates that for ternary n-alkane mixtures when the two components having the minimum difference in the number of carbon atoms were chosen as reference fluids, the AAD is the smallest.

Table 5.16

Testing GCSP (ξ_{ij}=1) with Experimental Ternary n-Alkane Viscosity Data

System	r1=1, r2=2		r1=1, r2=3		r1=2, r2=3	
	AAD	MAX	AAD	MAX	AAD	MAX
Temperature = 293.15 K						
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.40	0.69	1.16	-1.42	0.34	1.41
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	0.84	1.74	2.45	-3.14	1.08	-1.69
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.35	0.68	0.59	-0.83	0.27	-0.46
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.35	-2.15	2.20	-2.92	0.71	1.79
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.62	-1.00	1.12	-1.50	0.41	-0.76
Temperature = 298.15 K						
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.51	-0.90	1.14	-1.66	0.40	1.37
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.01	1.89	2.90	-3.52	1.23	-1.93
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.50	-0.76	0.90	-1.15	0.50	-0.76
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.64	-2.70	2.72	-3.44	0.99	1.88
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.65	-1.18	1.37	-1.77	0.55	-0.88
Temperature = 308.15 K						
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.69	-1.52	1.82	-2.35	0.36	1.34
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.07	2.52	3.30	-3.94	1.23	-1.92
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.54	-0.81	0.86	-1.13	0.55	-0.82
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.88	-2.70	3.08	-3.87	1.06	1.60
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.77	-1.19	1.40	-1.85	0.64	-0.98
Temperature = 313.15 K						
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.72	1.60	1.96	-2.50	0.45	2.35
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.11	3.32	3.47	-4.19	1.31	-1.98
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.45	-0.74	0.79	-1.10	0.51	-0.83
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.95	-3.24	3.19	-4.06	1.08	1.56
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.81	-1.25	1.48	-1.94	0.67	-1.03

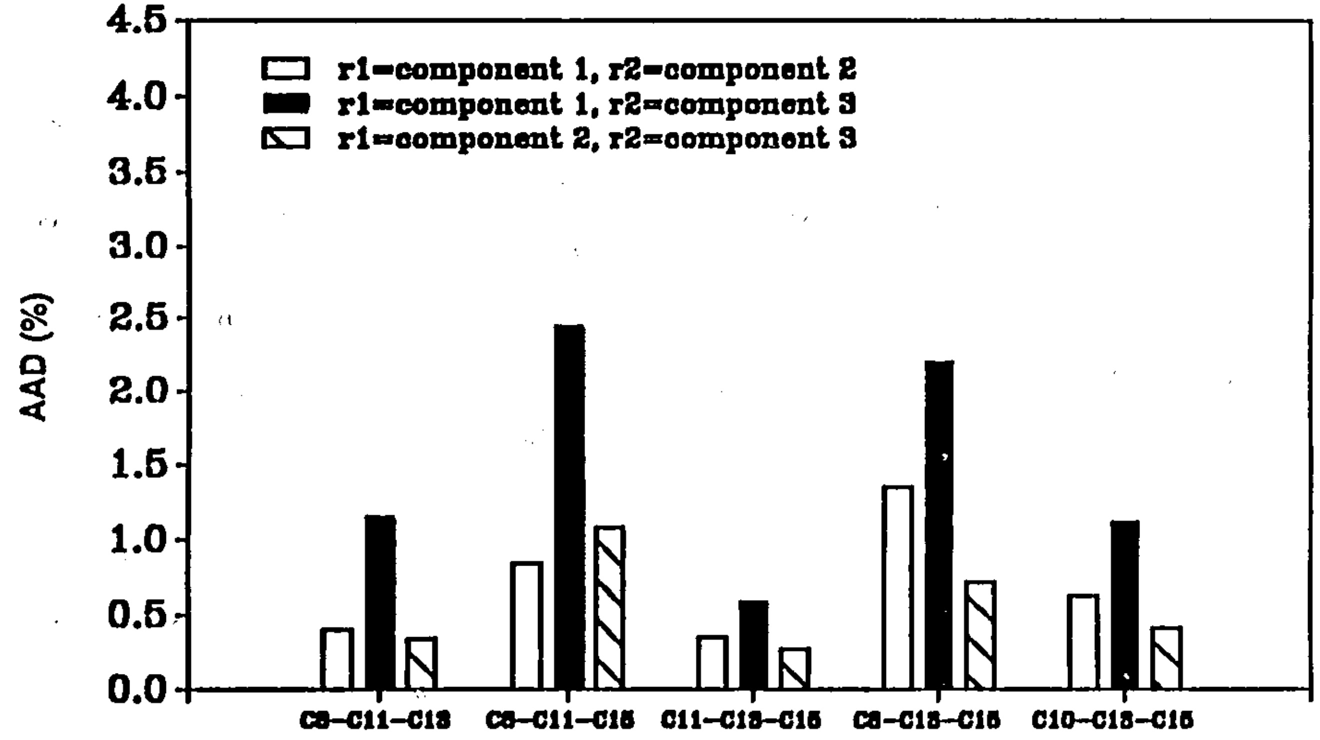


Figure 5.12. Results of Applying GCSP to n-Alkane Ternary Systems at 293.15 K

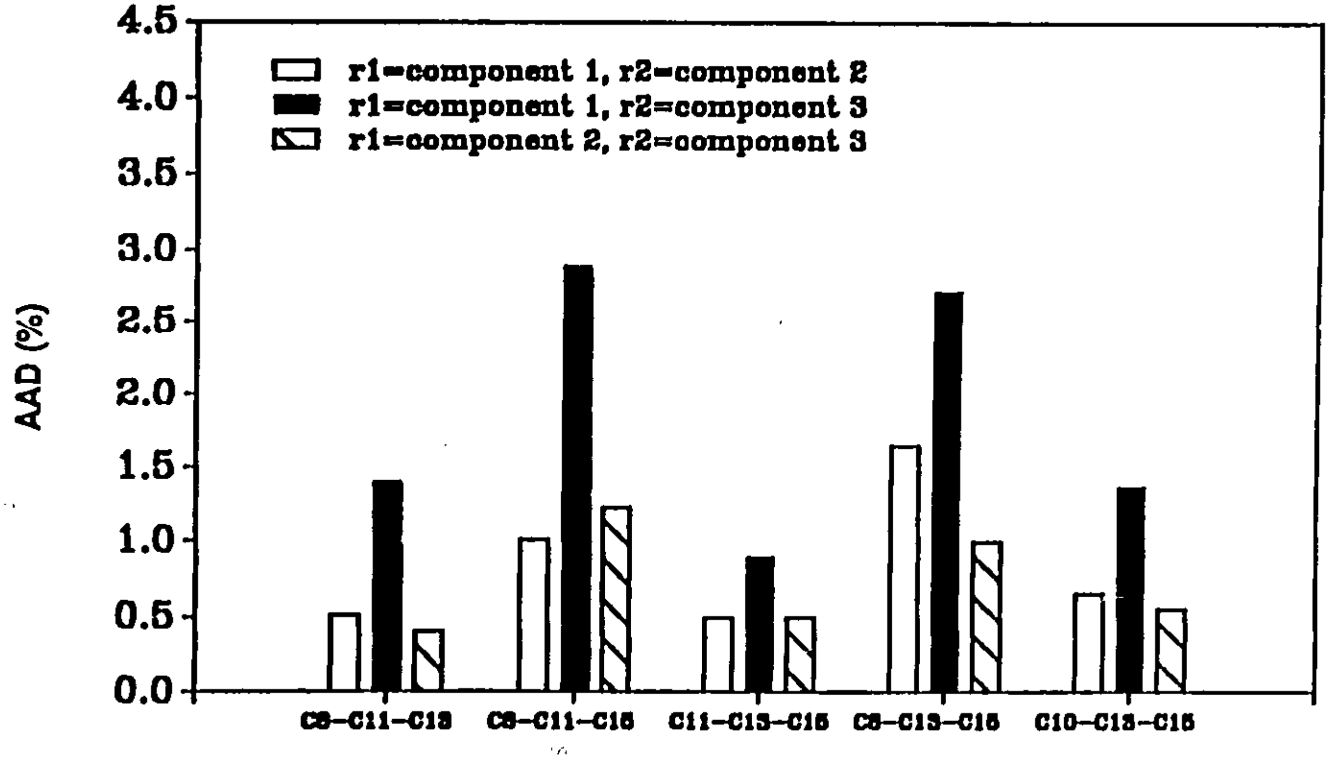


Figure 5.13. Results of Applying GCSP to n-Alkane Ternary Systems at 298.15 K

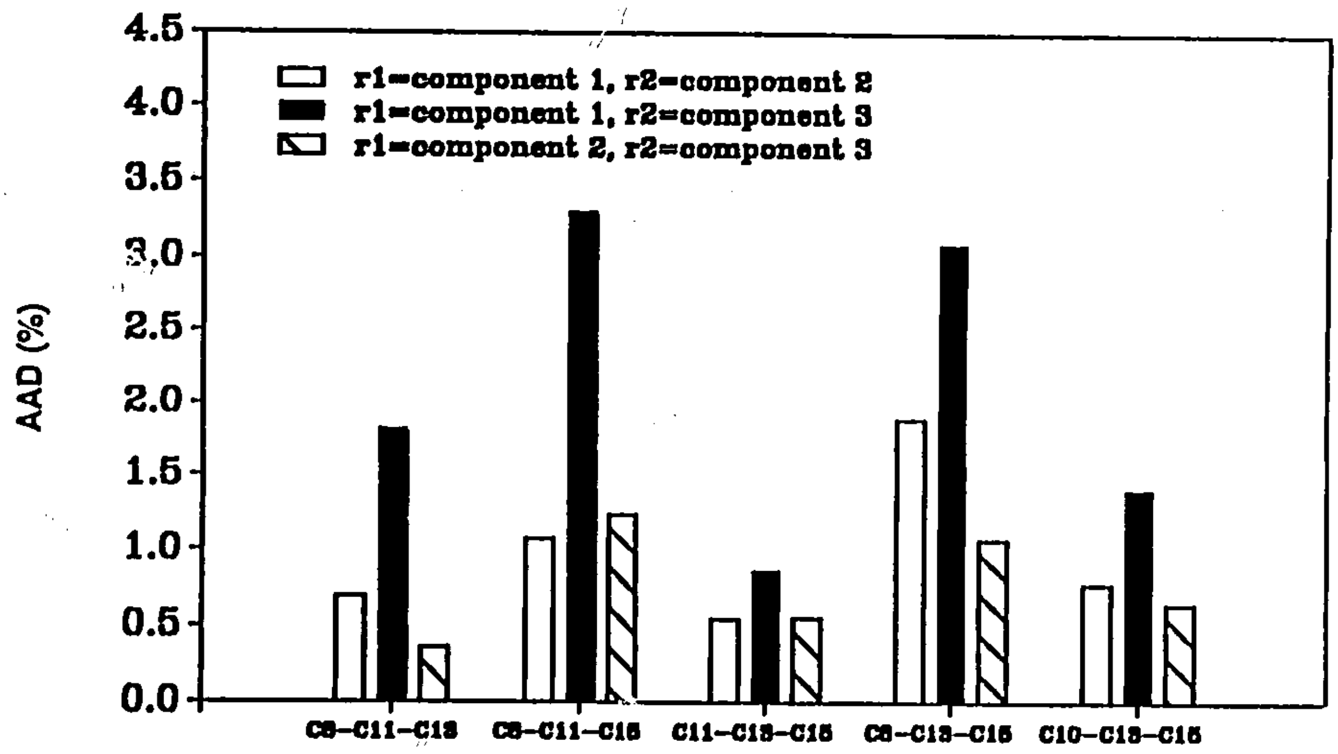


Figure 5.14. Results of Applying GCSP to n-Alkane Ternary Systems at 308.15 K

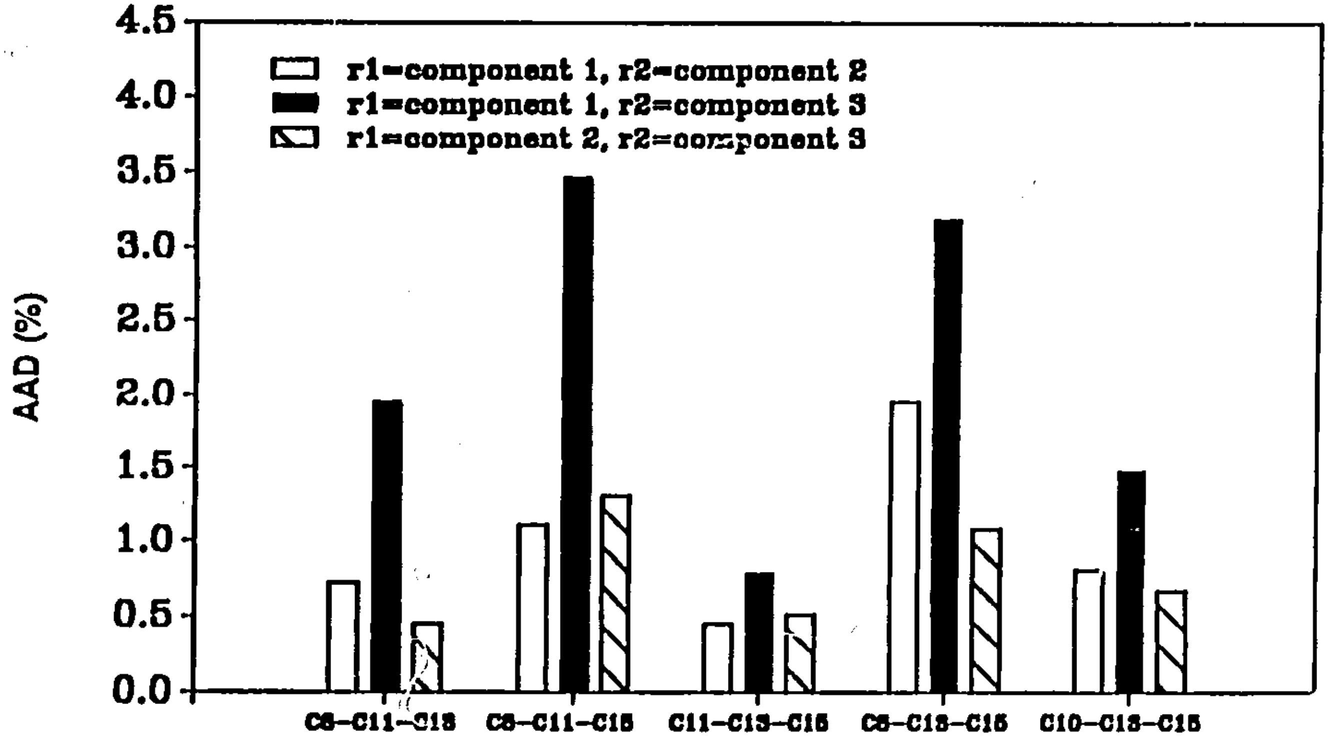


Figure 5.15. Results of Applying GCSP to n-Alkane Ternary Systems at 313.15 K

5.5.3 Test of the GCSP method using literature n-alkane quaternary viscosity data

The GCSP method was also tested by some n-alkane quaternary viscosity data taken from the literature (Wakefield, 1988). Two quaternary systems were used at 303 and 308 K. These are n-heptane(1)-n-nonane(2)-n-dodecane(3)-n-hexadecane(4) and n-hexane(1)-n-decane(2)-n-dodecane(3)-n-hexadecane(4). These data are the only n-alkane quaternary viscosity data reported in the literature. The binary interaction coefficients, ξ_{ij} were set equal to unity. The results for the two systems are shown in Figures 5.16 and 5.17, respectively. Different bars represent different temperatures and the numbers above the bars are their values of maximum error (MAX). Each quaternary mixture has six combinations of r1 and r2. The accuracy of prediction from each combination differs significantly. Unlike the case of ternary systems, the values of the maximum error for several combinations of r1 and r2 are relatively high. For example, for the system C_7-C_9-C_{12}-C_{16} at 303 K, when component 3 was chosen as r1 and component 4 as r2, the value of MAX is 18.64%. Such a high deviation value is an indication of the very poor prediction capability of mixture viscosity. Thus, the effect of the selection of reference fluids is more significant on quaternary n-alkane systems than on ternary systems. From Figure 5.16, it seems that eq. (5.17) is also applicable to quaternary n-alkane systems, since the minimum AAD of prediction was obtained when component 1 (C_7) was chosen as r1 and component 2 (C_9) as r2, and these two components have the minimum difference in the number of carbon atoms. But as can be seen from Figure 5.17, the expected best combination of r1=C_{10} and r2=C_{12}, i.e. the combination with the two

