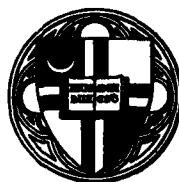


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COMPOSITION AND TEMPERATURE
DEPENDENCE OF SHEAR VISCOSITY
OF HYDROCARBON MIXTURES.



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VITREOUS STATE LABORATORY

The Catholic University of America
Washington, D.C. 20064

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DEPENDENCE OF SHEAR VISCOSITY
OF HYDROCARBON MIXTURES.

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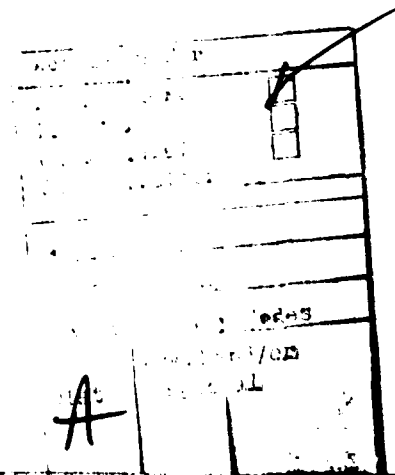
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I. INTRODUCTION

Characterization of the viscosity of hydrocarbon fuel mixtures is of considerable importance, because the viscosity directly controls atomization in combustors and affects the performance of other engine components such as fuel pumps. Typical aircraft fuels are blends and are exposed to operating temperatures which may range from -55 to 45°C , a temperature range which can correspond to viscosity changes of from 1 to 3 orders of magnitude, depending on the molecular weight of the fuel. Consequently an understanding of hydrocarbon fuel viscosities useful from an engineering standpoint demands an account of the combined effects of temperature and composition on the viscosity.

A truly fundamental theory would predict the viscosity, along with other thermodynamic and transport properties, from knowledge of the intermolecular forces and radial distribution functions alone. Such a theory has had appreciable success in application to pure, simple liquids such as the liquefied rare gases. For solutions, however, although the general theory has been formulated (1), it has not yet been reduced successfully to numerical results. One is thus forced to approximate approaches, of which two general types may be distinguished. The first in which the discrete molecular nature of the solvent is neglected has been remarkably

successful in explaining the viscosity of dilute solutions of high polymers. The second general approach is to correlate the viscosity of the mixture with the properties of the pure components and thermodynamic parameters characteristic of the interaction between components.

Prediction of the viscosity of liquid mixtures has been a goal of long standing. However, studies reported in the literature on viscosities of organic and hydrocarbon mixtures have never looked at the temperature dependence of additivity rules over the -50 to 50°C range important for fuels. Consequently we have carried out a study of the effects of temperature and composition on the shear viscosities of blends of hydrocarbons of the type found in jet fuel kerosenes. The combined temperature-composition dependence of viscosities of five pure C_{10} hydrocarbons and their different mixtures and of binary $C_{10} - C_{14}$ hydrocarbon system have been studied, and the results evaluated.*

II. REVIEW OF LITERATURE

The two aspects of liquid viscosity of interest here have usually been considered separately in the literature, namely, viscosity as a function of temperature for pure liquids, and the isothermal dependence of viscosity on composition for mixtures.

A. Simple Theories for Viscosity as a Function of Temperature for Pure Liquids

* The experimental viscosity measurement on the binary $C_{10} - C_{14}$ mixture (i.e., mixtures of HNN and XTHCDPD) were supported by Contract No. F33615-78-C-2000 from the Air Force Systems Command. They are reported here for the sake of completeness.

A correct molecular theory of liquid viscosity would be based on a detailed consideration of the characteristic molecular dynamics of the liquid state. Present theories of liquid viscosity leave a lot to be desired in this respect as has been pointed out in the review by Brush (2).

The fundamental question that must be addressed is how momentum is transferred from one molecule to another. In gases momentum is transferred primarily by molecular collisions and to a lesser extent by inter-atomic forces. In liquids it is an open question as to whether or not it is the attractive or the repulsive forces that are most important in controlling the viscosity. What is agreed upon and what is firmly established in the theories of Brownian motion of colloidal particles is that liquid viscosity is intimately associated with the diffusion of a colloidal particle. The diffusion coefficient D and the viscosity η are inversely proportional to one another. The Stokes - Einstein equation obtained from hydrodynamics and the theory of Brownian motion adequately correlates experimental liquid shear viscosities η and self-diffusion coefficient D for many liquids (3a, 3b)

$$D\eta/kT = (1/2\pi)(N_a/\bar{V})^{1/3} \quad (1)$$

where N_a is Avogadro's number, $\bar{V}$ the molar volume, T the temperature, and k the Boltzmann constant. Therefore, the problem of determining momentum transfer in a liquid can be converted into the conceptually simpler problem of determining the factors that govern molecular diffusion. Hence many viscosity theories for liquids are essentially theories of diffusion. One obvious goal of such theories is to obtain a relationship between equation-of-state properties (pressure-volume-temperature) and the viscosity of a liquid. This makes it possible to infer viscosity

behavior from PVT behavior. In this respect one of the early equations, and certainly one of the most popular, relating the shear viscosity of a liquid to temperature (4, 5, 6) is the so-called Andrade equation:

$$\eta = A \exp(B/T) \quad (1a)$$

where A and B are constants. A relationship of this kind was suggested empirically by Arrhenius (5) and by Guzman (6) and derived theoretically by Andrade (4).

Eyring (7) gave the equation added theoretical significance in the following form:

$$\eta = h(\lambda_1/\lambda_2\lambda_3\lambda^2)\exp(\Delta G^*/RT) \quad (2)$$

where λ_1 , λ_2 , λ_3 and λ are characteristic distances shown in Fig. 1, R is the ideal gas constant, ΔG^* the molar activation free energy, and h is Planck's constant.

If λ is assumed equal to λ_1 , and $\lambda_1\lambda_2\lambda_3$ is identified with molecular volume, Eq. (2) may be written as:

$$\eta = (hNa/\bar{V})\exp(\Delta G^*/RT) \quad (3)$$

For kinematic viscosity ν Eq. (3) can be written as:

$$\nu \equiv \eta/\rho = (hNa/M)\exp(\Delta G^*/RT) \quad (4)$$

where M is the molecular weight.

Eyring's picture of shear between two layers of liquid involves the successive passage of individual molecules from one equilibrium position to another as indicated in Fig. 1. Such a passage requires that either a hole or a site be available. The production of such a site requires the expenditure of energy to push back other molecules. The movement of the molecule may be regarded as the passage of the system over a free energy barrier of height ΔG^* .

Further, since ΔG^* , the Gibbs free energy of activation, may be replaced by $\Delta H^* - T\Delta S^*$, it is seen that

$$\eta = [(hNa/\bar{V})\exp(-\Delta S^*/R)]\exp(\Delta H^*/RT) \quad (4a)$$

where ΔH^* and ΔS^* are enthalpy and entropy of activation. Since the molar volume of a liquid $\bar{V}$ does not vary greatly with the temperature, if ΔH^* and ΔS^* are taken as constant, Eq. (4a) takes the form of Eq. (1a).

Eq. (1a) predicts a linear plot of $\ln \eta$ vs. $1/T$ for liquids. Experimentally, viscosity data which cover a sufficient range (>1 order of magnitude in η) do not give such plots. Rather, $\ln \eta$ versus $1/T$ plots are usually concave upwards.

Some other investigators (Lennard - Jones and Devonshire (8); Glasstone, Laidler and Eyring (7); Frenkel (9); Fowler and Guggenheim (10)) have employed the concept of the "Free Volume" in statistical thermodynamic theories of the liquid state. According to Glasstone, Laidler and Eyring the free volume may be regarded as the volume in which each molecule of a liquid moves in an average potential field due to its neighbours. However, theoretical estimates of free volume depend on postulates regarding the compressibilities of the molecules and the nature of their packing in the liquid state. A definition of free volume V_f often used is that employed by Doolittle (11):

$$V_f = V - V_o \quad (5)$$

where V is the measured volume at temperature T and V_o , which is termed the "occupied volume", is the close packed volume of the liquid. Generally V_f is presumed to vanish on isobaric cooling at some finite temperature T_o .