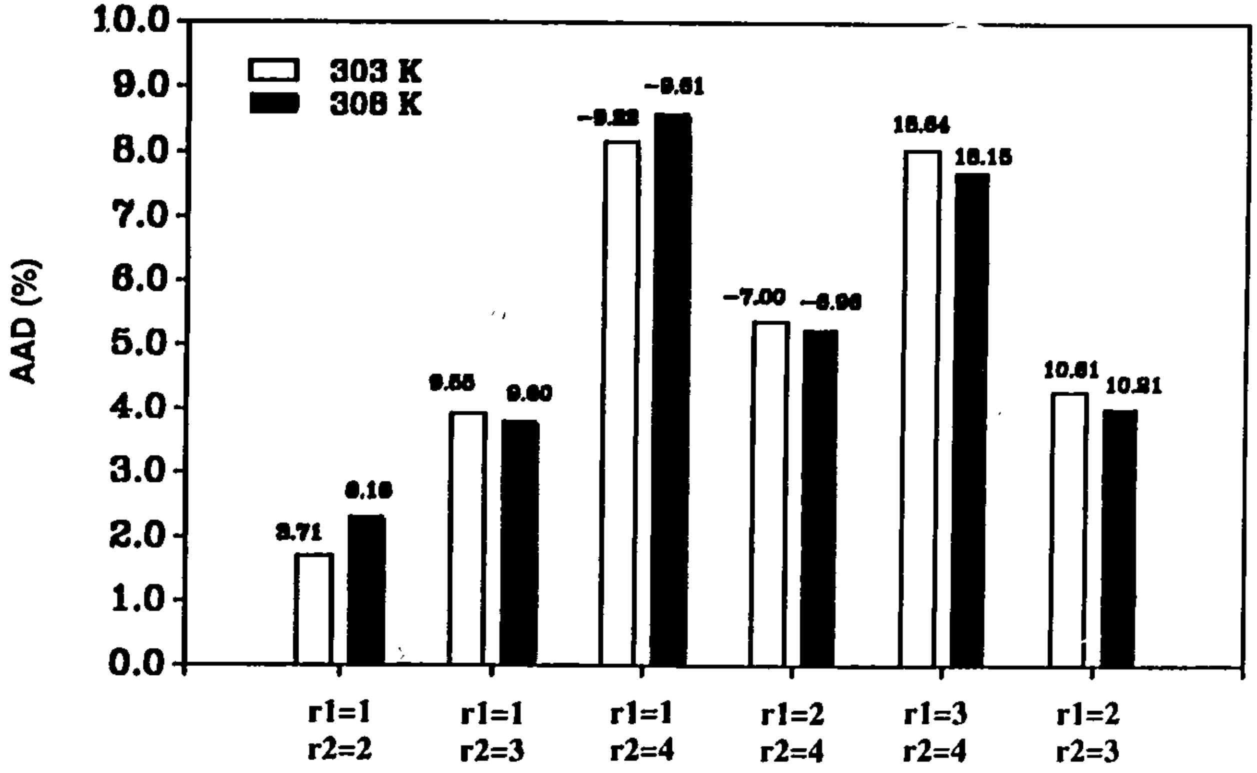


Figure 5.16. Results of Applying GCSP to C_7 - C_9 - C_{12} - C_{16} n-Alkane Quaternary System

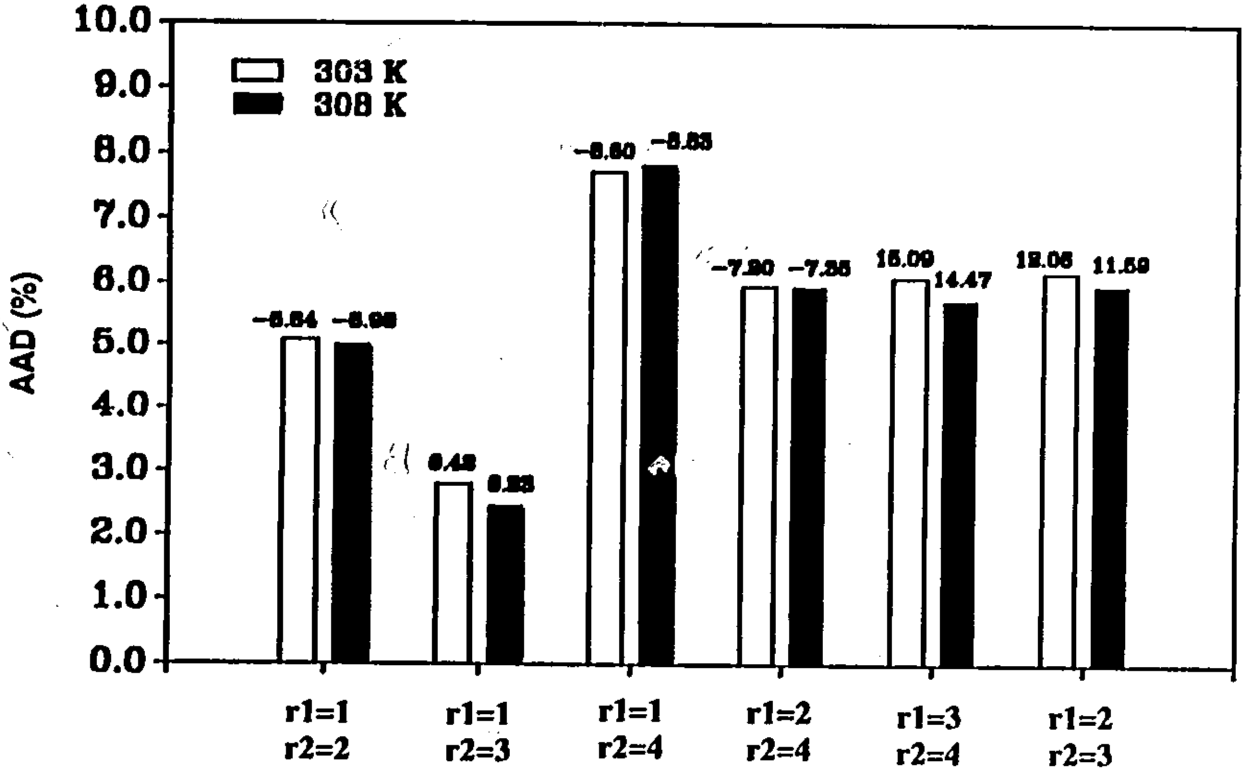


Figure 5.17. Results of Applying GCSP to C_6 - C_{10} - C_{12} - C_{16} n-Alkane Quaternary System

reference fluids having the minimum difference in carbon atom numbers, gave 6% AAD and 12% MAX at both temperatures, whereas the combination r1 = C_6 and r2 = C_{12} gave the minimum AAD, 3%, and 6.5% MAX. Therefore, eq. (5.17) cannot be used in quaternary n-alkane mixtures as a standard for choosing appropriate reference fluids.

The reason is probably that if two of the four components were chosen as r1 and r2 when the concentrations of the other two components are predominant, the mixture and the reference fluids have little similarity. Unfortunately, it is this similarity that provides the GCSP with the capability of predicting mixture viscosity. Therefore, in selecting reference fluids for quaternary mixtures, not only the difference of carbon atom numbers of the reference fluids is important, but also the concentration of the mixture should be considered.

The percent deviations given by the GCSP for the two quaternary mixtures, indicated above, at 303 K at different compositions are shown in Tables 5.17 and 5.18, respectively. The results of the two mixtures at 308 K have the same trends as those at 303 K as shown in Figures 5.16 and 5.17. Therefore, they have not been listed. One can conclude on the basis of the data reported in Tables 5.17 and 5.18 that when two of the four components have predominant concentrations, no high values of MAX are obtained if they are chosen as reference fluids, but such a selection does not guarantee obtaining minimum error. In other words, non-selection of the two components which have predominant concentrations as reference fluids in the quaternary mixture could cause very large errors, but selecting these two components does not necessarily give the best results although it can avoid the large values of deviation. It is a rather complicated situation.

Table 5.17

The Values of Percent Deviation of Testing GCSP ($\xi_{ij}=1$) with the Viscosity Data of n-Alkane Quaternary Mixture $C_7(1)$-$C_9(2)$-$C_{12}(3)$-$C_{16}(4)$ at 303.16 K

			Percent Deviation (%)					
x_1	x_2	x_3	r1=1 r2=2	r1=1 r2=3	r1=1 r2=4	r1=2 r2=4	r1=3 r2=4	r1=2 r2=3
0.07483	0.07466	0.42552	3.71	9.55	-8.58	-6.98	0.38	10.61
0.17416	0.17445	0.32635	0.77	5.44	-9.22	-7.00	3.40	5.67
0.25004	0.24921	0.25028	-0.58	3.26	-8.87	-6.14	6.85	2.87
0.32462	0.32501	0.17506	-1.62	1.38	-8.18	-4.89	10.98	0.38
0.42475	0.42538	0.07493	-1.86	0.001	-6.01	-1.86	18.64	-1.86

note: the data were taken from Wakefield, 1988.

Table 5.18

The Values of Percent Deviation of Testing GCSP ($\xi_{ij}=1$) with the Viscosity Data of n-Alkane Quaternary Mixture $C_6(1)$-$C_{10}(2)$-$C_{12}(3)$-$C_{16}(4)$ at 303.16 K

			Percent Deviation (%)					
x_1	x_2	x_3	r1=1 r2=2	r1=1 r2=3	r1=1 r2=4	r1=2 r2=4	r1=3 r2=4	r1=2 r2=3
0.08312	0.07472	0.42175	-4.43	6.42	-8.01	-6.99	-0.22	12.06
0.17435	0.17551	0.32506	-5.53	3.65	-8.60	-7.20	2.11	4.91
0.24928	0.24931	0.25090	-5.84	2.06	-8.50	-6.79	4.75	-0.05
0.32880	0.32389	0.17333	-5.68	0.88	-7.91	-5.85	8.27	-4.69
0.42524	0.42483	0.07514	-3.90	0.95	-5.57	-2.98	15.09	-9.13

note: the data were taken from Wakefield, 1988

There are mainly two cases with regard to the compositions of the mixture:

(i) In the case where the mole fractions of each component being approximately equal, the reference fluids can be chosen according to eq. (5.17). For example, when the mole fractions of the four components are all around 0.25, as shown in Table 5.17, the minimum deviation of -0.58% appeared when r1 = C_7 (component 1) and r2 = C_9 (component 2). Here the pair C_7 - C_9 has the minimum carbon atom number difference. Another example is that when the mole fractions of the four components are all around 0.25, as shown in Table 5.18, the minimum deviation of -0.05% was obtained when r1=C_{10} (components 2) and r2=C_{12} (component 3). Here again, the pair C_{10} - C_{12} has the minimum carbon atom number difference.

(ii) In the case where the mole fractions of each component are different, the selection of the reference fluids depends on both the kinds of components and the concentrations of such components as well as some unknown factors.

Therefore, it is really difficult to get correct predictions of the viscosity of quaternary mixtures by the GCSP.

For n-alkane mixtures with more than 4 components, the number of all possible combinations of r1 and r2 increases rapidly. Therefore, the selection of the best combination of r1 and r2 becomes more difficult and the effects of mixture concentration could be more complicated. Obviously, the probability of obtaining incorrect estimation of mixture viscosity will be higher. Therefore, it is desirable to develop a simpler method with reasonably accurate capability in predicting the viscosity of multicomponent n-alkane liquid mixtures.

5.5.4 Proposed modification of the GCSP method

To overcome the difficulties encountered with the existing GCSP method, which were discussed earlier, a pseudo-binary mixture model based on the Corresponding States Principle and the Congruence Principle is thus proposed in this study. It is a modification of the GCSP, which is especially useful for n-alkane mixtures with more than three components.

In this model, a multicomponent mixture is considered as a binary system consisting of pure component 1 and pseudopure component 2', where component 2' is a mixture of components 2, 3, ...n. The reference fluids in this case are r1=1 and r2=2'. Therefore, the acentric factor, the critical properties and the reduced viscosity of component 2' (as r2) are required. To do this in the special case of n-alkane liquid mixtures, the corresponding properties of the pure n-alkanes were first investigated. The acentric factor, ω, the critical temperature, T_c and the critical pressure, P_c, of the pure n-alkanes from C_5 to C_{17} which were taken from Reid *et al.* (1977) are listed in Table 5.19. The values of the absolute viscosities, η, of C_5 to C_{17} at several temperature levels taken from

Table 5.19

Physical Properties of Some Pure n-Alkanes Used in Pseudo-binary Mixture Model*

Compound	Molecular Weight	Critical Temperature K	Critical Pressure atm	Acentric Factor
n-Pentane	72.151	469.6	33.3	0.251
n-Hexane	86.178	507.4	29.3	0.296
n-Heptane	100.205	540.2	27.0	0.351
n-Octane	114.232	568.8	24.5	0.394
n-Nonane	128.259	594.6	22.8	0.444
n-Decane	142.286	617.6	20.8	0.490
n-Undecane	156.313	638.8	19.4	0.535
n-Dodecane	170.340	658.3	18.0	0.562
n-Tridecane	184.367	675.8	17.0	0.623
n-Tetradecane	198.394	694.0	16.0	0.679
n-Pentadecane	212.421	707.0	15.0	0.706
n-Hexadecane	226.448	717.0	14.0	0.742
n-Heptadecane	240.475	733.0	13.0	0.770

* data were taken from "The Properties of Gases and Liquids", Reid *et al.*, 1977

TRC Table (1988) are listed in Table 5.20. All these data were then plotted against their chain length, N (same as the number of carbon atoms). The plots of ω vs. N, T_c vs. N and P_c vs. N are shown in Figures 5.18, 5.19 and 5.20, respectively. The logarithm of the reduced viscosity, ln(ηε), was plotted versus N in Figure 5.21. All the figures show smooth curves. This means that these properties can be well represented by the polynomials of their chain lengths. The constants of these polynomials can be calculated by the least squares technique. Then according to the Congruence Principle (Brønsted and Koefoed, 1946), the properties of the n-alkane mixtures depend on the average chain length, which is given by

$$\bar{N} = \sum_i x_i N_i \tag{5.18}$$

therefore, it is reasonable to assume that the same polynomials derived from the properties of the pure n-alkanes can be used to estimate the same properties of the pseudopure component 2', with only the chain length of the pure n-alkane, N, replaced by the average chain length of 2', $\bar{N}$. Thus, the following polynomials are obtained:

$$\omega^{r2} = \sum_i A_i \bar{N}^i \tag{5.19}$$

$$T_{c,r2} = \sum_i B_i \bar{N}^i \tag{5.20}$$

$$P_{c,r2} = \sum_i C_i \bar{N}^i \tag{5.21}$$

and at each designated temperature

Table 5.20

Pure n-Alkane Data Used in Pseudo-binary Mixture Model

Compound	Absolute Viscosity* (10^{-3} Pa.s)				
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K
n-Pentane	0.234	0.224	0.215	0.206	
n-Hexane	0.3117	0.2976	0.2845	0.2724	0.2611
n-Heptane	0.4169	0.3955	0.3761	0.3581	0.3416
n-Octane	0.5450	0.5136	0.4850	0.4591	0.4355
n-Nonane	0.7139	0.6676	0.6260	0.5885	0.5545
n-Decane	0.9256	0.8588	0.7994	0.7465	0.6989
n-Undecane	1.185	1.092	1.009	0.9357	0.8707
n-Dodecane	1.503	1.374	1.261	1.163	1.078
n-Tridecane	1.880	1.706	1.555	1.424	1.310
n-Tetradecane	2.335	2.104	1.904	1.734	1.585
n-Pentadecane	2.863	2.560	2.303	2.084	1.894
n-Hexadecane	3.474	3.086	2.758	2.482	2.243
n-Heptadecane	4.196	3.700	3.286	2.938	2.642

* data were taken from TRC Tables, 1988.

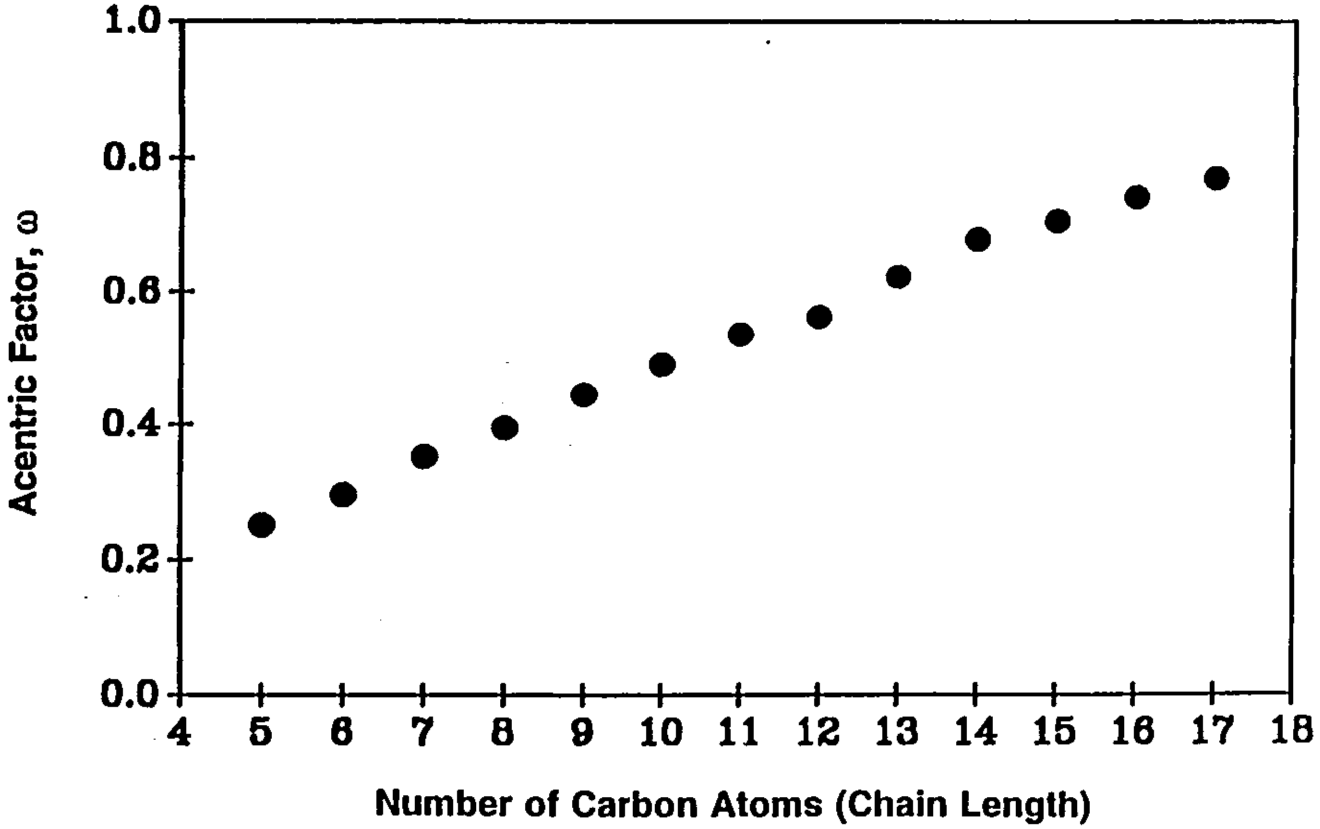


Figure 5.18. Acentric Factor vs. Chain Length for Pure n-Alkanes

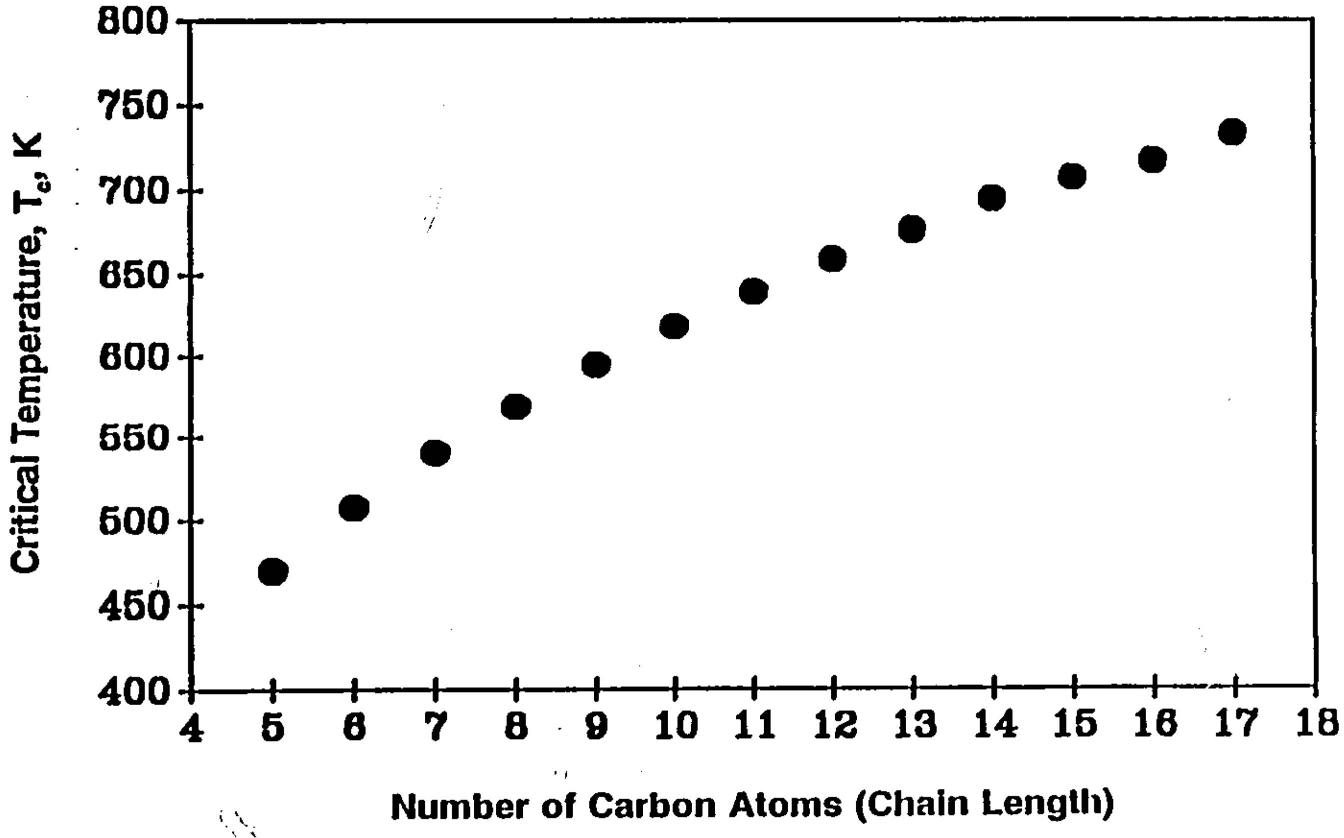


Figure 5.19. Critical Temperature vs. Chain length for Pure n-Alkanes

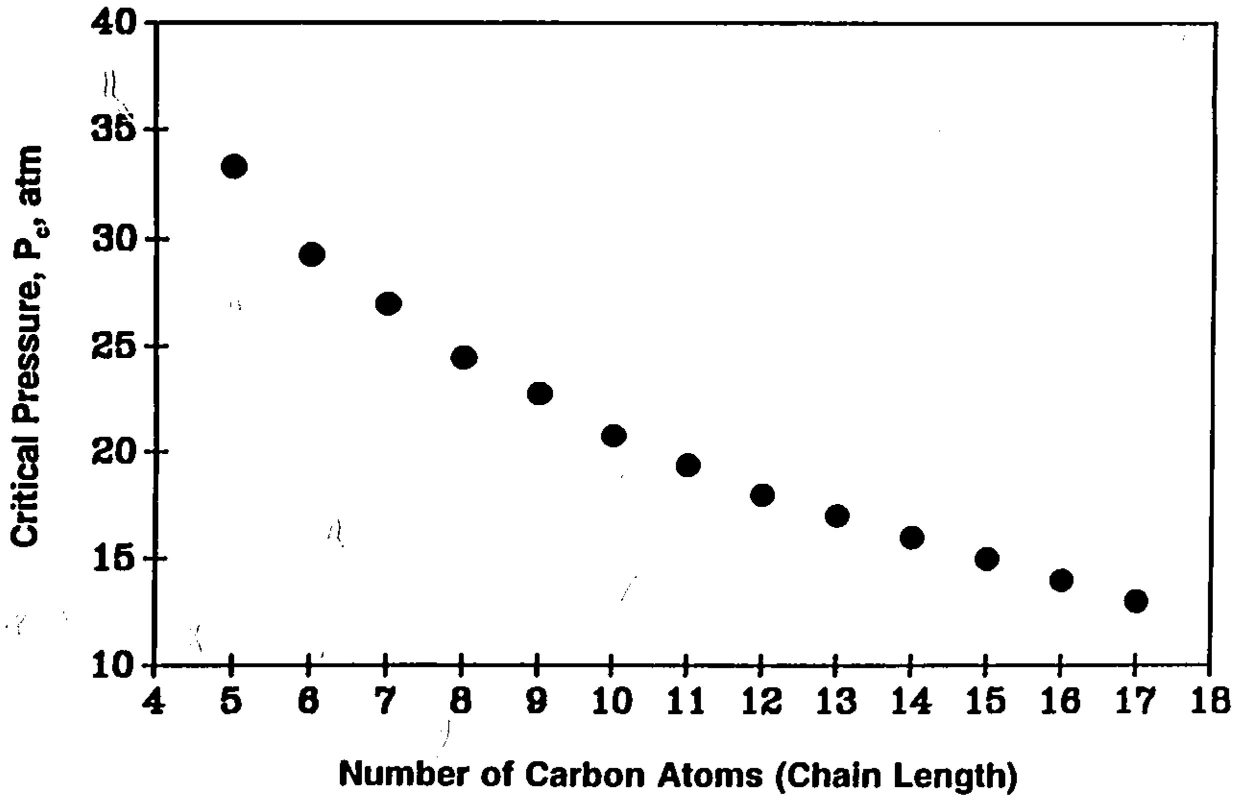


Figure 5.20. Critical Pressure vs. Chain Length for Pure n-Alkanes

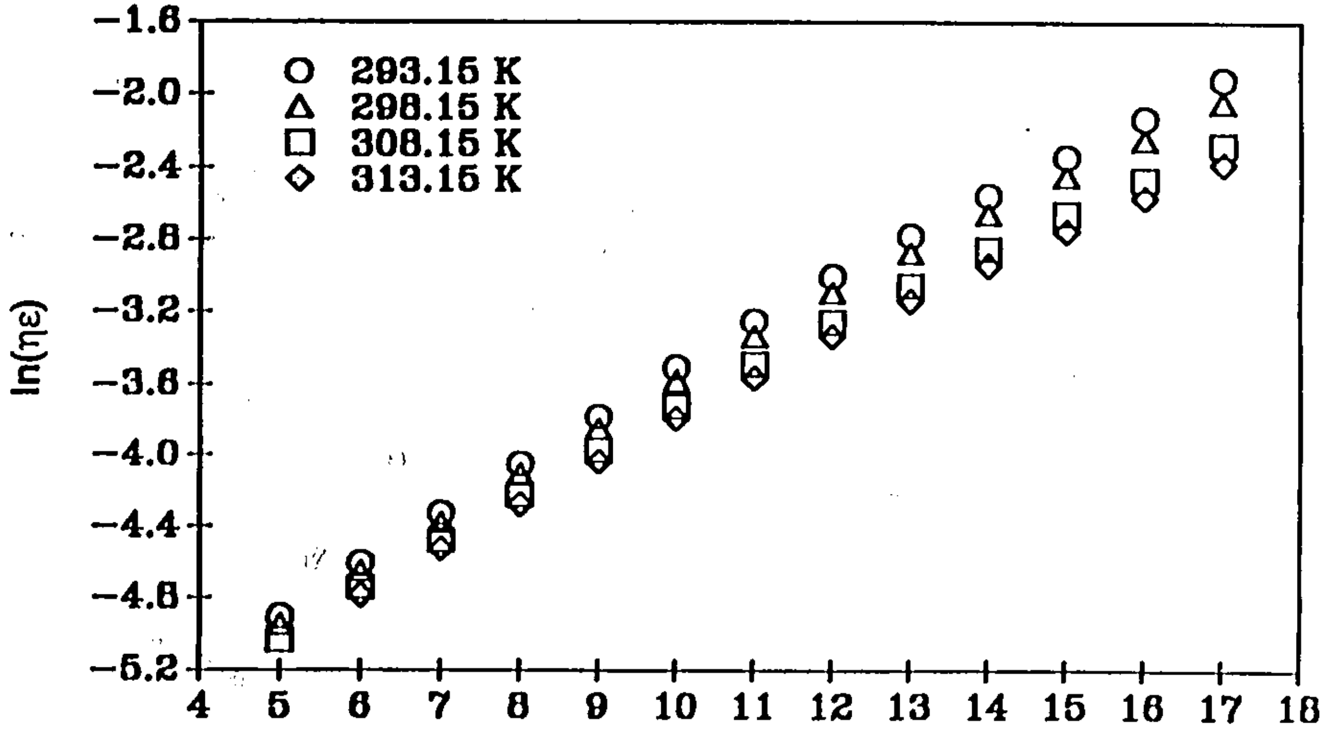


Figure 5.21. Logarithm of Reduced Viscosity vs. Chain Length for Pure n-Alkanes

$$\ln(\eta\epsilon)^{r2} = \sum_i D_i \bar{N}^i \tag{5.22}$$

The values of A_i, B_i and C_i in eqs. (5.19), (5.20) and (5.21) are listed in Table 5.21, they are calculated from the data in Table 5.19. Because ω_i, T_{ci} and P_{ci} are all temperature independent, A_i, B_i and C_i are also temperature independent. Eqs. (5.19), (5.20) and (5.21) can be used for any n-alkane liquid mixtures at any temperature level.

The values of D_i at 293.15, 298.15, 303.15, 308.15 and 313.15 are listed in Table 5.22. D_i are temperature dependent because the viscosity is temperature dependent. Since the viscosities, the critical properties and the molecular weights of the pure n-alkanes are available in the literature, then it is easy to calculate D_i at the temperatures of interest.

The values of all the constants above were calculated by the method of least squares. The order of the polynomials was selected as the one beyond which additional terms did not significantly improve the fit.

By considering the multicomponent n-alkane mixtures as the pseudo-binary mixtures and using eqs. (5.19) to (5.22) to calculate ω^{r2}, $T_{c,r2}$, $P_{c,r2}$ and $\ln(\eta\epsilon)^{r2}$, the modified-GCSP is ready to use.

5.5.5 Application of the modified-GCSP method

For the five ternary n-alkane liquid mixtures, the values of AAD and MAX of calculating the absolute viscosity by the proposed modification of the GCSP method in

Table 5.21

Least-Squares Constants for the Equations (5.19), (5.20) and (5.21)

	i=0	i=1	i=2	i=3
A_i	0.03990659	0.04422527		
B_i	226.48701	60.8026973	-2.63788711	0.04772727
C_i	58.584615	-6.898976	0.40398352	-0.00917832

Table 5.22

Least-Squares Constants for Equation (5.22)

T °K	D_o	D_1	D_2
293.15	-6.521234	0.344638	-0.00437817
298.15	-6.522899	0.336758	-0.00432932
303.15	-6.523873	0.329475	-0.00430982
313.15	-6.543745	0.325592	-0.00440440
318.15	-6.532410	0.316683	-0.00428502

the case of both $\xi_{ij} = 1$ and $\xi_{ij} \neq 1$ are listed in Table 5.23. Comparison of the data in Table 5.23 with those reported in Table 5.16 shows that when $\xi_{12} = 1$, the value of AAD given by the modified-GCSP is between the maximum and minimum values given by the original GCSP, but it is very close to the minimum one. When $\xi_{12} \neq 1$, the AAD value is significantly reduced. Also, it is clear that ξ_{12} is almost independent of temperature. The adjustable parameter ξ_{12} played a very important role in improving the accuracy of the method, but it can only be determined from experimental data. For the purpose of predicting it, an equation was proposed here which is based on the analysis of the relationship between ξ_{12} and carbon atom numbers of the components of the mixture. The equation is as follows

$$\xi_{12} = B_o + B_1(N_2 - N_1) + B_2(N_3 - N_2) \tag{5.23}$$

where $B_o = 0.8367$, $B_1 = 0.0328$ and $B_2 = 0.0426$. The values of these constants were calculated from the ξ_{12} values of five ternary mixtures at four temperatures (20 points) given in Table 5.23 by the method of least squares. The standard deviation of the fit is 0.015. The results of testing eq. (5.23) by using the literature viscosity data of the ternary system n-C_6-n-C_{14}-n-C_{16} reported by Heric and Brewer (1969) are given in Table 5.24. As shown in Table 5.24, if ξ_{12} was set to unity, the modified-GCSP method gave 5.16% AAD, but if ξ_{12} was predicted by eq. (5.23), then the value of AAD is reduced to 1.46%. The results support the prediction of ξ_{12} by eq. (5.23).