The free volume V_f is a time-average quantity and an equilibrium

property of the system. In a liquid-like system, however, the local free volume is continually being redistributed throughout the medium, the redistribution occurring simultaneously with the random thermal motions of the molecules. The basic idea underlying the free-volume approach is that the molecular mobility at any temperature is dependent on the available free volume at that temperature. As temperature increases, the free volume increases and molecular motions become more rapid.

Doolittle (11) found that the temperature dependence of the shear viscosity of low molecular weight hydrocarbon liquids could be represented by an empirical equation of the following form:

$$\eta = a \exp (b/f) \quad (6)$$

where a and b are constants and $f = V_f/V$ is the fractional free volume.

Cohen and Turnbull (12) gave this expression a theoretical basis by showing that Eq. (6) can be derived by assuming a distribution of hole sizes, so that the jump probability is determined only by the chance of a molecule finding an adjacent local free volume of sufficient size to jump into. Assuming a quasicrystalline liquid structure, a molecule can be pictured as vibrating about an equilibrium position until a combination of two events occurs: (1) the molecule attains sufficient energy to overcome the attractive forces holding it to its neighbors, and (2) an empty site is available into which the molecule can jump. Therefore, the probability of a transition or jump from one site to another P_j is given by the expression

$$P_j = P_E \times P_V$$

where P_E is the probability of attaining sufficient energy to break bonds and P_V is the probability that there is sufficient local free volume for

a jump to occur. Cohen and Turnbull consider a liquid consisting of hard spheres with only repulsive forces. Each molecule is confined in a cage made up of its neighbors. For diffusive motion to occur there must appear a fluctuation in the local density which opens up a hole large enough to permit a significant displacement of the molecule within the cage. They assume such fluctuations occur without energy change, i.e., $P_E = 1$. They associate a local free volume with each molecule and assume a distribution of local free volumes or hole sizes. In this way the molecules can be characterized by their associated free volumes. They next calculate an expression for the excess entropy due to this distribution of local free volume V and then find the distribution of v by maximizing this excess entropy holding the number of molecules and total free volume constant. They find the distribution of hole sizes $P(v)$ to be

$$P(v) = (\gamma/v_f) \exp(-\gamma v/v_f) \quad (7)$$

where $P(v)$ is the probability of finding the free volume v nearby. The average free volume per molecule is v_f . The constant γ is a numerical factor needed to correct for the overlap of free volume between molecules and lies between 1/2 and 1. Assuming that some minimum local free volume v^* is necessary for a jump to occur, one can calculate the probability of finding v^* and thus the jump probability. This is of course simply the quantity P_v defined above and is given by

$$P_v = \int_{v^*}^{\infty} P(v) dv = \exp(-\gamma v^*/v_f) \quad (8)$$

Since the quantity v^* should be close to v_0 , the close packed molecular volume, the hard-sphere viscosity equation takes the form

$$\eta = A/P_v = A \exp(\gamma v_0/v_f) \quad (9)$$

Cukierman, Lane, and Uhlmann (13) have adopted the formulation due to Turnbull and Cohen, and for the case of small free volume, they have expressed the self-diffusion coefficient for transport in a liquid by

$$D = (\bar{u}V^*/3)\exp(-V^*/V_f) \quad (10)$$

where $\bar{u}$ is the gas kinetic velocity and may be expressed by $\bar{u} = (3kT/m)^{1/2}$, where m is mass of the molecule; and α is a geometric factor relating the molecular displacement to the free volume.

In relating this expression to the shear viscosity, it is customary to assume that D is inversely proportional to η :

$$D = b/\eta \quad (11)$$

For relatively simple organic liquids, the Stokes - Einstein expression provides a useful estimate of b , at least in the range of high fluidity:

$$b = kT/3\pi a_0 \quad (12)$$

where a_0 is the molecular diameter. Using Eqs. (10) - (12), and taking $\alpha V^* = \beta a_0^3$ and $\gamma V^* = \delta V_m$ (V_m being the molecular volume). Cukierman, Lane and Uhlmann have expressed the viscosity as

$$\eta = (0.2/\beta a_0^2)(mkT)^{1/2}\exp(\delta V_m/V_f) \quad (13)$$

For a fractional free volume of f_d at temperature T_d , and assuming that the glassy expansion coefficient provides a good estimate of the van der Waals expansion in the liquid-liquid range, Eq. (13) becomes

$$\eta = (0.2/\beta a_0^2)(mkT)^{1/2}\exp\{c/[f_d - f_{d0}(T - T_d)]\} \quad (14)$$

This expression is equivalent to a modified Vogel - Fulcher expression

$$\log_{10} \eta = A + (1/2)\log T + B/(T - T_0) \quad (15)$$

where $10^A = (0.2/\beta a_0^2)(mk)^{1/2}$, $B = c/2$, and $T_0 = T_d - (f_d/f_{d0})$.

i.e. T_0 is the temperature where free volume vanishes on isobaric cooling.

Recently, Laughlin and Uhlmann (13) have reported a study of viscous flow in a number of relatively simple organic liquids. They have concluded that for none of the liquids investigated by them, the flow behavior over the full range of measured viscosity can be adequately represented by any of the standard theoretical models. However, they note that agreement is found between molecular dynamic calculations and a modified free volume theory. In addition, the free volume theory gives a good description of viscous flow at high temperature. Hence they suggest that free volume theories are most appropriately used to describe flow in the high-temperature region rather than in the region around the glass transition. They have also suggested that a second, easier flow process becomes dominant in the low temperature region.

III. VISCOSITY OF MIXTURES

The viscosity of liquid mixtures has attracted much attention in the literature, both from the practical and theoretical standpoints. But it should be mentioned that none of these papers have considered mixture viscosities over a sizable temperature range.

1. "Ideal" Equations

For ideal miscible liquid mixtures, many simple additivity rules for mixture viscosities have been suggested. Some of these expressions for a binary system are as follows (14, 15)

$$\eta_{\text{mix}}^{-1} = \eta_1^{-1} X_1 + \eta_2^{-1} X_2 \quad (16)$$

$$\eta_{\text{mix}} = \eta_1 X_1 + \eta_2 X_2 \quad (17)$$

$$\ln \eta_{\text{mix}} = X_1 \ln \eta_1 + X_2 \ln \eta_2 \quad (18)$$

Here η_{mix} , η_1 and η_2 are the viscosities of the mixture and pure components 1 and 2 respectively, X_1 and X_2 are the mole fractions of the components 1 and 2 in the mixture. It may be pointed out that these and similar other simpler forms have not proved successful even for the prediction of the viscosities of mixtures of gases at ordinary pressures.

2. Equations Based on Transition State Theory

a. One parameter equation for binary mixtures

Katti and Chaudhri (16) have assumed the transition state theory expression for viscosity and assumed that the activation free energies of mixtures can be approximated by a regular solution theory.

The general form of the transition state expression for viscosity obtained by rearranging Eq. (3) is

$$\Delta G^*/RT = \ln(\bar{V}/hNa)(\eta/\Lambda)^{-1} \quad (19)$$

For the mixture, they have assumed:

$$\Delta G_{\text{mix}}^*/RT = X_1(\Delta G_1^*/RT) + X_2(\Delta G_2^*/RT) + X_1 X_2(W/RT) \quad (20)$$

where W is the excess heat of mixing parameter that appears in regular solution theory. Also they assume:

$$\ln (\eta_{mix}/\lambda_{mix}) = X_1 \ln (\eta_1/\lambda_1) + X_2 \ln (\eta_2/\lambda_2) \quad (21)$$

Using Eq. (19) for the respective mixture and pure component viscosities then gives

$$\log \eta_{mix} V_{mix} = X_1 \log \eta_1 V_1 + X_2 \log \eta_2 V_2 + X_1 X_2 (W/RT) \quad (22)$$

where the V's are the molar volumes.

b. Two parameter equation

Heric (17) has extended the regular solution approach of Katti and Chaudhri to multicomponent systems and obtained the following expression for kinematic viscosity ν :

$$\log \nu = \sum_{i=1}^n X_i \log \nu_i + \sum_{i=1}^n X_i \log M_i - \log \sum_{i=1}^n X_i M_i + \delta_{i...n} \quad (23)$$

If only binary interaction terms are assumed necessary, then Katti and Chaudhri's theory gives:

$$\delta_{i...n} = (1/2) \sum_{i=1}^n \sum_{j=1}^n X_i X_j \alpha_{ij} \quad (24)$$

where $\delta_{i...n}$ is a deviation function, representing departure from a noninteracting system, α_{ij} is an interaction parameter with $\alpha_{ij} = \alpha_{ji}$ and $\alpha_{ii} = \alpha_{jj} = 0$.

Two aspects of Eq. (23) must now be taken into account. The first one - the use of a single α_{ij} term for representation of each binary interaction - has been considered by Heric, using the data of Katti and Chaudhri. He found that Eq. (24) did not give an adequate fit to the data and concluded that in a multicomponent system, assuming binary interaction only, a relationship containing more parameters than Eq. (24) was required:

$$\eta = \eta_0 \exp \left[\sum_{i,j} x_i x_j [\tau_{ij} + \alpha_{ij}(\lambda_i - \lambda_j)] \right] \quad (25)$$

The second aspect of Eq. (23) - the adequacy of assuming binary interactions only - has been considered again by Heric (17) with viscosity data on the ternary system reported by Kalidas and Laddha (18). He found that the assumption of only binary interactions inadequate to describe the data exactly and for a ternary system Eq. (25) must be expanded by inclusion of a term for ternary interaction.

It may be anticipated that, for systems higher in number of components, higher interaction terms may well be required. Thus in quaternary systems, for example, a term in $x_1 x_2 x_3 x_4$ would be required.

3. Free Volume Approach

It was mentioned previously that theories relate the viscosities of liquids either to the activation energy required for the molecule to overcome the attractive forces of its neighbors and flow to a new position (absolute reaction rate theory) or to the probability that an empty site exists near a molecule (free volume theory). Macedo and Litovitz (19) have made the hypothesis that the two effects are combined, so that the probability for viscous flow is taken as the product of probability for acquiring sufficient activation energy and of the occurrence of an empty site. Similar assumptions can be made for solutions.

Upon combining the absolute reaction rate and free volume theories, one obtains (19) for the viscosity of the solution

$$\eta = A \exp \left[\left(G^*/RT \right) + \left(V^*/V_f \right) \right] \quad (26)$$

An equation of the same form holds for the pure components

$$n_i = A \exp[(\Delta G_i^*/RT) + (V_i^*/V_{fi})] \quad (i=1,2) \quad (27)$$

where the subscript labels the property of pure component i . Considering the fundamental assumptions of absolute rate theory and following Rosevear, et al., (20) one now assumes

$$\Delta G^* = X_1 \Delta G_1^* + X_2 \Delta G_2^* - \alpha \Delta G^R \quad (28)$$

where ΔG^R , the excess free energy of activation, is closely related to the excess free energy of mixing, and α is a constant of order unity.

Substitution of Eq. (28) in equation (26), taking logarithms of the resulting equation and of Eq. (27), and simple manipulation then yields

$$\ln n = X_1 \ln n_1 + X_2 \ln n_2 - (\Delta G^R/RT) + V^*[(1/V_f) - (X_1/V_{f1}) - (X_2/V_{f2})] \quad (29)$$

The excess free energy of activation can, in turn, be broken down into enthalpy and entropy contributions

$$\Delta G^R = \Delta H_M - T\Delta S^R \quad (30)$$

ΔH_M is the enthalpy of mixing per mole of solution, and ΔS^R is the residual entropy per mole.

The reduced volume could be defined as:

$$\bar{V} = V/V^* \quad (31)$$

Substitution of Eqs. (5), (31) and (30) into Eq. (29) yields

$$\ln n = X_1 \ln n_1 + X_2 \ln n_2 - (\Delta H_M/RT) + (\Delta S^R/R) + ([1/(\bar{V} - 1)] - X_1/(\bar{V}_1 - 1) - X_2/(\bar{V}_2 - 1))] \ln n_{fd} + \ln n_H + \ln n_S + \ln n_V \quad (32)$$

This equation displays explicitly the various contributions to the mixture viscosity: the ideal viscosity, the enthalpy and residual entropy of mixing, and the difference in free volume between solution and pure components.

Jambon and Delmas (21) have obtained ΔG^R from the experimental ΔH_M and the calculated ΔS_M^R . The expression for ΔS_M^R , the excess entropy of mixing, is

$$\Delta S_M^R = 3X_1(P_1^*V_1^*/RT_1^*)\ln[(\bar{V}_1^{1/3}-1)/(\bar{V}^{1/3}-1)] + 3X_2(P_2^*V_2^*)/(RT_2^*)\ln[(\bar{V}_2^{1/3}-1)/(\bar{V}^{1/3}-1)] \quad (33)$$

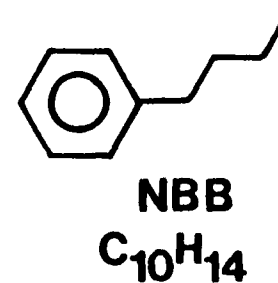
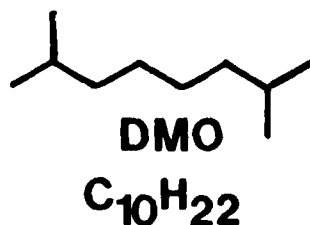
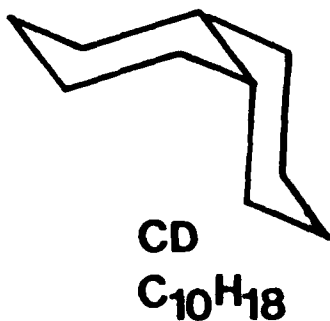
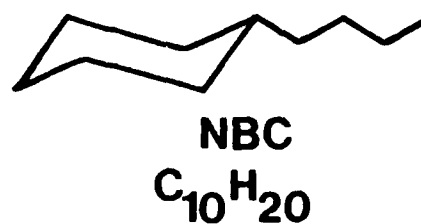
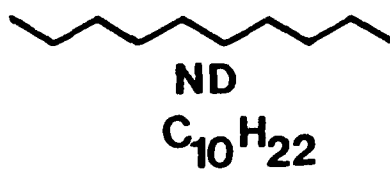
P^* , V^* , and T^* are respectively the reduced pressure, volume, temperature reduction parameters. They are obtained for the pure component through the equations $\bar{V} = (4\alpha T/3 + 1)/(\alpha T + 1)$ and $V^* = V/\bar{V}$ where α is the expansion coefficient. $\bar{T}$ is obtained from $\bar{T} = \bar{V}^{-1}(1 - \bar{V}^{-1/3})$ and $T^* = T/\bar{T}$. P^* is obtained from the thermal pressure coefficient, γ : $P^* = \gamma \bar{V}^2 T$.

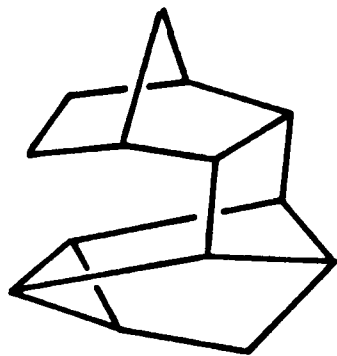
According to Bloomfield and Dewan (23), it is not clear a priori whether all of the contributions to the mixture viscosity in Eq. (32) are equally important and should be considered together in computing η . They have shown that the experimental viscosities are best reproduced by either the absolute rate theory alone or the free volume theory alone, for which agreement within theory and experiment is generally within a few percent.

IV. EXPERIMENTAL PROCEDURE

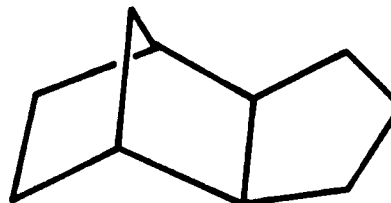
A. Materials.

Seven liquids were used in our study: six C_{10} hydrocarbon liquids - n-Decane (ND), n-Butylcyclohexane (NBC), cis-Decahydronaphthalene (CD), n-Butylbenzene (NBB), 2,7-Dimethyloctane (DMO), exo-tetrahydrodicyclopentadiene (XTHDCPD), and one hydrogenated dimer of norbornadiene (hexacyclic endo-endo-dihydrodinorbornadiene, HNN). The structures of these compounds and their formulae are given below.