For quaternary n-alkane mixtures, comparison of the modified-GCSP and the original GCSP for quaternary n-alkane mixtures C_7-C_9-C_{12}-C_{16} and C_6-C_{10}-C_{12}-C_{16} at 303.15 and 308.15 K (data were taken from Wakefield, 1988) is reported in Table 5.25. As can be

Table 5.23

Results of Using Modified-GCSP Method on Ternary n-Alkane Systems

System	$\xi_{12} = 1$		$\xi_{12} \neq 1$		
	AAD	MAX	AAD	MAX	ξ_{12}
Temperature = 293.15 K					
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.56	-0.93	0.12	0.32	1.0341
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.56	-2.40	0.29	-0.46	1.0986
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.32	1.13	0.35	1.06	0.9910
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.20	-2.11	0.30	0.95	1.0733
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.48	0.88	0.34	0.99	1.0150
Temperature = 298.15 K					
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.62	-1.08	0.16	0.37	1.0377
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.67	-2.44	0.22	0.43	1.1045
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.36	0.95	0.37	0.88	0.9917
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.35	-2.52	0.34	0.85	1.0844
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.47	0.89	0.43	0.95	1.0087
Temperature = 308.15 K					
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.80	-1.64	0.33	0.82	1.0443
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.68	-2.61	0.22	0.62	1.1051
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.57	1.29	0.46	1.10	0.9758
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.44	-2.72	0.48	1.09	1.0853
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.49	1.10	0.49	1.09	0.9988
Temperature = 313.15 K					
$C_8(1)$-$C_{11}(2)$-$C_{13}(3)$	0.88	-1.82	0.27	0.84	1.0512
$C_8(1)$-$C_{11}(2)$-$C_{15}(3)$	1.78	-2.84	0.23	0.82	1.1139
$C_{11}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.63	1.43	0.51	1.22	0.9721
$C_8(1)$-$C_{13}(2)$-$C_{15}(3)$	1.52	-2.93	0.52	1.11	1.0903
$C_{10}(1)$-$C_{13}(2)$-$C_{15}(3)$	0.50	1.08	0.50	1.09	1.0012

Table 5.24

Comparison of Using Calculated ξ_{12} Value with Using ξ_{12}=1 in Modified-GCSP for n-Alkane Ternary Mixture C_6(1)-C_{14}(2)-C_{16}(3) at 298.15 K

ξ_{12}=1				
x_3	x_2	Exptl Viscosity 10^{-3} Pa.s	Calcd Viscosity 10^{-3} Pa.s	Percent Deviation %
0.0650	0.0796	0.4631	0.4467	-3.5530
0.1028	0.1119	0.5654	0.5363	-5.1389
0.1046	0.1763	0.6538	0.6193	-5.2803
0.1521	0.1151	0.6582	0.6182	-6.0784
0.1650	0.1831	0.7862	0.7350	-6.5080
0.0878	0.3651	0.9111	0.8659	-4.9664
0.2118	0.2258	0.9491	0.8961	-5.5798
0.3274	0.0947	0.9851	0.9224	-6.3688
0.1297	0.4208	1.1144	1.0596	-4.9166
0.2648	0.2924	1.2078	1.1441	-5.2759
0.3995	0.1514	1.2787	1.2048	-5.7823
0.1105	0.5396	1.3054	1.2520	-4.0908
0.2522	0.3998	1.4001	1.3406	-4.2480
0.3777	0.2696	1.4749	1.4071	-4.5964
0.5182	0.1243	1.5625	1.4844	-4.9968
AAD=5.16%				

note: the experimental viscosity data were taken from Heric, 1969

Table 5.24 (cont'd)

Comparison of Using Calculated ξ_{12} value with Using ξ_{12}=1
in Modified-GCSP for n-Alkane Ternary Mixture $C_6(1)$-$C_{14}(2)$-$C_{16}(3)$ at 298.15 K

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

x_3	x_2	Exptl Viscosity 10^{-3} Pa.s	Calcd Viscosity 10^{-3} Pa.s	Percent Deviation %
0.0650	0.0796	0.4631	0.4620	-0.2314
0.1028	0.1119	0.5654	0.5587	-1.1807
0.1046	0.1763	0.6538	0.6471	-1.0226
0.1521	0.1151	0.6582	0.6458	-1.8813
0.1650	0.1831	0.7862	0.7688	-2.2194
0.0878	0.3651	0.9111	0.9037	-0.8131
0.2118	0.2258	0.9491	0.9356	-1.4186
0.3274	0.0947	0.9851	0.9634	-2.2083
0.1297	0.4208	1.1144	1.1008	-1.2229
0.2648	0.2924	1.2078	1.1877	-1.6641
0.3995	0.1514	1.2787	1.2508	-2.1835
0.1105	0.5396	1.3054	1.2922	-1.0110
0.2522	0.3998	1.4001	1.3830	-1.2190
0.3777	0.2696	1.4749	1.4517	-1.5729
0.5182	0.1243	1.5625	1.5315	-1.9839

AAD=1.46%

TABLE 5.25

Comparison of Modified-GCSP with GCSP for n-Alkane Quaternary Mixtures

Modified-GCSP		GCSP											
		r1=1, r2=2		r1=1, r2=3		r1=1, r2=4		r1=2, r2=4		r1=3, r2=4		r1=2, r2=3	
AAD	MAX	AAD	MAX	AAD	MAX	AAD	MAX	AAD	MAX	AAD	MAX	AAD	MAX
n-heptane(1)-n-nonane(2)-n-dodecane(3)-n-hexadecane(4) at 303.15 K													
4.21	-5.28	1.71	3.71	3.93	9.55	8.17	-9.22	5.37	-7.00	8.05	18.64	4.28	10.61
n-heptane(1)-n-nonane(2)-n-dedecane(3)-n-hexadecane(4) at 308.15 K													
4.41	-5.39	2.33	6.16	3.82	9.60	8.60	-9.61	5.26	-6.96	7.72	18.15	4.03	10.21
n-hexane(1)-n-decane(2)-n-dodecane(3)-n-hexadecane(4) at 303.15 K													
4.35	-5.46	5.08	-5.84	2.79	6.42	7.72	-8.60	5.96	-7.20	6.09	15.09	6.17	12.06
n-hexane(1)-n-decane(2)-n-dodecane(3)-n-hexadecane(4) at 308.15 K													
4.34	-5.34	5.01	-5.98	2.47	6.23	7.84	-8.83	5.96	-7.35	5.73	14.47	5.98	11.59

note: the data above were taken from Wakefield, 1988 seen, the modified-GCSP works well for the quaternary mixtures, the values of AAD of the modified-GCSP are always less than 4.5% and the values of MAX are always less than 5.5% for the two systems at both temperatures. There are no extremely high values of MAX as is the case in the original GCSP. Besides, it gives the minimum value of MAX in Table 5.25 except for the mixture C_7-C_9-C_{12}-C_{16} at 303.15 K with r1=C_7 and r2=C_9. Although the modified-GCSP does not give the smallest value of AAD, it gives the second smallest value of AAD for C_6-C_{10}-C_{12}-C_{16} mixture and the third smallest value of AAD for C_7-C_9-C_{12}-C_{16} mixture when compared with all the AAD values from different combinations of r1 and r2. Considering that for quaternary mixtures, the original GCSP sometimes gave a value of maximum error as high as 18% and there is no valid method to select the best combination of r1 and r2, the most important advantage of the proposed modification of the GCSP is that it provides a reliable means for predicting the viscosities of multicomponent (n>3) n-alkane liquid mixtures. There is no risk of obtaining an estimation of viscosity far from its real value over the entire composition range. A comparison of the results obtained by the proposed modification of the GCSP in the cases where $\xi_{12} = 1$ and $\xi_{12} \neq 1$ is given in Table 5.26. Correlation with the adjustable parameter ξ_{12} gave much better results than the prediction without ξ_{12}, but the viscosity data of the n-alkane quaternary are not enough to get an equation for predicting ξ_{12}. Therefore, there is no way to calculate this parameter by using pure component properties for quaternary systems. Moreover, as can be noticed from Table 5.26, the values of ξ_{12} do not change too much with the temperatures and the systems, the suggestion is thus that the value of ξ_{12} could be taken as 1.32.

Table 5.26

Results of Using the Modified-GCSP with and without the Interaction Coefficient for Quaternary n-Alkane Systems

System	$\xi_{12} = 1$		$\xi_{12} \neq 1$		
	AAD	MAX	AAD	MAX	ξ_{12}
Temperature = 303.15 K					
C_6(1)-C_{10}(2)-C_{12}(3)-C_{16}(4)	4.35	-5.46	1.25	2.86	1.3387
C_7(1)-C_9(2)-C_{12}(3)-C_{16}(4)	4.21	-5.28	1.16	2.68	1.3110
Temperature = 308.15 K					
C_6(1)-C_{10}(2)-C_{12}(3)-C_{16}(4)	4.34	-5.34	1.31	2.81	1.3382
C_7(1)-C_9(2)-C_{12}(3)-C_{16}(4)	4.41	-5.39	1.24	2.70	1.3231

As a matter of fact, it is the modified-GCSP method which provided the possibility and convenience of predicting ξ_{ij}, because there is only one ξ_{12} in the pseudo-binary mixture model no matter how many components the mixture actually contains. This is very important because there are $C_n^2 = n!/[2!(n-2)!]$ combinations of ξ_{ij} in the original GCSP. For example, a quaternary mixture has 6 ξ_{ij} and a quinary mixture has 10 and so on.

The pseudo-binary mixture model can be used for n-alkane liquid mixtures having unrestricted number of components.

Because there are no experimental data available for n-alkane liquid mixtures with more than four components, comparison cannot be made between the two methods for those mixtures in this study.

For mixtures of more than four components, the number of all possible combinations of r1 and r2 will increase rapidly, the selection of the proper combination becomes more difficult, and the probability to risk the incorrect estimation of mixture viscosity will be much higher. Therefore, the modified-GCSP method should be more meaningful for such mixtures.

5.6 Summary of the Viscosity Prediction Models

5.6.1 Binary n-alkane liquid systems

There are two choices of predicting mixture viscosities reliably for the binary n-alkane liquid systems. One was reported by Asfour *et al.* (1991), the other is reported in this study. The two methods and their accuracies are described below:

(i) The combination of the McAllister three-body model (eq. 2.25) with the technique of Asfour *et al.* (eqs. 2.27 and 2.28) can be used for systems with $| N_2 - N_1 | < 4$, which gives the AAD range of 0.02% ~ 0.4% (refer to Table 5.10), and the combination of the McAllister four-body model with the corresponding technique of Asfour *et al.*(eqs. 2.29 and 2.30) can be used for systems with $| N_2 - N_1 | \geq 4$, which gives the AAD range of 0.15% ~ 1.0% (refer to Table 5.11).

(ii) The combination of the original GCSP method with the improvement proposed in this study (eq. 5.15) can be used for any binary n-alkane systems without limits in the difference of carbon atom numbers between the two components. The AAD range of this combination is 0.63% ~ 1.23% (refer to Table 5.15).

The above two methods have close AAD ranges, the latter requires more physical properties of the pure components than the former, but it is simpler in calculation than the former.

5.6.2 Ternary n-alkane liquid systems

For the viscosity prediction of ternary n-alkane liquid systems, three reliable methods can be used. They all result from the present study.

(i) The combination of the ternary McAllister model (eq. 2.31) with the extension of the technique of Asfour *et al.* (1991), developed in this study (eq. 5.13), when tested by the only available literature ternary n-alkane viscosity data of C_6 -C_{14} -C_{16} (Heric and Brewer, 1969), gives an AAD of 0.36%.

(ii) The combination of the original GCSP with the supplementary equation developed in this study, eq. (5.17), gives AAD range of 0.27% ~ 1.11% (refer to Table 5.16).

(iii) The combination of the modified-GCSP with the prediction equation for the interaction coefficient, eq. (5.23), gives the AAD range of 0.12% ~ 1.46% (refer to Tables 5.23 and 5.24).

The above three methods have close AAD ranges. If more literature viscosity data of n-alkanes were available, method one and method three would be tested further to give more precise ranges of AAD.

5.6.3 Quaternary n-alkane liquid systems

According to the results of this study, the modified-GCSP method is the only reliable method for the prediction of the mixture viscosity for quaternary n-alkane liquid systems. When the binary interaction coefficient, ξ_{12}, is set to unity for prediction, the values of AAD are always less than 4.5% and the values of AAD when ξ_{12} is used as an adjustable parameter is in the range of 1.16% ~ 1.31% (refer to Table 5.26). Due to the scarcity of viscosity data of n-alkane quaternary systems, the only suggestion which can be made in this study on the basis of the results listed in Table 5.26 is that ξ_{12} can be set to 1.32 for more accurate viscosity prediction. It is believed that as long as more quaternary n-alkane viscosity data are collected, an equation similar to eq. (5.23) can be developed to predict ξ_{12} from the properties of pure components of the mixture.

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS

6.1 Conclusions

Viscosities of liquid mixtures are important tools required for engineering applications, but a reliable and generally valid theory for the prediction of liquid mixture viscosity from pure component properties has not been established yet. Thus, most of the theoretical investigations and industry designs are dependent on experimental viscosity data. For n-alkane liquid mixtures, their viscosity data are required for many purposes, especially in the petroleum industry, but there are insufficient data on these mixtures available in the literature. In this study, the viscosities of five selected n-alkane ternary liquid systems and their binary subsystems have been determined at 293.15, 298.15, 308.15 and 313.15 K over the entire composition range. The range of the mixture selected is representive of n-alkane liquid systems. The gathered experimental viscosity data can be considered as valuable addition to the literature.

The technique reported by Asfour *et al.* (1991) for predicting the McAllister binary parameters has been extended in this study to ternary n-alkane mixtures. An equation has been proposed in this study (eq. 5.13), which enables the McAllister model to predict the viscosities of ternary n-alkane mixtures accurately from pure component properties.

The Generalized Corresponding States Principle (GCSP) proposed by Teja and Rice (1981) was first tested by using experimental binary and ternary viscosity data. Then for binary mixtures, eq. (5.15) has been proposed for the prediction of the binary interaction coefficient, which is essential for reducing prediction errors. For ternary mixtures, eq. (5.17) has been presented for reference fluid selection which can minimize the errors in viscosity prediction.

A pseudo-binary mixture model has been developed in this study. This model modifies the GCSP method for multicomponent n-alkane liquid mixtures. It is applicable to mixtures having unrestricted number of components. Since the GCSP method can be either a predictive or a correlating technique depending on whether or not the binary interaction coefficient is set to unity, the developed modified-GCSP method has been tested for the above two kinds of capabilities by ternary and quaternary data. Results show the obvious superiority of the modified-GCSP over the original one. If the modified-GCSP is used as a predictive method, the significant errors resulting from the choice of the reference fluids, as required by the original GCSP, are avoided. If the modified-GCSP is used as a correlation technique, the number of the binary interaction coefficients which is essential for minimizing the deviations is reduced to one, therefore, less costly experimental data are required. Also, the prediction of the binary interaction coefficient becomes possible.

For ternary n-alkane mixtures, eq. (5.23) was proposed in this study to predict the unique coefficient in modified-GCSP with satisfactory accuracy. For quaternary n-alkane mixtures, the modified-GCSP provides the maximum safety and reasonable accuracy in predicting mixture viscosities. If more viscosity data were available, a similar relationship to eq. (5.23) could be obtained to reduce the deviations further. For mixtures containing more than four components, the selection of the proper reference fluids becomes more difficult and the probability to get wrong estimation of mixture viscosity from the original GCSP method becomes higher because of the increased number of components. Moreover, the increased number of the binary interaction coefficients required by the original GCSP need much more experimental data to determine their values. Therefore, the modified-GCSP method which does not involve the selection of reference fluids and which can make the binary interaction coefficient unique is more meaningful for such systems.

The densities of the selected five ternary n-alkane mixtures and eight binary subsystems were also measured at four temperature levels indicated above. Together with the viscosity data of these systems, some thermodynamic properties such as excess volume, excess activation energy and excess viscosity have been calculated and fitted to some literature models.

6.2 Recommendations

Because of the scarcity of viscosity data on n-alkane systems containing four components and non-existence of data on those containing five components, the proposed modification of the GCSP method in this study is only verified by relatively limited data base. Although the results obtained have shown the superiority of the modified-GCSP over the original GCSP, viscosity-composition data on quaternary and quinary n-alkane liquid systems should be collected and such data should be employed for further testing of the modified-GCSP method proposed in this study.

NOMENCLATURE

A	constant generally used as parameter; molar reduced residual Helmholtz free energy
A_{ij}	NRTL parameter; Heric's model parameter
AAD	average absolute deviation, %
B	constant generally used as parameter
C	constant generally used as parameter
D	constant generally used as parameter
E	constant generally used as parameter
f	shear stress for viscous flow
G_{ij}	NRTL nonrandomness factor
$\Delta^{*}G$	activation energy of viscous flow per g-mole, J/mol
$\Delta^{*}G_{o}$	activation energy of viscous flow per molecule
h	Planck's constant
H	enthalpy of mixing
i	index number in sum of the equations
j	index number in sum of the equations
k	Boltzmann's constant
m	number of experimental points
M	molecular weight, g/mol
MAX	maximum deviation, %

n	number of components of the mixture; nth component in the mixture
N	number of carbon atoms (chain length) of pure n-alkanes
$\tilde{N}$	average number of carbon atoms of n-alkane liquid mixture
N_o	Avogadro's number
P	pressure, atm
R	gas constant
r	rate of a liquid molecule moving under shear stress
r_o	rate of a liquid molecule moving as a result of thermal fluctuation
S	entropy of mixing
t	efflux time of the viscometers, sec
T	temperature, K; vibrating period of the density meter,sec
ν	effective volume occupied by one molecule
V	molar volume of the liquid, L/kmol
w	mass of the component, g
W_{visc}	interaction energy for the activation of flow
x	mole fraction
X	reduced compressibility, or reduced transport properties
x_{ji}	local mole fraction of component j around central molecule i
Z	compressibility factor

Greek Letters

α	equivalent to λ_1; NRTL nonrandomness parameter
ε	reciprocal of the fluid viscosity at critical temperature
η	absolute viscosity, 10^{-3} Pa.s
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 distances involved in Eyring's theory
ν	kinematic viscosity, 10^{-6} m^2/s
ν_{12}	McAllister three-body binary model interaction parameter
ν_{21}	McAllister three-body binary model interaction parameter
ν_{123}	McAllister three-body ternary model interaction parameter
ν_{1112}	McAllister four-body binary model interaction parameter
ν_{2221}	McAllister four-body binary model interaction parameter
ν_{1122}	McAllister four-body binary model interaction parameter
ξ	$\ln(\eta V)$
ξ_{ij}	binary interaction coefficient in the Generalized Corresponding States Principle; intermolecular viscous interaction term in Rowley's model
ξ_{loc}	ξ based on local composition
ξ_i^o	pure component ξ value
ρ	density, kg/L
σ	free energy mixing parameter in Rowley's model

ϕ	shape factor; volume fraction
ϕ_{ji}	local volume fraction of component j around central molecule i
ω	Pitzer acentric factor

Subscripts

1,2,3,4	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
n	refer to nth component in the mixture
o	refer to the reference fluid
r1	refer to reference fluid 1
r2	refer to reference fluid 2
α	refer to pure fluid α

Superscripts

'	refer to asymmetric parameter; refer to pseudo binary mixture
*	refer to composition when $\xi_{21} = \xi_{loc}$

calcd	calculated value
E	excess over the ideal solution property
exptl	experimental value
o	refer to the reference fluid
R	reduced properties
r1	refer to reference fluid 1
r2	refer to reference fluid 2
α	refer to pure fluid α

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APPENDICES

Appendix A

Raw Data of Viscosity and Density Measurements

Table A.1

Raw Data of Binary System n-Octane(1)-n-Undecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 293.15 K		
0.0000	0.339514	[9]*	[9]*
0.1007	0.339249	50B158	348.890
0.2018	0.338972	50B159	357.923
0.2995	0.338684	25A498	708.167
0.4108	0.338343	25A483	670.153
0.5076	0.338026	50B159	289.793
0.5964	0.337717	50B158	246.803
0.6995	0.337338	50B159	251.715
0.7954	0.336964	50B158	212.610
0.8978	0.336540	25A498	455.290
1.0000	0.336094	[1]*	[1]*
	Temperature = 298.15 K		
0.0000	0.339076	[10]*	[10]*
0.0992	0.338815	50B158	323.228
0.2059	0.338517	50B159	332.050
0.2978	0.338247	50B158	284.294
0.3916	0.337955	50B159	293.706
0.5019	0.337594	50B158	248.574
0.5959	0.337267	50B159	255.048
0.7154	0.336823	25A498	493.928
0.8041	0.336472	25A498	462.058
0.9011	0.336066	25A498	429.150
1.0000	0.335621	[2]*	[2]*

* see Table A.14 for both the types of viscometer and the values of efflux time

Table A.1 (cont'd)

Raw Data of Binary System n-Octane(1)-n-Undecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.338206	[11]*	[11]*
0.0969	0.337947	50B159	310.578
0.1954	0.337666	50B158	266.246
0.3001	0.337353	50B159	274.680
0.3970	0.337047	50B158	235.002
0.5094	0.336667	50B159	240.518
0.5981	0.336352	25A483	488.958
0.6968	0.335979	25A498	447.140
0.7992	0.335568	25A483	425.280
0.8998	0.335137	25A498	386.600
1.0000	0.334675	[3]*	[3]*
Temperature = 313.15 K			
0.0000	0.337775	[12]*	[12]*
0.0953	0.337515	50B158	264.815
0.1962	0.337226	50B158	249.775
0.2979	0.336920	50B159	258.703
0.3995	0.336592	50B159	243.247
0.5061	0.336231	25A498	480.058
0.5914	0.335924	25A483	465.460
0.7010	0.335506	25A483	431.577
0.8001	0.335105	25A498	394.267
0.8999	0.334672	25A483	375.434
1.0000	0.334206	[4]*	[4]*

* see Table A.14 for both the types of viscometer and the values of efflux time

Table A.2

Raw Data of Binary System n-Octane(1)-n-Tridecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K			
0.0000	0.340972	[13]*	[13]*
0.1023	0.340640	75J29	254.170
0.1966	0.340309	50B159	522.153
0.3001	0.339914	75J29	207.563
0.3813	0.339592	50B158	390.593
0.4993	0.339077	50B158	342.463
0.6002	0.338588	50B159	334.977
0.6949	0.338086	50B158	271.327
0.7976	0.337475	50B159	262.060
0.9047	0.336782	50B159	227.310
1.0000	0.336094	[1]*	[1]*
Temperature = 298.15 K			
0.0000	0.340551	[14]*	[14]*
0.1246	0.340139	75J29	227.570
0.1988	0.339876	50B158	434.755
0.3028	0.339478	50B159	430.880
0.3936	0.339104	50B159	392.300
0.4971	0.338640	50B158	318.908
0.5956	0.338160	50B159	313.916
0.6970	0.337618	25A498	588.783
0.7898	0.337073	50B158	226.846
0.8988	0.336365	50B159	216.198
1.0000	0.335627	[2]*	[2]*

* see Table A.14 for both the types of viscometer and the values of efflux time

Table A.2 (cont'd)

Raw Data of Binary System n-Octane(1)-n-Tridecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.339703	[15]*	[15]*
0.0968	0.339380	75J29	199.028
0.1961	0.339026	50B158	373.645
0.2969	0.338636	50B159	374.085
0.3966	0.338216	50B158	308.680
0.4942	0.337770	50B159	307.570
0.5983	0.337252	50B158	250.440
0.7006	0.336693	50B159	246.163
0.7985	0.336102	50B158	199.538
0.9019	0.335410	50B159	193.590
1.0000	0.334679	[3]*	[3]*
Temperature = 313.15 K			
0.0000	0.339282	[16]*	[16]*
0.0995	0.338950	50B158	377.497
0.1969	0.338600	50B158	346.910
0.2992	0.338200	50B159	347.963
0.3967	0.337786	50B159	317.518
0.4967	0.337325	50B158	261.605
0.5993	0.336805	50B158	235.355
0.6949	0.336282	50B159	233.823
0.7971	0.335661	25A498	439.097
0.8964	0.334988	25A483	398.200
1.0000	0.334206	[4]*	[4]*

* see Table A.14 for both the types of viscometer and the values of efflux time

Table A.3

Raw Data of Binary System n-Octane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 293.15 K		
0.0000	0.342067	[17]*	[17]*
0.1032	0.341704	75J29	371.270
0.2005	0.341332	50B159	739.947
0.2833	0.340982	75J29	293.707
0.3826	0.340521	50B159	576.980
0.5038	0.339891	50B158	435.510
0.6008	0.339327	50B159	414.137
0.7150	0.338570	50B158	308.830
0.7994	0.337937	50B158	268.023
0.8985	0.337093	25A498	516.110
1.0000	0.336094	[1]*	[1]*
	Temperature = 298.15 K		
0.0000	0.341649	[18]*	[18]*
0.1114	0.341254	75J29	331.684
0.1978	0.340916	75J29	298.172
0.2962	0.340495	75J29	262.822
0.3951	0.340030	50B158	472.123
0.4927	0.339522	50B159	451.900
0.5978	0.338911	50B158	350.588
0.7035	0.338210	50B158	295.928
0.8008	0.337477	50B158	250.400
0.9026	0.336598	50B159	228.550
1.0000	0.335619	[2]*	[2]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.3 (cont'd)

Raw Data of Binary System n-Octane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.340820	[19]*	[19]*
0.1015	0.340456	75J29	279.050
0.1973	0.340079	75J29	250.238
0.2986	0.339641	75J29	221.783
0.3975	0.339166	50B158	401.810
0.4973	0.338635	50B159	389.990
0.6065	0.337980	50B158	301.213
0.6944	0.337387	50B198	263.740
0.8053	0.336536	50B159	242.785
0.9040	0.335664	25A498	430.500
1.0000	0.334679	[3]*	[3]*
Temperature = 313.15 K			
0.0000	0.340404	[20]*	[20]*
0.1274	0.339940	75J29	248.795
0.1958	0.339664	75J29	230.720
0.2936	0.339242	50B158	423.313
0.3949	0.338752	50B158	374.243
0.4956	0.338215	50B159	361.603
0.6113	0.337511	50B159	308.355
0.6990	0.336913	25A483	584.963
0.8018	0.336113	50B159	230.680
0.9006	0.335232	50B159	195.123
1.0000	0.334206	[4]*	[4]*

* see Table A.14 for both the types of viscometer and the values of efflux time

Table A.4

Raw Data of Binary System n-Decane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 293.15 K		
0.0000	0.342063	[17]*	[17]*
0.0958	0.341822	75J29	386.260
0.1989	0.341551	75J29	351.183
0.2947	0.341280	50B159	722.367
0.3865	0.341005	75J29	291.620
0.4839	0.340692	50B159	595.347
0.5780	0.340367	50B158	489.270
0.6958	0.339927	50B159	469.317
0.7980	0.339511	50B159	417.680
0.9126	0.339003	50B158	329.240
1.0000	0.338581	[5]*	[5]*
	Temperature = 298.15 K		
0.0000	0.341647	[18]*	[18]*
0.0973	0.341408	75J29	347.590
0.1999	0.341135	75J29	316.763
0.2983	0.340854	75J29	289.140
0.3925	0.340570	75J29	264.070
0.4933	0.340240	50B158	489.763
0.6025	0.339857	50B159	480.760
0.7033	0.339475	50B158	391.550
0.8079	0.339042	50B158	347.316
0.9014	0.338624	50B159	341.510
1.0000	0.338138	[6]*	[6]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.4 (cont'd)

Raw Data of Binary System n-Decane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.340820	[19]*	[19]*
0.1015	0.340563	75J29	287.027
0.1942	0.340313	75J29	265.473
0.3018	0.340004	75J29	242.000
0.4028	0.339689	75J29	220.860
0.5033	0.339356	75J29	201.008
0.6010	0.339006	50B158	375.043
0.7040	0.338609	50B158	337.268
0.8051	0.338184	50B158	302.705
0.9044	0.337730	50B159	297.673
1.0000	0.337251	[7]*	[7]*
Temperature = 313.15 K			
0.0000	0.340411	[20]*	[20]*
0.0999	0.340155	75J29	263.110
0.1932	0.339903	75J29	244.125
0.3013	0.339592	75J29	222.983
0.4013	0.339281	75J29	204.403
0.5059	0.338929	50B158	381.425
0.5986	0.338595	50B158	349.315
0.7028	0.338187	50B159	346.505
0.8009	0.337772	50B158	284.548
0.8988	0.337320	50B159	218.097
1.0000	0.336813	[8]*	[8]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.5

Raw Data of Binary System n-Undecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 293.15 K		
0.0000	0.342058	[17]*	[17]*
0.1013	0.341860	75J29	390.997
0.2015	0.341651	75J29	362.580
0.3004	0.341436	75J29	335.840
0.4036	0.341198	50B158	635.380
0.5046	0.340951	50B159	645.840
0.6010	0.340705	50B158	537.670
0.7004	0.340433	50B159	541.767
0.8000	0.340144	50B158	449.617
0.8974	0.339845	50B159	451.500
1.0000	0.339507	[9]*	[9]*
	Temperature = 298.15 K		
0.0000	0.341649	[18]*	[18]*
0.1026	0.341447	75J29	351.723
0.2004	0.341243	75J29	327.570
0.2994	0.341025	75J29	304.253
0.3983	0.340797	75J29	281.765
0.5079	0.340528	75J29	258.545
0.5973	0.340295	75J29	240.128
0.7022	0.340008	50B158	453.315
0.7975	0.339730	50B158	415.736
0.9019	0.339405	50B158	377.105
1.0000	0.339079	[10]*	[10]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.5 (cont'd)

Raw Data of Binary System n-Undecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.340820	[19]*	[19]*
0.1005	0.340618	75J29	291.563
0.1976	0.340414	75J29	273.023
0.3007	0.340185	75J29	254.325
0.4013	0.339946	75J29	236.475
0.5074	0.339683	50B158	448.712
0.5964	0.339446	50B159	461.100
0.7036	0.339151	50B198	384.923
0.7972	0.338869	50B158	356.418
0.9014	0.338541	50B159	358.605
1.0000	0.338206	[11]*	[11]*
Temperature = 313.15 K			
0.0000	0.340407	[20]*	[20]*
0.0989	0.340205	75J29	267.103
0.1959	0.340000	75J29	250.765
0.2996	0.339767	75J29	233.932
0.3974	0.339538	75J29	218.515
0.4945	0.339294	50B158	418.483
0.5890	0.339046	50B159	429.348
0.7019	0.338726	50B158	359.580
0.7983	0.338439	50B158	331.410
0.9013	0.338107	50B159	334.650
1.0000	0.337773	[12]*	[12]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.6

Raw Data of Binary System n-Tridecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 293.15 K		
0.0000	0.342058	[17]*	[17]*
0.1030	0.341959	50B158	832.163
0.2020	0.341860	75J29	389.820
0.3023	0.341758	75J29	375.347
0.3967	0.341659	75J29	361.117
0.5078	0.341539	75J29	345.607
0.5972	0.341441	50B158	683.353
0.7003	0.341325	50B158	656.183
0.8019	0.341206	50B158	628.407
0.9018	0.341086	50B159	662.303
1.0000	0.340965	[13]*	[13]*
	Temperature = 298.15 K		
0.0000	0.341649	[18]*	[18]*
0.1000	0.341551	75J29	364.398
0.2001	0.341452	75J29	351.340
0.2975	0.341351	75J29	338.610
0.3984	0.341246	75J29	325.870
0.5000	0.341136	50B158	643.187
0.5985	0.341026	50B159	680.810
0.7037	0.340909	50B158	592.233
0.8004	0.340794	50B158	570.550
0.8983	0.340674	50B158	549.283
1.0000	0.340551	[14]*	[14]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.6 (cont'd)

Raw Data of Binary System n-Tridecane(1)-n-Pentadecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.340822	[19]*	[19]*
0.1004	0.340721	75J29	301.085
0.2017	0.340620	75J29	290.718
0.2979	0.340520	75J29	281.065
0.3944	0.340416	75J29	271.770
0.4994	0.340303	75J29	261.583
0.5935	0.340196	50B158	518.743
0.7015	0.340073	50B158	498.343
0.8030	0.339951	50B159	527.465
0.9036	0.339826	50B158	461.175
1.0000	0.339706	[15]*	[15]*
Temperature = 313.15 K			
0.0000	0.340405	[20]*	[20]*
0.1024	0.340304	75J29	274.935
0.1963	0.340206	75J29	266.390
0.3028	0.340095	75J29	256.968
0.4019	0.339989	75J29	248.400
0.4997	0.339878	75J29	240.118
0.5970	0.339770	50B159	523.473
0.7040	0.339646	50B158	458.063
0.8017	0.339526	50B159	486.030
0.8985	0.339409	50B159	468.860
1.0000	0.339279	[16]*	[16]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.7