HNN
C₁₄H₁₈



XTHDCPD
C₁₀H₁₆

Samples of n-Decane (99+%), n-Butylcyclohexane (99+%), cis-Decahydronaphthalene (99%), and n-Butylbenzene (99+%) were obtained from the Aldrich Chemical Company, Milwaukee, Wisconsin. 2,7-Dimethyloctane (99%) was obtained from Chemical Samples Company, Columbus, Ohio. XTHDCPD (99.9%) was obtained from Dr. A. Schneider of Suntech, Inc. These were used without further purification. Impure HNN was obtained from Dr. A. Schneider of Suntech, Inc. Pure HNN (99.9%) was prepared from M. Shahriari and R. Mossadegh by triple recrystallization from acetone at dry ice temperature, followed by vacuum distillation.

For the first five C₁₀ hydrocarbons (ND, NBC, CD, NBB and DMO)

shear viscosities η and densities ρ were measured for the five neat liquids, all ten binary 0.5 - 0.5 mixtures, four ternary 1/3 - 1/3 - 1/3 mixtures, one quaternary 0.25 - 0.25 - 0.25 - 0.25 mixture, and one quinternary 0.2 - 0.2 - 0.2 - 0.2 - 0.2 mixture. In addition, η and ρ were determined for the 0.25 - 0.75 and 0.75 - 0.25 CD/DMO and NBC/DMO binary mixtures. Finally η and ρ were determined for the pure HNN XTHDCPD and four different (0.8 - 0.2, 0.6 - 0.4, 0.4 - 0.6, and 0.2 - 0.8) HNN/XTHDCPD binary mixtures. All of the above compositions are in mole fraction. The temperature ranges of measurement were from 50°C down to either about -55°C or the temperature above this where the sample crystallized. The melting points of pure NBC (-75°C), DMO (-55°C), NBB (-88°C), and XTHDCPD (-91°C) are at the lower limit of or below the temperature range of this study. The melting points of CD (-43°C) and of HNN (8°C) are within this range, but CD and HNN were stable under supercooling. CD was never observed to crystallize, while measurements on pure HNN could be extended down to -51°C without crystallization. HNN-XTHDCPD mixtures did not crystallize. ND (mp = -30°C) and most of the mixtures containing substantial amounts of it could be supercooled very little, if at all, and crystallized quite reproducibly whenever the temperature dropped below a certain critical value. These temperatures were about -31°C for pure ND, -40°C for 0.5 ND - 0.5 NBC, -44°C for 0.5 ND - 0.5 CD, -45°C for 0.5 ND - 0.5 NBB, -51°C for 1/3 ND - 1/3 NBC - 1/3 CD and -51°C for 1/3 ND - 1/3 CD - 1/3 NBB.

B. Experimental Apparatus and Procedure

Kinematic viscosities ν were measured with factory calibrated Cannon - Ubbelohde semi-micro capillary viscometers from the Cannon Instrument Co., State College, Pennsylvania. Six different viscometers of constants 0.004142, 0.01681, 0.0937, 0.2515, 1.099 and 7.98 cSt/s were used in this study. To prevent the condensation of moisture inside the viscometers at low temperature, all openings of the viscometers were protected with small drying tubes containing anhydrous calcium sulfate (Drierite).

For highly viscous liquids such as HNN and some of its mixtures the viscometer efflux times were in the range 100 to 1700 s. For lower viscosity liquids, the efflux times were usually between 60 and 600 s. Each efflux time was measured at least twice and, more commonly, at least three times with a repeatability of 0.1 to 0.2%. Prior to each experiment the viscometers were rinsed with acetone to dissolve any hydrocarbon remaining from a previous experiment, followed by cleaning with detergent, rinsing with deionized water and drying at 110°C.

Densities ρ were measured at the same time as the viscosities in a dilatometer of about 11 mL volume constructed by sealing a portion of a graduated 5 mL pipette stem to a 10 mL volumetric flask. Liquid volumes could be read to the nearest 0.01 mL. The dilatometer was calibrated using deionized water as a density standard.

The viscometers and dilatometer were thermostatted to $\pm 0.05^\circ\text{C}$ or better in a stirred and regulated water bath above room temperature and in a stirred and regulated refrigerated 4-liter methanol bath contained in an unsilvered dewar flask below room temperature. Except for a narrow window to allow observation of the dilatometer and viscometer,

the methanol bath was fully covered with aluminum foil for better prevention of heat leakage.

Bath temperatures were determined with an accuracy - 0.02°C with a copper-constantan thermocouple which had been calibrated by us against a Leeds & Northrup model 8167-25 Pt resistance thermometer, serial no, 1812215. The Pt thermometer had been calibrated at National Bureau of Standards at the triple point of H_2O and the tin and zinc melting points. Thermocouple emf's were measured with a Leeds & Northrup type K-4 potentiometer.

Mixtures with a specified composition (in mole fraction) were prepared by direct weighing from the pure components.

Experimental Results and Data Analysis

Density

The experimental values of density ρ for all the liquids studied are given in Table I. The liquid densities ρ of all the pure compounds and their mixtures were found to vary linearly with temperature. Density results are given in Table II in the form of parameters obtained from least squares fits of the experimental data to the equation

$$\rho(\text{g}/\text{cm}^3) = \rho(0) - bT(^{\circ}\text{C}) \quad (34)$$

where $\rho(0)$ is the density at 0°C . The volume measurement is the only important source of error in the density. The overall accuracy of the density is estimated to be 0.1%. Duplicate density results (Runs I and II) were obtained for five of the entries in Table II and agree within our estimated uncertainty of 0.1% or better, as may be seen by

Comparison of results of duplicate viscosity experiments

Molar Composition	T(°C)	η (CP) Run 1	Run 2	% diff
ND	50	0.612	0.614	0.3
	0	1.291	1.292	0.1
	-30	2.572	2.569	0.1
NBC	50	0.836	0.839	0.4
	0	1.889	1.890	0.1
	-30	4.16	4.16	0.0
	-55	11.35	11.37	0.2
CD	50	1.848	1.844	0.2
	0	5.53	5.51	0.4
	-30	15.67	15.59	0.5
	-55	57.1	56.7	0.7
NBB	30	0.897	0.905	0.9
	0	1.469	1.467	0.1
	-30	3.036	3.010	0.9
	-55	7.69	7.66	0.4
0.5ND - 0.5NBC	50	0.703	0.700	0.4
	0	1.523	1.524	0.1
	-30	3.147	3.140	0.2
	-40	4.32	4.29	0.7

comparing the values of $\rho(o)$ and the density temperature coefficients b .

Shear Viscosities

The experimental values of shear viscosity η are listed in Table I for all the liquids studied. In Figs. 2, 2a, 2b are shown the typical examples of the temperature dependence of the shear viscosity in form of Arrhenius plots of $\log \eta$ versus $10^3/T(K)$. The plots are curved, but may be described well by the semi-empirical Vogel-Tammann-Fulcher (VTF) equation.

$$\ln \eta(P) = A + B/[T(K) - T_0] \quad (35)$$

where A , B , and T_0 are constants. The η versus T data for all liquids studied were fit to Eq. (35) in the following fashion. A value of T_0 was chosen, the corresponding parameters A and B evaluated by a linear least squares fit of $\ln \eta$ versus $1/(T - T_0)$ and the standard deviation, Std. Dev. $\ln \eta$, of the experimental data from the least squares line calculated. The value of T_0 was changed and the fit repeated until the value of T_0 (to the nearest 1 K) correspond to a minimum in Std. Dev. $\ln \eta$ was located.

In Table III the best fit parameters for Eq. (35) are listed for the liquids whose viscosity was studied. Std. Dev. $\ln \eta$ is typically about 0.003 for the 5 C_{10} hydrocarbons and their mixtures, and about 0.004 for HNN, XTHDCPD and their mixtures. Since $\Delta \ln \eta = \Delta \eta / \eta$, this means that the scatter of the data from the best fit curves is typically about 0.3% and 0.4%, so that the parameters of Table III may be used to calculate the viscosities of the respective liquids rather precisely over the temperature ranges indicated.

The shear viscosities reported here are accurate to within 0.5%.

This accuracy has been estimated from the corresponding uncertainties in temperature control and in the measurements on the times of flow, the densities and the viscometer calibration constants.