Raw Data of Binary System n-Decane(1)-n-Tridecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 293.15 K		
0.0000	0.340965	[13]*	[13]*
0.0987	0.340777	75J29	264.757
0.1990	0.340576	50B159	561.793
0.2995	0.340366	75J29	233.233
0.3997	0.340146	75J29	218.797
0.4986	0.339917	75J29	204.963
0.5974	0.339681	50B159	432.773
0.7015	0.339418	50B158	365.643
0.7973	0.339161	50B158	341.683
0.9014	0.338870	50B158	316.753
1.0000	0.338578	[5]*	[5]*
	Temperature = 298.15 K		
0.0000	0.340551	[14]*	[14]*
0.0939	0.340369	50B158	497.328
0.1985	0.340159	75J29	227.590
0.2931	0.339958	75J29	214.913
0.3988	0.339725	50B158	413.060
0.5031	0.339481	50B158	386.300
0.5995	0.339247	50B159	399.274
0.7013	0.338987	25A498	787.557
0.8001	0.338724	50B158	316.533
0.9011	0.338436	50B159	324.230
1.0000	0.338140	[6]*	[6]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.7 (cont'd)

Raw Data of Binary System n-Decane(1)-n-Tridecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
	Temperature = 308.15 K		
0.0000	0.339702	[15]*	[15]*
0.0991	0.339510	75J29	205.120
0.1956	0.339313	75J29	194.280
0.2991	0.339090	50B158	375.895
0.3993	0.338863	50B158	354.730
0.5073	0.338608	50B158	332.440
0.5962	0.338388	50B159	346.455
0.6991	0.338123	50B158	295.153
0.8037	0.337836	50B159	303.424
0.8994	0.337559	50B158	258.898
1.0000	0.337250	[7]*	[7]*
	Temperature = 313.15 K		
0.0000	0.339279	[16]*	[16]*
0.1001	0.339080	75J29	189.620
0.1979	0.338878	75J29	179.944
0.2988	0.338661	75J29	170.288
0.3996	0.338433	75J29	160.863
0.5025	0.338187	50B159	341.958
0.5994	0.337945	50B158	293.498
0.6995	0.337683	50B158	276.043
0.7980	0.337409	50B159	285.585
0.9013	0.337109	50B159	267.130
1.0000	0.336803	[8]*	[8]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.8

Raw Data of Binary System n-Undecane(1)-n-Tridecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K			
0.0000	0.340972	[13]*	[13]*
0.1389	0.340795	75J29	265.317
0.2035	0.340710	75J29	258.245
0.3044	0.340574	50B158	507.903
0.4076	0.340430	50B159	534.580
0.4924	0.340308	75J29	228.248
0.5897	0.340167	50B158	448.793
0.7108	0.339983	50B159	468.095
0.8027	0.339837	50B158	407.253
0.8980	0.339682	50B159	429.030
1.0000	0.339514	[9]*	[9]*
Temperature = 298.15 K			
0.0000	0.340541	[14]*	[14]*
0.1023	0.340411	75J29	245.895
0.2031	0.340277	75J29	235.780
0.2991	0.340147	75J29	226.763
0.3981	0.340008	75J29	217.503
0.4981	0.339863	50B158	428.020
0.5976	0.339715	50B158	410.757
0.7028	0.339554	50B159	432.000
0.8000	0.339401	50B158	376.227
0.9004	0.339235	50B159	395.630
1.0000	0.339068	[10]*	[10]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.8 (cont'd)

Raw Data of Binary System n-Undecane(1)-n-Tridecane(2)

Mole Fraction x_1	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K			
0.0000	0.339706	[15]*	[15]*
0.1051	0.339568	75J29	208.080
0.1986	0.339445	75J29	200.968
0.2953	0.339311	50B158	397.448
0.3954	0.339168	50B158	382.680
0.5039	0.339008	50B158	366.633
0.5908	0.338878	50B159	389.673
0.6994	0.338710	50B159	373.075
0.7972	0.338554	50B158	325.410
0.9027	0.338378	50B158	311.303
1.0000	0.338209	[11]*	[11]*
Temperature = 313.15 K			
0.0000	0.339283	[16]*	[16]*
0.1040	0.339146	75J29	192.570
0.1964	0.339023	75J29	186.180
0.2972	0.338882	75J29	179.465
0.3982	0.338738	50B158	354.720
0.5078	0.338575	50B159	374.283
0.5968	0.338440	50B159	361.565
0.7018	0.338277	50B158	315.363
0.7985	0.338117	50B159	333.833
0.9009	0.337947	50B159	320.240
1.0000	0.337774	[12]*	[12]*

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.9

Raw Data of Ternary System n-Octane(1)-n-Undecane(2)-n-Tridecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.336090	[1]*	[1]*	1.0000	0.0000	0.335615	[2]*	[2]*
0.0000	1.0000	0.339513	[9]*	[9]*	0.0000	1.0000	0.339068	[10]*	[10]*
0.0000	0.0000	0.340972	[13]*	[13]*	0.0000	0.0000	0.340542	[14]*	[14]*
0.1043	0.1018	0.340494	75J29	243.133	0.1053	0.1002	0.340057	75J29	222.855
0.1087	0.7938	0.339402	50B159	398.963	0.1052	0.7940	0.338971	50B158	337.367
0.2058	0.2046	0.339969	75J29	209.235	0.2045	0.2002	0.339541	75J29	193.257
0.2096	0.3973	0.339645	50B159	393.127	0.2062	0.3907	0.339227	50B158	364.300
0.2055	0.5981	0.339321	50B158	391.370	0.1979	0.6038	0.338895	50B159	366.660
0.3124	0.2921	0.339386	50B158	367.010	0.2998	0.2950	0.338990	50B158	344.597
0.3067	0.3977	0.339225	50B158	351.190	0.3058	0.3990	0.338780	50B159	358.780
0.4072	0.1998	0.339124	50B158	344.787	0.3981	0.1976	0.338725	50B158	323.817
0.3990	0.3049	0.338969	50B159	363.650	0.4031	0.3008	0.338510	50B159	337.317
0.4007	0.4024	0.338774	50B158	314.153	0.4020	0.4011	0.338318	50B158	292.218
0.6048	0.2009	0.338131	50B159	299.907	0.5925	0.2097	0.337726	50B159	283.679
0.7999	0.1002	0.337211	50B159	246.817	0.7978	0.1025	0.336752	25A483	520.160

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.9 (cont'd)

Raw Data of Ternary System n-Octane(1)-n-Undecane(2)-n-Tridecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.334676	[3]*	[3]*	1.0000	0.0000	0.334199	[4]*	[4]*
0.0000	1.0000	0.338203	[11]*	[11]*	0.0000	1.0000	0.337765	[12]*	[12]*
0.0000	0.0000	0.339696	[15]*	[15]*	0.0000	0.0000	0.339274	[16]*	[16]*
0.1062	0.0985	0.339206	75J29	190.003	0.0964	0.0975	0.338818	75J29	177.923
0.1048	0.7952	0.338100	50B158	293.453	0.1026	0.8003	0.337667	50B159	302.267
0.2060	0.1951	0.338682	75J29	166.808	0.1981	0.1988	0.338280	50B158	320.800
0.2060	0.3903	0.338365	50B158	315.523	0.1993	0.3896	0.337959	50B158	296.790
0.2033	0.6018	0.338009	50B159	317.740	0.2063	0.5973	0.337561	50B159	297.002
0.3072	0.2969	0.338085	50B159	326.657	0.2961	0.2976	0.337696	50B158	281.073
0.3030	0.3960	0.337922	50B159	314.117	0.3028	0.4014	0.337474	50B159	293.878
0.3973	0.1979	0.337854	25A498	657.610	0.4045	0.1996	0.337381	50B159	289.663
0.3922	0.3072	0.337669	50B158	270.847	0.4032	0.2947	0.337199	50B159	278.438
0.4007	0.4006	0.337444	50B159	283.020	0.4016	0.4002	0.336995	50B158	241.743
0.6069	0.1954	0.336781	50B159	248.667	0.6117	0.1972	0.336299	50B159	233.483
0.7962	0.1023	0.335850	50B159	208.810	0.8004	0.1002	0.335367	25A498	415.183

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.10

Raw Data of Ternary System n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.336091	[1]*	[1]*	1.0000	0.0000	0.335615	[2]*	[2]*
0.0000	1.0000	0.339518	[9]*	[9]*	0.0000	1.0000	0.339068	[10]*	[10]*
0.0000	0.0000	0.342070	[17]*	[17]*	0.0000	0.0000	0.341638	[18]*	[18]*
0.1038	0.1022	0.341483	75J29	342.830	0.1108	0.0995	0.341031	75J29	308.490
0.1091	0.7909	0.339580	75J29	186.333	0.1096	0.7909	0.339124	50B159	388.883
0.2082	0.2010	0.340808	75J29	274.940	0.2131	0.2016	0.340346	75J29	249.383
0.2019	0.3999	0.340293	75J29	233.217	0.2068	0.3952	0.339836	75J29	213.547
0.2111	0.5953	0.339632	75J29	191.220	0.2087	0.5964	0.339192	50B158	364.303
0.3068	0.2995	0.340043	50B158	447.980	0.3025	0.2971	0.339625	75J29	202.800
0.3029	0.3988	0.339748	75J29	199.777	0.3083	0.3947	0.339286	50B158	377.543
0.4042	0.1993	0.339823	50B159	466.117	0.4039	0.2016	0.339364	75J29	190.237
0.4007	0.3008	0.339505	50B158	385.760	0.4023	0.2988	0.339053	50B158	357.647
0.4051	0.3987	0.339136	50B158	348.120	0.4027	0.4017	0.338687	50B158	323.767
0.6019	0.2054	0.338548	50B159	335.043	0.5903	0.2059	0.338172	50B159	318.567
0.8005	0.1016	0.337454	50B158	237.943	0.8034	0.1012	0.336966	50B158	223.677

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.10 (cont'd)

Raw Data of Ternary System n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.334676	[3]*	[3]*	1.0000	0.0000	0.334201	[4]*	[4]*
0.0000	1.0000	0.338202	[11]*	[11]*	0.0000	1.0000	0.337766	[12]*	[12]*
0.0000	0.0000	0.340815	[19]*	[19]*	0.0000	0.0000	0.340401	[20]*	[20]*
0.1208	0.0986	0.340156	75J29	255.158	0.1018	0.0951	0.339823	75J29	240.138
0.1086	0.7899	0.338272	50B159	338.155	0.1030	0.7965	0.337849	75J29	140.566
0.2077	0.2035	0.339521	75J29	212.768	0.2004	0.1964	0.339148	75J29	199.805
0.2102	0.3937	0.338972	50B158	374.203	0.1992	0.3971	0.338590	50B158	352.377
0.2025	0.6004	0.338358	50B158	317.467	0.2014	0.5952	0.337944	50B158	298.427
0.3060	0.3009	0.338741	50B159	390.423	0.2998	0.2988	0.338352	50B158	334.127
0.3043	0.4040	0.338417	50B159	358.567	0.2939	0.4029	0.338050	50B159	340.037
0.3956	0.2061	0.338542	50B158	339.920	0.4021	0.1961	0.338103	50B158	316.988
0.3983	0.3050	0.338193	50B159	341.793	0.4011	0.2953	0.337771	50B159	321.117
0.4032	0.3977	0.337824	50B158	283.500	0.3947	0.4066	0.337407	50B159	294.093
0.6003	0.2049	0.337219	50B159	276.167	0.6050	0.1965	0.336775	50B158	236.403
0.7969	0.1060	0.336092	50B159	220.216	0.8015	0.0977	0.335630	25A498	439.730

* see Table A.14 for the types of viscometer and the values of efflux time.

Table A.11

Raw Data of Ternary System n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.339510	[9]*	[9]*	1.0000	0.0000	0.339067	[10]*	[10]*
0.0000	1.0000	0.340971	[13]*	[13]*	0.0000	1.0000	0.340542	[14]*	[14]*
0.0000	0.0000	0.342057	[17]*	[17]*	0.0000	0.0000	0.341637	[18]*	[18]*
0.1007	0.1011	0.341761	75J29	374.240	0.1023	0.0994	0.341330	75J29	339.033
0.1031	0.7990	0.340964	50B158	576.697	0.0985	0.8009	0.340539	50B159	527.640
0.2021	0.2018	0.341434	75J29	332.530	0.2025	0.2008	0.341004	75J29	302.413
0.2019	0.3946	0.341214	75J29	307.990	0.2038	0.3949	0.340775	75J29	279.397
0.1997	0.6040	0.340966	75J29	281.818	0.1998	0.6008	0.340536	50B158	528.783
0.3002	0.2943	0.341098	75J29	296.340	0.3023	0.2957	0.340659	75J29	269.363
0.3019	0.4009	0.340963	75J29	282.917	0.3033	0.3999	0.340527	75J29	257.763
0.4029	0.2007	0.340959	50B158	579.900	0.3984	0.2034	0.340535	50B159	585.046
0.4006	0.3009	0.340839	50B158	557.030	0.3991	0.3013	0.340408	50B159	561.113
0.4001	0.4047	0.340705	50B159	585.660	0.4024	0.4038	0.340265	50B158	486.797
0.6052	0.1973	0.340428	50B158	488.203	0.6028	0.1982	0.339995	50B159	494.437
0.7995	0.0998	0.339998	50B158	428.140	0.8027	0.0970	0.339549	50B158	394.427

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.11 (cont'd)

Raw Data of Ternary System n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.338208	[11]*	[11]*	1.0000	0.0000	0.337768	[12]*	[12]*
0.0000	1.0000	0.339700	[15]*	[15]*	0.0000	1.0000	0.339278	[16]*	[16]*
0.0000	0.0000	0.340819	[19]*	[19]*	0.0000	0.0000	0.340402	[20]*	[20]*
0.1030	0.1021	0.340504	75J29	280.947	0.0957	0.0937	0.340111	75J29	259.290
0.0996	0.8060	0.339696	75J29	216.678	0.1027	0.7995	0.339272	75J29	200.023
0.2019	0.1981	0.340182	75J29	253.563	0.2012	0.2007	0.339761	75J29	232.737
0.2021	0.3938	0.339952	75J29	235.580	0.1956	0.3991	0.339537	75J29	217.540
0.2005	0.6019	0.339701	50B159	490.900	0.1959	0.6024	0.339282	50B158	413.623
0.3030	0.2983	0.339824	75J29	226.740	0.2922	0.2978	0.339426	75J29	210.833
0.2996	0.4028	0.339702	75J29	218.213	0.2987	0.3959	0.339286	50B158	414.957
0.3968	0.2000	0.339709	50B159	495.823	0.3950	0.2005	0.339288	75J29	203.137
0.3732	0.3142	0.339623	50B159	482.560	0.3973	0.2983	0.339155	50B159	440.870
0.4028	0.3998	0.339430	50B158	414.190	0.3945	0.4067	0.339017	50B158	384.847
0.6018	0.1983	0.339156	50B159	422.877	0.6018	0.1966	0.338728	50B159	392.700
0.8003	0.1013	0.338700	50B159	373.680	0.7985	0.1008	0.338275	50B158	317.117

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.12

Raw Data of Ternary System n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.336087	[1]*	[1]*	1.0000	0.0000	0.335615	[2]*	[2]*
0.0000	1.0000	0.340968	[13]*	[13]*	0.0000	1.0000	0.340542	[14]*	[14]*
0.0000	0.0000	0.342060	[17]*	[17]*	0.0000	0.0000	0.341638	[18]*	[18]*
0.1051	0.0982	0.341588	50B158	726.570	0.1047	0.0988	0.341161	75J29	322.077
0.1085	0.7960	0.340746	75J29	263.856	0.1049	0.7983	0.340326	75J29	242.027
0.2089	0.2018	0.341048	75J29	296.917	0.2045	0.1972	0.340643	75J29	273.153
0.2045	0.3899	0.340833	75J29	275.943	0.2084	0.3920	0.340379	75J29	250.427
0.2074	0.5969	0.340542	75J29	250.057	0.2074	0.5969	0.340107	50B159	518.257
0.3024	0.2956	0.340502	75J29	250.443	0.2986	0.2974	0.340081	50B158	473.550
0.3061	0.4020	0.340333	50B158	486.260	0.3028	0.4005	0.339912	75J29	218.987
0.4009	0.1998	0.340139	50B159	509.733	0.4034	0.1996	0.339689	50B159	469.263
0.4030	0.2986	0.339978	50B158	442.383	0.4092	0.2977	0.339506	50B158	404.003
0.4037	0.4025	0.339810	50B158	419.883	0.3998	0.4065	0.339384	50B158	390.443
0.6185	0.1931	0.338858	50B158	330.040	0.6052	0.1990	0.338485	50B158	313.837
0.8009	0.1005	0.337690	50B159	276.833	0.8034	0.0973	0.337217	50B159	259.450