In Table IV shear viscosities and densities of some of the liquids studied in this work are compared with those of a number of different investigators. At a given temperature the densities measured in different laboratories agree with ours well within the experimental error. The disagreement between our viscosity measurements and those of other investigators is at worst 2%. This is acceptable in view of the many possible sources of error in viscosity measurements.

V. DISCUSSION OF TEMPERATURE / COMPOSITION DEPENDENCE OF VISCOSITY

A. Preliminary Comments

We consider here only simple equations with few adjustable parameters to describe composition and combined composition/temperature dependence of viscosity.

B. "Ideal" Equations

We tested three simple additivity rules for expressing mixture viscosities in terms of the viscosities of the pure components.

1. Additivity of viscosities:

$$\eta = \sum X_i \eta_i \quad (36)$$

2. Additivity of fluidities:

$$1/\eta = \sum X_i / \eta_i \quad (37)$$

3. Logarithmic additivity of viscosities or fluidities:

$$\ln \eta = \sum X_i \ln \eta_i \quad (38)$$

where X_i is the mole fraction and η_i the viscosity of pure component i .

Eq. (36) grossly overestimated mixture viscosities and will not be discussed further. Tests of Eqs. (37) and (38) are illustrated in Table V, where ratios of calculated to experimental mixture viscosities are tabulated at two temperatures and in Figs. 3, 4, 5, where isotherms of $\log \eta$ vs. X_{CD} , X_{NBC} and X_{HNN} are plotted for the CD-DMO, NBC-DMO and HNN-XTHDCPD binary systems.

Eq. (38) uniformly tends to overestimate mixture viscosities, and the errors become larger the greater the difference between the viscosities of the pure components, i.e., the lower the temperature.

Eq. (37), which assumes isothermal additivity of fluidities, gives the best agreement with the experimental viscosities. Two general trends are immediately apparent from the ratios of calculated to experimental viscosity in Table V.

1. For binary mixtures of the saturated hydrocarbons (ND, NBC, CD, DMO) and also for the binaries of HNN-XTHDCPD, Eq. (37) uniformly underestimates viscosities, and the errors become larger the larger the differences in the pure component viscosities, i.e., the lower the temperature.

2. For binary mixtures of the aromatic hydrocarbon NBB with the saturated hydrocarbons Eq. (37) overestimates the mixture viscosities.

These two effects, i.e., viscosity underestimates which become larger the greater the difference in component viscosities and viscosity overestimates when a saturated and aromatic hydrocarbon are mixed, compensate one another in mixtures in which both effects are important. For example, in the quaternary mixture Eq. (37) correctly predicts the viscosity within 2% or better. This is apparently due to the compensating effects of the high viscosity component CD and the aromatic NBB.

C. Katti and Chaudhri Approach for Binary Mixtures

The Katti and Chaudhri model (16) should be capable of accounting for the combined temperature/composition dependence of binary mixture viscosities using a single adjustable parameter, W . We can rewrite their expression (Eq. (22)) in the form:

$$W/R = [T/X_2(1 - X_2)][\ln Y - X_1 \ln Y_1 - X_2 \ln Y_2]$$

where Y stands for either η or ηV . According to Katti and Chaudhri's theory, W/R should be a constant over the full range of temperature for any binary mixture.

We have calculated values of W/R from the experimental viscosity data for three binaries. The results are shown in the Tables VI, VII, and VIII. To estimate the uncertainty in W/R , we have used the expression,

$$[T/X_2(1 - X_2)][\sigma^2 + X_1^2 \sigma_1^2 + X_2 \sigma_2^2]^{1/2}$$

where σ is the uncertainty in $\ln Y$ and similarly for σ_1 and σ_2 . σ , σ_1 and σ_2 were set equal to the standard deviations from the best fit expressions to the VTF equation (Table III).

For the two binaries (CD-DMO and HNN-XTHDPCD) in which there are

large differences in the viscosities of the two pure components and in which there are large deviations from additivity in $\ln \eta$, W/R is not constant as predicted by this model, but shows definite trends within experimental error. In particular, at constant temperature W/R increases with increasing mole fraction of the more viscous component. At constant composition, W/R increases with decreasing temperature.

$\ln \eta$ in the NBC-DMO system is nearly additive, so that the W/R values are small. Hence in this system no trend in W/R can be detected within experimental error.

D. Free Volume Additivity Equations

1. Introduction

Of the simple viscosity theories the free volume treatment has been most successful in accounting for the temperature dependence of organic liquid viscosities in the 10^{-2} to 10^4 P range. According to this theory, the shear viscosity is given by Eq. (13):

$$\ln \eta = \ln[(0.2/\beta a_0^2)(MkT)^{1/2}] + \gamma V^*/V_f \quad (13)$$

The free volume V_f of the liquid is given by

$$V_f = V\alpha(T - T_0) \quad (39)$$

where V is the specific volume. If we combine Eqs. (13) and (39), and ignore the weak temperature dependence due to the $T^{1/2}$ in the first term on the right hand side of Eq. (13), we obtain an expression of the form of the VTF Eq., (Eq.(35)).

Granted that molecular diameters, a_0 , of low molecular weight organic molecules are all of comparable magnitude, Eq. (13) shows that

Hence we will introduce the further simplification that all of the B_i terms in Eqs. (41) and (42) are the same, so that

$$\ln \eta = \sum X_i A_i + B/(T - \sum X_i T_{0i}) \quad (43)$$

and

$$\begin{aligned} 1/\eta &= [\sum X_i \exp(-A_i)] \exp[-B/(T - \sum X_i T_{0i})] \\ &= (\sum X_i a_i) \exp[-B/(T - \sum X_i T_{0i})] \end{aligned} \quad (44)$$

where $a_i = \exp(-A_i)$ in Eq. (44). We will call these, the master viscosity Eqs. They can be justified roughly as follows.

We have mentioned that the B parameters of the best fits to the VTF Eq. in Table III for the three binaries and for the mixtures of all C_{10} hydrocarbons are nearly the same. The mean values of B are:

581.26	for ND, NBC, CD, NBB, DMO and all their mixtures
536.26	for NBC-DMO mixtures
637.48	for CD-DMO mixtures
832.45	for HNN-XTHDCPD mixtures

Using these B values fits to the viscosities of the respective liquids could be obtained which were only slightly worse than the best fits of Table III. Parameter for VTF Eq. fits using the mean values of B are given in Table IX.

Inspection of these parameters shows that forcing a constant value of B orders the A and T_0 parameters, such that the A and T_0 values for the mixtures are intermediate between the respective A and T_0 parameters for the pure components. This means that the A and T_0 parameters for any of the liquids are given approximately by

$$A = \sum X_i A_i \quad (45)$$

$$T_o = \sum X_i T_{oi} \quad (46)$$

where subscript i refers as before to component i .

A_i and T_{oi} values were obtained by linear least squares fits to Eqs. (45) and (46) of the A and T_o values in Table IX. For the five C_{10} hydrocarbons these fits gave:

$A_{ND} = -7.701$	$T_{o\ ND} = 96.6$	
$A_{NBC} = -7.505$	$T_{o\ NBC} = 107.8$	
$A_{CD} = -7.003$	$T_{o\ CD} = 124.5$	(47a)
$A_{DMO} = -7.782$	$T_{o\ DMO} = 97.5$	
$A_{NBB} = -7.618$	$T_{o\ NBB} = 97.6$	
STD. DEV. = 0.030	STD. DEV. = 1.9	

For the HNN-XTHDCPD binary the fits to Eqs. (45) and (46) gave:

$A_{HNN} = -7.077$	$T_{o\ HNN} = 152.3$	
$A_{XTHDCPD} = -7.599$	$T_{o\ XTHDCPD} = 92.1$	(47b)
STD. DEV. = 0.015	STD. DEV. = 0.8	

Substituting Eqs. (45) and (46) into Eq. (35) justifies the semi-empirical Eq. (43) which accounts for the combined composition/temperature dependence of the viscosity.

2. Fits to Master Viscosity Equation (43)

We have carried out a fit to Eq. (43) of the experimental viscosity data for all temperatures and compositions of each of the three binary systems (NBC-DMO, CD-DMO and HNN-XTHDCPD) and of the combined data (1, 2, 3, 4, and 5 - component liquids) for all five C_{10}

hydrocarbons. Eq. (43) is linear in the A_i and B parameters, so that these were evaluated by a linear least squares computer fit. The T_{oi} parameters were set equal to those obtained in the previous section (Eq. (47)). The relative precision or % error for all viscosity determinations are about the same and independent of the magnitude of η , that is, $\Delta\eta/\eta \approx \Delta\ln \eta \approx \text{constant}$. Hence in fits to Eq. (43) all values of $\ln \eta$ were weighted equally. The parameters for the fits to Eq. (43) are given in Table X.