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.12 (cont'd)

Raw Data of Ternary System n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.334673	[3]*	[3]*	1.0000	0.0000	0.334204	[4]*	[4]*
0.0000	1.0000	0.339699	[15]*	[15]*	0.0000	1.0000	0.339275	[16]*	[16]*
0.0000	0.0000	0.340813	[19]*	[19]*	0.0000	0.0000	0.340403	[20]*	[20]*
0.1061	0.1009	0.340326	75J29	267.818	0.0998	0.1140	0.339913	75J29	246.360
0.1086	0.7972	0.339464	50B159	462.117	0.1048	0.7955	0.339060	75J29	190.565
0.2076	0.1961	0.339796	75J29	229.678	0.2059	0.2011	0.339372	75J29	211.820
0.2048	0.3941	0.339552	75J29	213.255	0.2003	0.3951	0.339149	75J29	198.225
0.2113	0.5947	0.339249	50B158	399.957	0.2037	0.5988	0.338852	50B158	373.890
0.3057	0.2932	0.339204	50B159	440.880	0.3116	0.3083	0.338727	75J29	178.760
0.3113	0.3989	0.339019	50B158	380.680	0.2998	0.4051	0.338646	50B158	358.087
0.4065	0.1978	0.338823	50B159	401.913	0.4000	0.1977	0.338427	75J29	167.047
0.4081	0.2967	0.338656	50B159	383.937	0.3978	0.3018	0.338277	50B159	361.953
0.4017	0.4051	0.338520	50B158	335.613	0.3959	0.4064	0.338118	50B159	346.930
0.6080	0.1958	0.337601	50B159	301.070	0.6015	0.1995	0.337192	50B159	284.947
0.8052	0.0976	0.336305	50B159	230.087	0.8047	0.0986	0.335847	50B159	217.755

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.13

Raw Data of Ternary System n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.338585	[5]*	[5]*	1.0000	0.0000	0.338128	[6]*	[6]*
0.0000	1.0000	0.340973	[13]*	[13]*	0.0000	1.0000	0.340542	[14]*	[14]*
0.0000	0.0000	0.342060	[17]*	[17]*	0.0000	0.0000	0.341638	[18]*	[18]*
0.1002	0.1025	0.341707	75J29	367.938	0.0814	0.1024	0.341328	75J29	339.160
0.0993	0.8003	0.340911	75J29	276.787	0.0836	0.8163	0.340504	75J29	254.350
0.1979	0.2021	0.341329	75J29	321.737	0.1700	0.2119	0.340968	75J29	299.510
0.1999	0.3961	0.341099	75J29	296.503	0.1652	0.4175	0.340742	75J29	277.083
0.2022	0.6023	0.340835	75J29	271.587	0.1696	0.6245	0.340473	75J29	253.173
0.2970	0.3010	0.340916	75J29	280.595	0.2514	0.3155	0.340604	75J29	265.686
0.2981	0.4012	0.340784	50B158	549.660	0.2547	0.4316	0.340447	50B158	518.010
0.3953	0.1983	0.340731	50B158	541.687	0.3479	0.2183	0.340421	50B159	568.240
0.4052	0.2985	0.340562	75J29	249.877	0.3490	0.3270	0.340276	50B158	492.563
0.3976	0.4006	0.340450	50B158	494.830	0.3494	0.4377	0.340123	50B158	468.897
0.6004	0.2004	0.339995	50B158	433.017	0.5485	0.2266	0.339709	50B158	417.420
0.8006	0.1023	0.339330	50B159	394.507	0.7675	0.1177	0.338999	50B159	377.630

* see Table A.14 for the types of viscometer and the values of efflux time

Table A.13 (cont'd)

Raw Data of Ternary System n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec	Mole Fraction x_1	Mole Fraction x_2	Density Meter Reading sec	Viscometer	Efflux Time sec
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.337248	[7]*	[7]*	1.0000	0.0000	0.336803	[8]*	[4]*
0.0000	1.0000	0.339697	[15]*	[15]*	0.0000	1.0000	0.339275	[16]*	[16]*
0.0000	0.0000	0.340816	[19]*	[19]*	0.0000	0.0000	0.340403	[20]*	[20]*
0.1003	0.1008	0.340455	75J29	277.255	0.1006	0.1034	0.340038	75J29	253.985
0.0996	0.7999	0.339634	75J29	213.390	0.0998	0.8028	0.339210	75J29	197.040
0.2013	0.1990	0.340062	75J29	245.507	0.1981	0.2033	0.339648	75J29	226.190
0.1997	0.4042	0.339818	50B158	466.207	0.1987	0.3985	0.339410	75J29	210.410
0.2000	0.6013	0.339567	50B159	474.770	0.2005	0.6017	0.339142	50B159	438.727
0.3010	0.3019	0.339625	75J29	215.450	0.2941	0.2992	0.339229	75J29	200.643
0.2998	0.4028	0.339498	50B159	467.817	0.3010	0.4011	0.339073	50B158	393.590
0.4008	0.2012	0.339429	50B158	419.243	0.3971	0.1948	0.339029	50B158	390.593
0.4006	0.3003	0.339293	50B159	442.827	0.3995	0.3033	0.338868	50B159	410.530
0.4008	0.3996	0.339152	50B158	386.230	0.4001	0.4007	0.338728	50B159	394.407
0.6014	0.2009	0.338694	50B159	377.627	0.6018	0.1979	0.338268	50B159	352.187
0.8038	0.0978	0.338012	50B159	318.270	0.8038	0.1000	0.337572	50B159	297.817

* see Table A.14 for the types of viscometer and the values of efflux time

Tabel A.14

Raw Viscosity Data of Pure Components

No.	Substance	Temperature K	Viscometer	Efflux Time sec
[1]	n-Octane	293.15	50B158	180.820
[2]	n-Octane	298.15	25A498	396.818
			25A483	405.678
[3]	n-Octane	308.15	25A498	358.447
			25A483	336.337
[4]	n-Octane	313.15	25A483	348.720
			25A498	341.103
[5]	n-Decane	293.15	25A483	699.000
			50B158	294.187
[6]	n-Decane	298.15	50B159	301.753
			50B158	274.323
[7]	n-Decane	308.15	50B158	241.820
			50B159	265.770
[8]	n-Decane	313.15	25A498	528.077
			25A483	539.577
[9]	n-Undecane	293.15	50B158	371.482
			50B159	408.435
[10]	n-Undecane	298.15	50B159	378.062
			50B158	343.762
[11]	n-Undecane	308.15	50B158	298.710
			50B159	328.640
			25A498	693.367
[12]	n-Undecane	313.15	25A483	663.920
			25A498	649.713
			50B159	307.323
			50B158	279.353

Tabel A.14 (cont'd)

Raw Viscosity Data of Pure Components

No.	Substance	Temperature K	Viscometer	Efflux Time sec
[13]	n-Tridecane	293.15	50B158	576.413
			50B159	634.700
			75J29	280.970
[14]	n-Tridecane	298.15	50B158	525.274
			50B159	577.642
			75J29	255.858
[15]	n-Tridecane	308.15	50B158	443.773
			50B159	488.227
			75J29	216.325
[16]	n-Tridecane	313.15	75J29	199.815
			50B159	451.047
			50B158	410.110
[17]	n-Pentadecane	293.15	50B158	865.637
			75J29	421.635
[18]	n-Pentadecane	298.15	50B159	853.708
			75J29	377.990
[19]	n-Pentadecane	308.15	50B159	702.813
			75J29	311.147
[20]	n-Pentadecane	313.15	75J29	284.230
			50B158	583.333

Appendix B

Excess Properties of Ternary n-Alkane Systems

Table B.1

The Excess Properties of Ternary System n-Octane(1)-n-Undecane(2)-n-Tridecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^{*}G^{E}$ J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^{*}G^{E}$ J/mol
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1043	0.1018	-0.0549	-0.0399	74.8043	0.1053	0.1002	-0.0435	-0.0381	80.3708
0.1087	0.7938	-0.0281	-0.0336	42.7693	0.1052	0.7940	-0.0227	-0.0339	44.6241
0.2058	0.2046	-0.0842	-0.0654	120.9099	0.2045	0.2002	-0.0695	-0.0673	123.5702
0.2096	0.3973	-0.0727	-0.0629	109.9878	0.2062	0.3907	-0.0598	-0.0638	113.5044
0.2055	0.5981	-0.0532	-0.0591	84.1156	0.1979	0.6038	-0.0430	-0.0499	85.9025
0.3124	0.2921	-0.0911	-0.0825	142.1149	0.2998	0.2950	-0.0736	-0.0806	145.9690
0.3067	0.3977	-0.0785	-0.0780	126.8695	0.3058	0.3990	-0.0645	-0.0814	131.3808
0.4072	0.1998	-0.1039	-0.0909	167.2177	0.3981	0.1976	-0.0862	-0.1015	172.0650
0.3990	0.3049	-0.0898	-0.0889	147.4283	0.4031	0.3008	-0.0745	-0.0991	153.6354
0.4007	0.4024	-0.0745	-0.0824	126.9276	0.4020	0.4011	-0.0625	-0.0830	129.8821
0.6048	0.2009	-0.0823	-0.0945	147.9234	0.5925	0.2097	-0.0683	-0.1072	155.5715
0.7999	0.1002	-0.0533	-0.0705	100.8534	0.7978	0.1025	-0.0433	-0.0686	110.2246

Table B.1 (cont'd)

The Excess Properties of Ternary System n-Octane(1)-n-Undecane(2)-n-Tridecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1062	0.0985	-0.0317	-0.0561	79.5725	0.0964	0.0975	-0.0245	-0.0645	75.5701
0.1048	0.7952	-0.0165	-0.0292	44.6901	0.1026	0.8003	-0.0137	-0.0482	44.2267
0.2060	0.1951	-0.0485	-0.0802	128.9975	0.1981	0.1988	-0.0391	-0.1001	129.9460
0.2060	0.3903	-0.0421	-0.0818	116.1777	0.1993	0.3896	-0.0349	-0.0867	116.9876
0.2033	0.6018	-0.0307	-0.0756	89.1893	0.2063	0.5973	-0.0256	-0.0731	94.4552
0.3072	0.2969	-0.0519	-0.1022	151.6222	0.2961	0.2976	-0.0426	-0.1113	154.4400
0.3030	0.3960	-0.0456	-0.0856	136.0390	0.3028	0.4014	-0.0378	-0.0979	139.2172
0.3973	0.1979	-0.0573	-0.1240	187.9659	0.4045	0.1996	-0.0495	-0.1513	187.4802
0.3922	0.3072	-0.0516	-0.1063	159.9660	0.4032	0.2947	-0.0430	-0.1254	170.6162
0.4007	0.4006	-0.0429	-0.0981	140.7301	0.4016	0.4002	-0.0358	-0.1071	145.7026
0.6069	0.1954	-0.0466	-0.1207	170.0592	0.6117	0.1972	-0.0375	-0.1359	176.8483
0.7962	0.1023	-0.0275	-0.0852	132.6377	0.8004	0.1002	-0.0260	-0.0980	117.4748

Table B.2

The Excess Properties of Ternary System n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^{*}G^{E}$ J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^{*}G^{E}$ J/mol
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1038	0.1022	-0.1425	-0.0539	144.9157	0.1108	0.0995	-0.1157	-0.0796	157.9899
0.1091	0.7909	-0.0627	-0.0428	79.0635	0.1096	0.7909	-0.0504	-0.0344	83.1347
0.2082	0.2010	-0.2096	-0.0995	236.2205	0.2131	0.2016	-0.1696	-0.1165	246.9844
0.2019	0.3999	-0.1802	-0.0943	213.5744	0.2068	0.3952	-0.1472	-0.0965	221.9782
0.2111	0.5953	-0.1216	-0.0642	150.7777	0.2087	0.5964	-0.0974	-0.0641	159.3681
0.3068	0.2995	-0.2092	-0.1179	262.1321	0.3025	0.2971	-0.1704	-0.1163	269.6904
0.3029	0.3988	-0.1803	-0.0955	225.2426	0.3083	0.3947	-0.1456	-0.1147	237.6372
0.4042	0.1993	-0.2344	-0.1445	304.4397	0.4039	0.2016	-0.1906	-0.1339	312.0720
0.4007	0.3008	-0.1993	-0.1206	260.6655	0.4023	0.2988	-0.1615	-0.1308	273.2954
0.4051	0.3987	-0.1511	-0.1149	214.7435	0.4027	0.4017	-0.1232	-0.1191	220.7158
0.6019	0.2054	-0.1666	-0.1344	251.9539	0.5903	0.2059	-0.1419	-0.1446	265.9033
0.8005	0.1016	-0.1027	-0.1053	170.4723	0.8034	0.1012	-0.0798	-0.1107	185.5553

Table B.2 (cont'd)

The Excess Properties of Ternary System n-Octane(1)-n-Undecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1208	0.0986	-0.0817	-0.0807	171.4958	0.1018	0.0951	-0.0610	-0.0842	152.5232
0.1086	0.7899	-0.0360	-0.0530	83.5004	0.1030	0.7965	-0.0297	-0.0605	82.0088
0.2077	0.2035	-0.1144	-0.1298	248.8132	0.2004	0.1964	-0.0932	-0.1338	249.0913
0.2102	0.3937	-0.1017	-0.1225	227.2196	0.1992	0.3971	-0.0829	-0.1290	228.6767
0.2025	0.6004	-0.0671	-0.0981	161.9793	0.2014	0.5952	-0.0573	-0.0963	166.6137
0.3060	0.3009	-0.1172	-0.1510	274.3943	0.2998	0.2988	-0.0974	-0.1699	280.0605
0.3043	0.4040	-0.0987	-0.1381	240.9752	0.2939	0.4029	-0.0832	-0.1579	245.3421
0.3956	0.2061	-0.1304	-0.1804	319.1648	0.4021	0.1961	-0.1102	-0.1965	329.2422
0.3983	0.3050	-0.1102	-0.1667	280.0259	0.4011	0.2953	-0.0939	-0.1767	289.2971
0.4032	0.3977	-0.0865	-0.1379	230.1564	0.3947	0.4066	-0.0711	-0.1507	234.8816
0.6003	0.2049	-0.0948	-0.1672	275.0149	0.6050	0.1965	-0.0806	-0.1910	284.8650
0.7969	0.1060	-0.0555	-0.1353	199.4959	0.8015	0.0977	-0.0504	-0.1360	196.2919

Table B.3

The Excess Properties of Ternary System n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1007	0.1011	-0.0643	-0.0270	27.1088	0.1023	0.0994	-0.0389	-0.0008	43.7219
0.1031	0.7990	-0.0245	-0.0093	19.9036	0.0985	0.8009	-0.0176	-0.0092	23.2771
0.2021	0.2018	-0.0871	-0.0273	54.4593	0.2025	0.2008	-0.0615	-0.0116	67.3570
0.2019	0.3946	-0.0695	-0.0246	56.0259	0.2038	0.3949	-0.0544	-0.0005	59.7426
0.1997	0.6040	-0.0493	-0.0183	37.9797	0.1998	0.6008	-0.0361	-0.0032	45.3176
0.3002	0.2943	-0.0845	-0.0279	69.4559	0.3023	0.2957	-0.0646	-0.0156	75.8224
0.3019	0.4009	-0.0704	-0.0245	62.9571	0.3033	0.3999	-0.0549	-0.0105	67.1963
0.4029	0.2007	-0.0997	-0.0285	76.5313	0.3984	0.2034	-0.0738	-0.0211	87.7062
0.4006	0.3009	-0.0827	-0.0275	69.9371	0.3991	0.3013	-0.0629	-0.0153	77.4867
0.4001	0.4047	-0.0651	-0.0213	57.4484	0.4024	0.4038	-0.0496	-0.0111	63.5534
0.6052	0.1973	-0.0744	-0.0280	67.1828	0.6028	0.1982	-0.0573	-0.0154	74.6263
0.7995	0.0998	-0.0471	-0.0167	45.1125	0.8027	0.0970	-0.0363	-0.0014	49.5778

Table B.3 (cont'd)