In Table XI a comparison is made of experimental η values with those calculated via Eq. (43) from the parameters of Table X. The quantity tabulated is $100(\ln \eta_{\text{calc}} - \ln \eta_{\text{exp}})$ which is approximately the % deviation of the experimental value from the best fit value. Clearly Eq. (43) does not fit the data to within experimental error.

Pure component viscosities are underestimated for the five C_{10} hydrocarbons and overestimated for HNN and XTHDCPD. Mixture viscosities are generally overestimated for five C_{10} 's and underestimated for HNN-XTHDCPD. The disagreement between experimental and calculated viscosities is largest at low temperatures and where the differences in pure component viscosities are largest. That is, the fit is better for the NBC-DMO binary, where the pure component viscosities are not very different, than for the CD-DMO and HNN-XTHDCPD binaries, where the opposite is true.

On the other hand Eq. (43) gives a remarkably good account of the data. For systems of N components it describes the viscosities of both pure liquids and mixtures as a function of temperature using $(N - 1)$ fewer adjustable parameters than are required by the VTF Eq. to account for η as a function of T of the pure liquids alone. For the HNN-XTHDCPD

binary system, for example, the experimental data cover a range in $\ln \eta$ of 8.61. The standard deviation in $\ln \eta$ from the fit to Eq. (43) is 0.056, i.e., only 0.7% of the range in $\ln \eta$. The error in $\ln \eta$ for the worst data point is 0.15, only 1.7% of the range in $\ln \eta$.

3. Fits to Master Viscosity Equation (44)

Fits of the experimental viscosity data, similar to those described in the previous section, were also carried out for Eq. (44). The T_{0j} values were again set equal to those of Eq. (47). Eq. (44) is not linear in B, so the best fit B parameter was obtained by trial and error. For each trial value of B the corresponding a_j values were obtained from a linear least squares fit. As mentioned in the previous section, the experimental error in each value of $\ln \eta$ is approximately the same. Consequently, the error in the fluidity, $\Delta(1/\eta)$, is not constant but is proportional to $1/\eta$. Hence in carrying out the fits to Eq. (44) each fluidity point was weighted by the fluidity itself. That is, the a_j and B parameters were chosen to minimize the following quantity:

$$\chi^2 = \sum_{j=1}^n (1/\eta_j - 1/\eta_{j,calc})^2 / (1/\eta_j)^2$$

where n is the number of data points included in the fit.

The parameters for the best fits to Eq. (44) are also given in Table X. In Table XII are tabulated values of $100(\ln \eta_{exp} - \ln \eta_{calc})$ for Eq. (44).

For the CD-DMO binary system Eq. (44) gives a markedly better fit to the data than does Eq. (43). For the other two binaries and for the combined data for all five C_{10} hydrocarbons there is not a great deal of difference between the goodness of the fits obtained from the two master viscosity equations.

4. Free Volume Interpretation of Viscosity

The free volume theory assumes that a molecule can undergo diffusive transport only when the free volume in its vicinity is equal to or greater than a certain critical value v^* . The shear viscosity is given by the Eq. (14):

$$\begin{aligned}\ln \eta &= \ln[(1/\beta a_0^2)(km/8\pi)^{1/2}] + (1/2)\ln T + \{\gamma v^*/[v\Delta\alpha(T - T_0)]\} \\ &= A' + (1/2)\ln T + B'/(T - T_2)\end{aligned}$$

If we equate this to the VTF Eq., $\ln \eta = A + B/(T - T_0)$ and also equate the temperature derivations of the two expressions

$$d\ln \eta/dT = -B/(T - T_2)^2 = 1/2T - B'/(T - T_2)^2$$

we get:

$$A = \ln[(1/\beta a_0^2)(km/8\pi)^{1/2}] + (1/2)[1 + \ln T - (T_2/T)] \quad (48)$$

$$B = \gamma v^*/v\Delta\alpha - (T - T_2)^2/2T \quad (49)$$

Over the temperature range covered by this study the second term on the right hand side of Eq. (48) is a very weak function of temperature, while the second term on the right hand side of Eq. (49) is quite small compared to our experimental B values. For example $1/2(1 + \ln T - T_2/T)$ for $T_2 \sim 100K$ changes from 2.98 to 3.23 between -50 and $50^\circ C$, compared to a typical value of A of around -7 (Table X). $(T - T_2)^2/2T$ for $T_2 \sim 100K$ is about 77 for $T = 50^\circ C$ and about 34 for $T = -50^\circ C$, compared to $B = 600$.

The molecular diameters a_0 calculated at $0^\circ C$ for five C_{10} hydrocarbons, HNN and XTHDCPD from Eq. (48), and the A_i parameters of Eq. (47) using 0.2 for the value of β are shown in Table XIII.

These are compared with and seen to be of the same magnitude as the mean molecular diameters calculated from the densities at 0°C, $(M/\rho_0 Na)^{1/3}$, where M is molecular weight and Na Avogadro's number.

These molecules are not spherical. To obtain alternative estimates of their dimensions we constructed scale frame work molecular models of them. The molecular model set is manufactured by Prentice - Hall, Inc., Englewood Cliffs, N.J. By examining the projections of each model on a background area when viewed from different aspects, we determined the dimensions (L, W, H) of a right parallopiped which would just enclose the entire molecule; these are also listed in Table XII. The CD, HNN and XTHDCPD molecules are rigid, but ND, NBC, DMO and NBB are flexible and passes a number of configurations. For ND the molecular dimensions were averaged over four different configurations expressed in terms of the staggered (s) or eclipsed (e) arrangement of groups bonded to adjacent carbons along the backbone of the molecule. These four ND configurations are (1) all (s), (2) (e) at carbons 5 - 6, (3) (e) at carbons 3 - 4 and 7 - 8 and (4) (e) at carbons 2 - 3, 5 - 6, and 8 - 9. For NBC the configuration is such that the n-butyl group is occupying an equatorial position on the cyclohexane ring and the terminal-CH₂CH₃ group and cyclohexane ring are in the trans positions relative to each other. For this general NBC configuration molecular dimensions were averaged for the (s) and (e) configurations of groups bonded to carbons 2 - 3 of the n-butyl side group. For DMO three different configurations have been considered, which are (1) all staggered, (2) eclipsed at carbons 2 - 3, 6 - 7, and (3) eclipsed at carbons 1 - 2, 7 - 8. Finally for NBB, two configurations were considered: (1) all staggered and (2) eclipsed at carbons 2 - 3 of n-butyl group.

The molecular diameters from viscosity a_i are remarkably close to realistic molecular diameters from the models.

They correlate best with the maximum molecular dimensions L obtained from the models as shown by the near constancy of the a_{oi}/L ratio.

As noted in Ref. 7, the constancy of the a_{oi}/L values implies, via the free volume theory, that the critical sized void into which a molecule makes a diffusive step must be of sufficient size to accomodate the longest dimension of the molecule. This suggests in turn that the molecules are freely rotating in the viscosity range studied here.

The differences in viscosities of the C_{10} molecules studied in this work and that of the HNN and XTHDCPD, are mostly due to differences both in the A_i and T_{oi} parameters. From Eqs. (43) and (44), what is not accounted for by A_i or a_i , is accounted for by T_{oi} . Of the five C_{10} liquids, CD and DMO differ most in viscosity, by a factor of 3.28 at the upper temperature limit of 50°C and by a factor of 10.79 at the lower limit of about -55°C . The differences in the A_i of CD and DMO (and hence the differences in L) account for a factor of about 2.0 difference in viscosity, i.e., for a substantial fraction of the viscosity difference in the experimental range. The difference between the A_i of HNN and of XTHDCPD accounts for a factor of about 1.6 difference in viscosities of these components.

In view of the simplicity of the free volume model for viscosity of liquids, we feel that this is a very good correlation between molecular structure and viscosity, and it appears then that the free volume model gives a surprisingly good account of the shear viscosity of these liquids and their mixtures in the low viscosity range.