The Excess Properties of Ternary System n-Undecane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1030	0.1021	-0.0258	-0.0084	44.4774	0.0957	0.0937	-0.0210	-0.0160	40.5311
0.0996	0.8060	-0.0115	-0.0170	22.5813	0.1027	0.7995	-0.0113	-0.0127	20.4901
0.2019	0.1981	-0.0397	-0.0234	69.9167	0.2012	0.2007	-0.0350	-0.0326	66.0977
0.2021	0.3938	-0.0352	-0.0234	62.2473	0.1956	0.3991	-0.0299	-0.0178	59.3690
0.2005	0.6019	-0.0249	-0.0322	43.9667	0.1959	0.6024	-0.0208	-0.0097	44.1239
0.3030	0.2983	-0.0438	-0.0376	75.2479	0.2922	0.2978	-0.0370	-0.0335	73.5391
0.2996	0.4028	-0.0378	-0.0393	65.2549	0.2987	0.3959	-0.0321	-0.0323	65.6657
0.3968	0.2000	-0.0504	-0.0329	87.9114	0.3950	0.2005	-0.0422	-0.0374	87.4012
0.3732	0.3142	-0.0426	-0.0302	75.8661	0.3973	0.2983	-0.0365	-0.0346	76.6477
0.4028	0.3998	-0.0343	-0.0241	63.4933	0.3945	0.4067	-0.0291	-0.0286	61.8644
0.6018	0.1983	-0.0396	-0.0304	73.7624	0.6018	0.1966	-0.0341	-0.0324	72.4615
0.8003	0.1013	-0.0259	-0.0096	46.1014	0.7985	0.1008	-0.0223	-0.0235	46.2302

Table B.4

The Excess Properties of Ternary System n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	$\Delta^* G^E$ J/mol
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1051	0.0982	-0.1346	-0.0594	109.7461	0.1047	0.0988	-0.0946	-0.0518	129.1918
0.1085	0.7960	-0.0699	-0.0554	85.6110	0.1049	0.7983	-0.0548	-0.0396	87.2030
0.2089	0.2018	-0.1930	-0.0894	205.7855	0.2045	0.1972	-0.1489	-0.0914	216.5600
0.2045	0.3899	-0.1641	-0.0893	190.6378	0.2084	0.3920	-0.1326	-0.0829	199.9955
0.2074	0.5969	-0.1335	-0.0727	158.8920	0.2074	0.5969	-0.1048	-0.0644	170.3795
0.3024	0.2956	-0.2083	-0.1264	254.2039	0.2986	0.2974	-0.1666	-0.1181	262.0784
0.3061	0.4020	-0.1879	-0.1188	234.5260	0.3028	0.4005	-0.1512	-0.1038	243.7009
0.4009	0.1998	-0.2456	-0.1406	299.1287	0.4034	0.1996	-0.1950	-0.1470	318.7903
0.4030	0.2986	-0.2143	-0.1347	285.9683	0.4092	0.2977	-0.1755	-0.1365	294.0088
0.4037	0.4025	-0.1860	-0.1238	256.6140	0.3998	0.4065	-0.1476	-0.1187	270.9268
0.6185	0.1931	-0.1973	-0.1558	293.2410	0.6052	0.1990	-0.1628	-0.1515	311.1641
0.8009	0.1005	-0.1283	-0.1174	209.9505	0.8034	0.0973	-0.1049	-0.1171	220.0776

Table B.4 (cont'd)

The Excess Properties of Ternary System n-Octane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1061	0.1009	-0.0637	-0.0844	134.5120	0.0998	0.1140	-0.0512	-0.0523	128.5754
0.1086	0.7972	-0.0376	-0.0512	93.3709	0.1048	0.7955	-0.0310	-0.0571	92.8101
0.2076	0.1961	-0.1011	-0.1345	225.8880	0.2059	0.2011	-0.0848	-0.1225	225.0343
0.2048	0.3941	-0.0888	-0.1155	204.0085	0.2003	0.3951	-0.0732	-0.1216	203.6092
0.2113	0.5947	-0.0719	-0.1107	178.3614	0.2037	0.5988	-0.0590	-0.1055	176.7987
0.3057	0.2932	-0.1153	-0.1548	273.3484	0.3116	0.3083	-0.0959	-0.1673	276.6790
0.3113	0.3989	-0.1037	-0.1362	256.2871	0.2998	0.4051	-0.0851	-0.1563	255.0528
0.4065	0.1978	-0.1334	-0.1966	331.0244	0.4000	0.1977	-0.1123	-0.1989	332.4031
0.4081	0.2967	-0.1202	-0.1751	305.2209	0.3978	0.3018	-0.0999	-0.1913	307.4868
0.4017	0.4051	-0.1037	-0.1697	274.7217	0.3959	0.4064	-0.0869	-0.1771	278.5784
0.6080	0.1958	-0.1130	-0.2105	321.8547	0.6015	0.1995	-0.0951	-0.2052	330.2678
0.8052	0.0976	-0.0698	-0.1686	237.6542	0.8047	0.0986	-0.0580	-0.1583	245.5354

Table B.5

The Excess Properties of Ternary System n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol
Temperature = 293.15 K					Temperature = 298.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1002	0.1025	-0.0823	-0.0179	48.8507	0.0814	0.1024	-0.0472	-0.0025	54.3527
0.0993	0.8003	-0.0341	-0.0123	36.1522	0.0836	0.8163	-0.0257	0.0054	31.8426
0.1979	0.2021	-0.1186	-0.0233	87.8415	0.1700	0.2119	-0.0804	-0.0175	91.3940
0.1999	0.3961	-0.0999	-0.0337	83.3020	0.1652	0.4175	-0.0695	-0.0109	82.2407
0.2022	0.6023	-0.0664	-0.0264	74.2278	0.1696	0.6245	-0.0517	-0.0058	65.7117
0.2970	0.3010	-0.1169	-0.0435	117.3676	0.2514	0.3155	-0.0897	-0.0259	109.3107
0.2981	0.4012	-0.1043	-0.0388	103.6052	0.2547	0.4316	-0.0777	-0.0209	99.6963
0.3953	0.1983	-0.1429	-0.0526	132.1690	0.3479	0.2183	-0.1063	-0.0365	137.0568
0.4052	0.2985	-0.1235	-0.0415	119.0225	0.3490	0.3270	-0.0930	-0.0348	123.1802
0.3976	0.4006	-0.0981	-0.0446	109.2928	0.3494	0.4377	-0.0779	-0.0217	104.6465
0.6004	0.2004	-0.1114	-0.0417	123.6224	0.5485	0.2266	-0.0939	-0.0363	132.7614
0.8006	0.1023	-0.0708	-0.0223	81.2279	0.7675	0.1177	-0.0640	-0.0266	97.6979

Table B.5 (cont'd)

The Excess Properties of Ternary System n-Decane(1)-n-Tridecane(2)-n-Pentadecane(3)

Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol	Mole Fraction x_1	Mole Fraction x_2	Excess Viscosity 10^{-3} Pa.s	Excess Volume L/kmol	Δ^*G^E J/mol
Temperature = 308.15 K					Temperature = 313.15 K				
1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000
0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1003	0.1008	-0.0365	-0.0205	64.3680	0.1006	0.1034	-0.0298	-0.0327	65.9388
0.0996	0.7999	-0.0194	-0.0156	36.1574	0.0998	0.8028	-0.0156	-0.0287	36.9564
0.2013	0.1990	-0.0577	-0.0413	106.5707	0.1981	0.2033	-0.0478	-0.0467	105.5612
0.1997	0.4042	-0.0510	-0.0353	92.4779	0.1987	0.3985	-0.0419	-0.0493	94.9095
0.2000	0.6013	-0.0375	-0.0375	73.6574	0.2005	0.6017	-0.0315	-0.0418	73.8569
0.3010	0.3019	-0.0651	-0.0449	120.0112	0.2941	0.2992	-0.0537	-0.0515	121.2881
0.2998	0.4028	-0.0566	-0.0479	108.6108	0.3010	0.4011	-0.0464	-0.0546	112.0789
0.4008	0.2012	-0.0749	-0.0621	145.3748	0.3971	0.1948	-0.0625	-0.0788	148.3196
0.4006	0.3003	-0.0661	-0.0557	128.6335	0.3995	0.3033	-0.0546	-0.0642	130.3855
0.4008	0.3996	-0.0549	-0.0514	111.5819	0.4001	0.4007	-0.0459	-0.0627	112.3793
0.6014	0.2009	-0.0623	-0.0597	127.3654	0.6018	0.1979	-0.0520	-0.0717	131.0186
0.8038	0.0978	-0.0399	-0.0446	85.8556	0.8038	0.1000	-0.0324	-0.0529	88.5880

Appendix C

Error Analysis of Density Measurements and Excess Molar Volumes

C.1 Density Measurements

The equation used for calculating densities is given in Chapter 3 as

$$\rho = \frac{AT^2}{1 - BT^2} - C \tag{3.3}$$

To calculate the predicted error in the density value due to an error in T, the period of vibration, eq. (3.3) is differentiated to obtain

$$\frac{d\rho}{dT} = \frac{2AT}{(1 - BT^2)^2}$$

or

$$d\rho = \frac{2AT}{(1 - BT^2)^2} \tag{C.1}$$

The maximum value of T is 0.3421 seconds and the maximum fluctuation in density meter readings is 1 x 10^{-5} seconds in this study. The values of the calibration constants which are A = 16.35, B = -0.024 and C = 1.14.

Substituting all the values above into eq. (C.1), then the predicted error in density measurements is

$$d\rho = \frac{2 \times 16.35 \times 0.3421}{[1 - (-0.024) \times 0.3421^2]^2} \times 1 \times 10^{-5}$$

$$= 1.11 \times 10^{-4} \ \ kg/L$$

Therefore, the conclusion is

$$d\rho < 1.5 \times 10^{-4} \ \ kg/L \tag{C.2}$$

C.2 Excess Molar Volume of Mixing

The equation used to calculate excess molar volumes of mixing is given in Chapter 2 as

$$V^E = \frac{\sum_i x_i M_i}{\rho} - \sum_i \frac{x_i M_i}{\rho_i} \tag{2.106}$$

for a binary mixture, eq. (2.106) becomes

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \tag{C.3}$$

Subject to the constraint that

$$x_1 + x_2 = 1 \tag{C.4}$$

eq.(C.3) is rearranged as

$$V^E = \frac{M_1 x_1 + M_2(1 - x_1)}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{(1 - x_1)M_2}{\rho_2}$$
$$= \frac{x_1(M_1 - M_2) + M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{M_2 - x_1 M_2}{\rho_2} \tag{C.5}$$

The error in excess molar volume is due to the density measurements of the solution and the weight measurements in making the solution, that is, V^E is a function of ρ, ρ_1, ρ_2, w_1 and w_2, where w_1 and w_2 are the mass of component 1 and 2, respectively.

To calculate the predicted error in the excess molar volume of mixing, V^E is differentiated with respect to all its variables as

$$dV^E = \frac{\partial V^E}{\partial \rho}d\rho + \frac{\partial V^E}{\rho_1}d\rho_1 + \frac{\partial V^E}{\partial \rho_2}d\rho_2 + \frac{\partial V^E}{\partial w_1}dw_1 + \frac{\partial V^E}{\partial w_2}dw_2 \tag{C.6}$$

where

$$\frac{\partial V^E}{\partial w_1} = \frac{\partial V^E}{\partial x_1} \cdot \frac{\partial x_1}{\partial w_1} \tag{C.7}$$

$$\frac{\partial V^E}{\partial w_1} = \frac{\partial V^E}{\partial x_1} \cdot \frac{\partial x_1}{\partial w_2} \tag{C.8}$$

Substituting eqs. (C.7) and (C.8) into (C.6) yields

$$dV^E = \frac{\partial V^E}{\partial \rho}d\rho + \frac{\partial V^E}{\rho_1}d\rho_1 + \frac{\partial V^E}{\partial \rho_2}d\rho_2$$
$$+ \frac{\partial V^E}{\partial x_1} \cdot \frac{\partial x_1}{\partial w_1}dw_1 + \frac{\partial V^E}{\partial x_1} \cdot \frac{\partial x_1}{\partial w_2}dw_2 \tag{C.9}$$

In fact

$$d\rho_1 = d\rho_2 = d\rho \tag{C.10}$$

and

$$dw_1 = dw_2 = dw \tag{C.11}$$

Thus eq. (C.9) can be simplified as

$$dV^E = (\frac{\partial V^E}{\partial \rho} + \frac{\partial V^E}{\rho_1} + \frac{\partial V^E}{\partial \rho_2}) d\rho + [\frac{\partial V^E}{\partial x_1}(\frac{\partial x_1}{\partial w_1} + \frac{\partial x_1}{\partial w_2})] dw \tag{C.12}$$

From eq. (C.5):

$$\frac{\partial V^E}{\partial \rho} = \frac{-x_1(M_1 - M_2) - M_2}{\rho^2} \tag{C.13}$$

$$\frac{\partial V^E}{\partial \rho_1} = \frac{x_1 M_1}{\rho_1^2} \tag{C.14}$$

$$\frac{\partial V^E}{\partial \rho_2} = \frac{M_2 - x_1 M_2}{\rho_2^2} \tag{C.15}$$

$$\frac{\partial V^E}{\partial x_1} = \frac{M_1 - M_2}{\rho} - \frac{M_1}{\rho_1} + \frac{M_2}{\rho_2} \tag{C.16}$$

From the definition of mole fraction:

$$x_1 = \frac{\frac{w_1}{M_1}}{\frac{w_1}{M_1} + \frac{w_2}{M_2}} = \frac{w_1 M_2}{w_1 M_2 + w_2 M_1} \tag{C.17}$$

the differentiation of x_1 with respect to w_1 is

$$\frac{\partial x_1}{\partial w_1} = \frac{w_2 M_1 M_2}{(w_1 M_2 + W_2 M_1)^2} \tag{C.18}$$

and the differentiation of x_1 with respect to w_2 is

$$\frac{\partial x_1}{\partial w_2} = \frac{-w_1 M_1 M_2}{(w_1 M_2 + W_2 M_1)^2} \tag{C.19}$$

Substituting eqs. (C.13), (C.14), (C.15), (C.16), (C.17), (C.18) and (C.19) into (C.12) yields

$$\begin{aligned} dV^E = {} & [\frac{-x_1(M_1 - M_2) - M_2}{\rho^2} + \frac{x_1 M_1}{\rho_1^2} + \frac{M_2 - x_1 M_2}{\rho_2^2}]d\rho \\ & + [(\frac{M_1 - M_2}{\rho} - \frac{M_1}{\rho_1} + \frac{M_2}{\rho_2})[\frac{M_1 M_2 (w_2 - w_1)}{(w_1 M_2 + w_2 M_1)^2}]]dw \end{aligned} \tag{C.20}$$

Eq. (C.20) is the general equation used for error analysis of excess molar volumes.

From Figures 5.1 to 5.4, it is clear that the maximum value of V^E appears at system n-octane(1)-n-pentadecane(2) when temperature = 313.15 K and x_1 = 0.5. Therefore, data at this point are chosen to calculate the predicted error of excess molar volume. They are listed as follows:

$M_1 = 114.23$ g/mol $\rho_1 = 0.68647$ g/cm^3 $x_1 = 0.4956$ g/cm^3

$M_2 = 212.42$ g/mol $\rho_2 = 0.75454$ g/cm^3 $\rho = 0.73036$

$w_1 = 6.3222$ g $w_2 = 11.9656$ g

From eq. (C.2) $d\rho$ can take the value of 1.5×10^{-4} g/cm^3 and as indicated in Chapter 3 $dw = 1 \times 10^{-7}$ kg $= 1 \times 10^{-4}$ g. Substituting all the data above into eq. (C.20) yields

$$
\begin{aligned}
dV^E = {} & [\frac{-0.4956(114.23 - 212.42) - 212.42)}{0.73036^2} + \frac{0.4956 \times 114.23}{0.68647^2} \\
& + \frac{212.42 - 0.4956 \times 212.42}{0.75454^2}] \times 1.5 \times 10^{-4} \\
& + \{(\frac{114.23 - 212.42}{0.73036} - \frac{114.23}{0.68647} + \frac{212.42}{0.75454}) \cdot \\
& [\frac{114.23 \times 212.42 \ (11.9656 - 6.3222)}{(6.3222 \times 212.42 + 11.9656 \times 114.23)^2}]\} \times 1 \times 1^{-4} \\
= {} & 1.65 \times 10^{-4} \ cm^3/mol
\end{aligned}
$$

Therefore, the conclusion is

$$dV^E < 2 \times 10^{-4} \ cm^3/mol = 2 \times 10^{-4} \ L/kmol \qquad (C.21)$$

Appendix D

The Results of Testing Purity of the Chemicals by Gas Chromatographic Analysis

The purities of the n-alkanes used in this study were checked by gas chromatography. The detector type is the Flame Ionization Detector (FID) and the column type is a 5 m x 0.53 mm methyl - silicon capillary column. The results are listed as follows:

Reagent	Stated Purity (mol%)	Actual Purity (mol%)
n-octane	99+	99.7
n-decane	99+	99.8
n-undecane	99	99.9
n-tridecane	99+	99.5
n-pentadecane	99+	99.9

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