Table I. Experimental viscosities and densities of n-Decane

Run 1		
<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.90	0.00611	0.7075
39.79	0.00694	0.7151
30.06	0.00790	0.7223
14.86	0.00994	0.7336
0.00	0.01291	0.7447
- 9.92	0.01577	0.7522
-21.02	0.02038	0.7605
-30.53	0.02610	0.7672
-32.00	(Frozen)--	--

Run 2		
<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.88	0.00613	0.7075
39.80	0.00703	0.7149
30.50	0.00788	0.7218
14.88	0.00998	0.7335
- 0.08	0.01294	0.7450
-10.24	0.01585	0.7518
-20.33	0.01996	0.7594
-31.31	0.02670	0.7676

Table I. Experimental viscosities and densities of n-Butylcyclohexane

Run 1		
<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.24	0.00829	0.7767
30.32	0.01094	0.7910
14.74	0.01424	0.8028
- 0.33	0.01910	0.8139
-15.15	0.02707	0.8246
-30.22	0.04185	0.8359
-40.13	0.05923	0.8430
-50.16	0.08982	0.8503
-56.61	0.12374	0.8553

Run 2		
<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.04	0.00832	0.7772
30.25	0.01102	0.7912
15.22	0.01414	0.8026
- 0.34	0.01910	0.8144
-15.19	0.02702	0.8251
-30.10	0.04161	0.8361
-40.22	0.05936	0.8436
-50.63	0.09213	0.8512
-58.51	0.13670	0.8570

Table I. Experimental viscosities and densities of Cis-Decahydro-naphthalene.

Run 1		
<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.96	0.01846	0.8734
30.05	0.02673	0.8885
12.98	0.03926	0.9017
- 0.09	0.05596	0.9118
-15.47	0.08969	0.9229
-30.31	0.15828	0.9343
-40.18	0.24844	0.9418
-50.25	0.42821	0.9494
-54.77	0.56464	0.9533
Run 2		
<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.53	0.01852	0.8726
30.28	0.02660	0.8865
- 0.77	0.05687	0.9100
-15.13	0.08849	0.9209
-32.68	0.17438	0.9339
-40.43	0.25020	0.9401
-50.57	0.43170	0.9482
-55.78	0.59974	0.9516

Table I. Experimental viscosities and densities of Dimethyloctane

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.03	0.00559	0.7012
39.97	0.00632	0.7085
30.14	0.00720	0.7160
14.98	0.00901	0.7273
- 0.29	0.01172	0.7386
-14.76	0.01567	0.7495
-30.09	0.02278	0.7607
-40.00	0.03051	0.7677
-50.13	0.04367	0.7752
-55.34	0.05396	0.7790

Table I. Experimental viscosities and densities of n-Butylbenzene

Run 1		
$T^{\circ} (C)$	η (P)	ρ (g/Cm ³)
49.92	0.00694	0.8360
28.94	0.00919	0.8525
- 0.14	0.01470	0.8753
-15.65	0.02044	0.8876
-32.38	0.03238	0.9010
-40.32	0.04202	0.9070
-50.31	0.06192	0.9148
-53.61	0.07173	0.9173

Run 2		
$T^{\circ} (C)$	η (P)	ρ (g/Cm ³)
49.90	0.00696	0.8366
40.14	0.00786	0.8441
30.12	0.00897	0.8518
15.11	0.01126	0.8635
- 0.08	0.01474	0.8751
-15.11	0.02031	0.8870
-29.95	0.03131	0.8984
-40.17	0.04332	0.9070
-50.18	0.06205	0.9144
-54.84	0.07652	0.9179

Table I. Experimental viscosities and densities of the mixture of
50% n-Decane + 50% n-Butylcyclohexane

Run 1		
$T^{\circ}(\text{C})$	η (P)	ρ (g/Cm ³)
49.92	0.00702	0.7414
39.84	0.00799	0.7485
30.26	0.00914	0.7557
15.39	0.01152	0.7666
- 0.59	0.01542	0.7782
-10.75	0.01913	0.7860
-20.81	0.02438	0.7937
-30.51	0.03190	0.8007
-40.20	0.04352	0.8072
-42.00	Frozen -	--
Run 2		
$T^{\circ}(\text{C})$	η (P)	ρ (g/Cm ³)
49.91	0.00705	0.7407
39.81	0.00799	0.7477
29.65	0.00920	0.7553
14.77	0.01163	0.7662
- 0.03	0.01524	0.7771
-10.52	0.01897	0.7849
-20.69	0.02453	0.7925
-30.41	0.03267	0.7999
-40.35	0.04474	0.8067

Table I. Experimental viscosities and densities of the mixture of
50% n-Decane + 50% cis-Decahydronaphthalene

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/cm³)</u>
49.86	0.00927	0.7829
39.77	0.01066	0.7901
29.96	0.01242	0.7974
14.40	0.01631	0.8090
- 0.27	0.02188	0.8198
-10.73	0.02788	0.8277
-20.32	0.03584	0.8346
-30.33	0.04852	0.8420
-40.19	0.06835	0.8488

Table I. Experimental viscosities and densities of the mixture of
50% n-Decane + 50% Dimethyloctane

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.10	0.00584	0.7048
39.97	0.00662	0.7121
30.03	0.00755	0.7197
15.06	0.00944	0.7307
- 0.26	0.01233	0.7424
-15.00	0.01666	0.7529
-30.11	0.02426	0.7638
-39.90	0.03263	0.7711
-50.04	0.04686	0.7783

Table I. Experimental viscosities and densities of the mixture of 50% n-Decane + 50% n-Butylbenzene.

<u>T°(C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.03	0.00622	0.7642
40.60	0.00695	0.7709
30.67	0.00792	0.7790
14.77	0.00998	0.7910
- 0.56	0.01293	0.8026
-15.07	0.01730	0.8135
-29.96	0.02481	0.8248
-40.10	0.03345	0.8322
-44.66	0.03899	0.8359

Table I. Experimental viscosities and densities of the mixture of
50% n-Butylcyclohexane + 50% cis-Decahydronaphthalene.

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.86	0.01162	0.8227
30.00	0.01593	0.8379
14.70	0.02129	0.8489
- 0.37	0.02974	0.8599
-14.73	0.04338	0.8708
-30.36	0.07177	0.8824
-41.77	0.10861	0.8902
-51.37	0.17644	0.8978
-55.18	0.21745	0.9007

Table I. Experimental viscosities and densities of the mixture
75% n-Butylcyclohexane + 25% Dimethyloctane

$T^{\circ}(\text{C})$	η (P)	ρ (g/Cm ³)
50.09	0.00739	0.7506
40.21	0.00848	0.7637
29.93	0.00982	0.7706
15.04	0.01248	0.7824
0.00	0.01652	0.7929
-15.10	0.02317	0.8042
-30.03	0.03501	0.8149
-39.97	0.04907	0.8223
-50.00	0.07336	0.8290
-53.63	0.08641	0.8317

Table I. Experimental viscosities and densities of the mixture of
50% n-Butylcyclohexane + 50% Dimethyloctane

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.21	0.00673	0.7379
40.16	0.00766	0.7452
30.16	0.00880	0.7523
15.26	0.01111	0.7641
- 0.09	0.01466	0.7748
-14.84	0.02014	0.7853
-29.97	0.03008	0.7963
-39.89	0.04132	0.8030
-50.00	0.06073	0.8102
-55.96	0.07950	0.8146

Table I. Experimental viscosities and densities of the mixture of
25% n-Butylcyclohexane + 75% Dimethyloctane

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.07	0.00613	0.7194
40.19	0.00694	0.7266
30.07	0.00794	0.7339
14.89	0.00994	0.7455
0.04	0.01295	0.7564
-15.22	0.01782	0.7675
-30.10	0.02603	0.7778
-40.01	0.03545	0.7857
-50.56	0.05232	0.7934
-54.65	0.06238	0.7960

Table I. Experimental viscosities and densities of the mixture of
50% n-Butylbenzene + 50% n-Butylcyclohexane

$T^{\circ}(\text{C})$	η (P)	ρ (g/cm ³)
49.98	0.00726	0.8040
30.06	0.00944	0.8196
14.84	0.01192	0.8307
- 0.41	0.01575	0.8430
-15.61	0.02175	0.8544
-30.69	0.03239	0.8658
-40.68	0.04443	0.8728
-50.55	0.06457	0.8806
-52.45	0.06999	0.8822

Table I. Experimental viscosities and densities of the mixture of
25% Dimethyloctane + 75% cis-Decahydronaphthalene

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.03	0.01239	0.8244
40.01	0.01450	0.8314
30.01	0.01722	0.8390
14.75	0.02311	0.8501
- 0.22	0.03254	0.8616
-15.13	0.04877	0.8725
-29.96	0.08003	0.8836
-40.21	0.12045	0.8912
-50.10	0.19201	0.8984
-54.38	0.24133	0.9013

Table I. Experimental viscosities and densities of the mixture of
50% cis-Decahydronaphthalene + 50% Dimethyloctane

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.04	0.00906	0.7796
40.25	0.01044	0.7867
30.10	0.01211	0.7939
14.82	0.01585	0.8050
- 0.10	0.02147	0.8161
-15.20	0.03097	0.8271
-30.15	0.04811	0.8380
-40.00	0.06854	0.8447
-50.39	0.10550	0.8523
-55.26	0.13530	0.8557

Table 1. Experimental viscosities and densities of mixtures of
75% Dimethyloctane + 25% cis-Decahydronaphthalene

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.11	0.00696	0.7386
40.12	0.00855	0.7460
29.87	0.00918	0.7536
15.28	0.01159	0.7644
- 0.38	0.01545	0.7759
-15.02	0.02133	0.7865
-30.16	0.03206	0.7974
-39.91	0.04405	0.8052
-50.18	0.06528	0.8122
-53.33	0.07481	0.8144

Table I. Experimental viscosities and densities of the mixture of
50% cis-Decahydronaphthalene + 50% n-Butylbenzene

$T^{\circ}(\text{C})$	η (P)	ρ (g/cm ³)
50.14	0.01000	0.8534
29.98	0.01309	0.8692
14.51	0.01699	0.8810
- 0.12	0.02268	0.8920
-14.87	0.03229	0.9037
-30.79	0.05118	0.9157
-40.25	0.07159	0.9226
-50.25	0.10889	0.9305
-53.72	0.12891	0.9334

Table I. Experimental viscosities and densities of the mixture of
50% n-Butylbenzene + 50% Dimethyloctane

$T(^{\circ}\text{C})$	η (P)	ρ (g/cm ³)
50.24	0.00597	0.7608
40.11	0.00673	0.7688
30.06	0.00766	0.7762
15.11	0.00949	0.7875
0.05	0.01222	0.7990
-14.94	0.01640	0.8106
-30.00	0.02360	0.8218
-40.04	0.03175	0.8296
-50.52	0.04572	0.8375
-54.68	0.05387	0.8409

Table I. Experimental viscosities and densities of the mixture of
33.3% n-Decane + 33.3% n-Butylcyclohexane + 33.3% cis-
Decahydronaphthalene

<u>T°(C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.91	0.00892	0.7811
30.26	0.01187	0.7951
15.24	0.01531	0.8063
- 0.23	0.02085	0.8177
-15.11	0.02963	0.8288
-30.65	0.04642	0.8402
-40.29	0.06503	0.8473
-50.41	0.09862	0.8545

Table 1. Experimental viscosities and densities of the mixture of
33.3% n-Butylbenzene + 33.3% n-Butylcyclohexane + 33.3%
n-Decane

$T^{\circ}(\text{C})$	η (P)	ρ (g/Cm ³)
49.72	0.00679	0.7686
39.75	0.00768	0.7763
30.11	0.00873	0.7831
14.79	0.01103	0.7951
- 0.17	0.01433	0.8064
-14.85	0.01941	0.8169
-29.93	0.02851	0.8285
-40.01	0.03885	0.8359
-50.52	0.05619	0.8438
-54.56	0.06653	0.8465

Table I. Experimental viscosities and densities of the mixture of
33.3% cis-Decahydronaphthalene + 33.3% n-Butylbenzene + 33.3%
n-Decane

$T^{\circ} (C)$	η (P)	ρ (g/Cm ³)
49.73	0.00806	0.7979
39.87	0.00918	0.8056
30.10	0.01054	0.8126
14.88	0.01339	0.8240
- 0.09	0.01771	0.8354
-15.35	0.02475	0.8466
-32.88	0.04021	0.8601
-40.31	0.05119	0.8651
-50.54	0.07564	0.8729

Table I. Experimental viscosities and densities of the mixture of
33.3% n-Butylbenzene + 33.3% n-Butylcyclohexane + 33.3% cis-
Decahydronaphthalene

T^0 (C)	η (P)	ρ (g/Cm ³)
50.04	0.00917	0.8265
30.19	0.01218	0.8414
14.87	0.01573	0.8526
0.04	0.02106	0.8644
-15.14	0.03001	0.8753
-30.13	0.04594	0.8870
-40.30	0.06538	0.8945
-49.97	0.09736	0.9018
-51.07	0.10242	0.9026
-54.94	0.12284	0.9051

Table I. Experimental viscosities and densities of the mixture of
25% n-Decane + 25% n-Butylcyclohexane + 25% cis-Decahydro-
naphthalene + 25% n-Butylbenzene

$T^{\circ}(\text{C})$	η (P)	ρ (g/Cm ³)
49.93	0.00809	0.7925
39.86	0.00926	0.8001
30.13	0.01062	0.8071
15.41	0.01345	0.8181
- 0.07	0.01791	0.8297
-14.92	0.02492	0.8406
-30.15	0.03765	0.8521
-40.03	0.05200	0.8594
-50.03	0.07645	0.8671
-54.87	0.09449	0.8710

Table 1. Experimental viscosities and densities of the mixture of 20% of 5 C₁₀ components

Run 1		
<u>T°(c)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.05	0.00747	0.7725
40.24	0.00848	0.7794
30.17	0.00975	0.7868
14.88	0.01242	0.7985
- 0.10	0.01635	0.8096
-14.91	0.02257	0.8206
-30.00	0.03373	0.8319
-39.89	0.04639	0.8390
-49.88	0.06786	0.8463
-55.66	0.08762	0.8504
Run 2		
<u>T°(C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
51.83	0.00731	0.7714
41.56	0.00833	0.7790
30.91	0.00965	0.7868
9.34	0.01369	0.8029
- 1.68	0.01686	0.8113
-12.68	0.02141	0.8197
-24.68	0.02897	0.8284
-37.12	0.04227	0.8372
-49.20	0.06588	0.8463
-54.44	0.08328	0.8503

Table I. Experimental viscosities and densities of HNN

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.55	0.10568	1.0690
39.74	0.14357	1.0760
29.85	0.20099	1.0831
14.94	0.36634	1.0941
0.04	0.76796	1.1048
-10.26	1.44592	1.1128
-15.12	2.02257	1.1169
-21.69	3.38706	1.1215
-29.79	7.02662	1.1274
-39.93	21.73532	1.1358
-50.50	96.66266	1.1443
-53.61	Frozen	

Table I. Experimental viscosities and densities of mixture of
80% HNN + 20% XTHDCPD

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.00	0.07283	1.0415
29.90	0.12759	1.0551
15.28	0.21149	1.0664
- 0.04	0.40261	1.0791
-14.94	0.88599	1.0903
-30.07	2.53050	1.1018
-40.09	5.77212	1.1094
-50.39	18.32347	1.1171
-54.92	33.46471	1.1207

Table I. Experimental viscosities and densities of mixture of
60% HNN + 40% XTHDCPD

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.85	0.05071	1.0132
30.09	0.08266	1.0281
15.10	0.12848	1.0396
- 0.04	0.22233	1.0509
-14.74	0.42766	1.0618
-30.08	0.98465	1.0730
-39.99	2.01006	1.0810
-50.52	5.00710	1.0890
-54.00	7.08425	1.0926

Table I. Experimental viscosities and densities of mixture of
40% HNN + 60% XTHDCPD

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.92	0.03525	0.9821
30.03	0.05451	0.9965
14.62	0.08201	1.0087
- 0.02	0.12821	1.0196
-15.00	0.22525	1.0313
-30.06	0.45584	1.0433
-41.40	0.87379	1.0522
-50.52	1.64076	1.0584
-53.73	2.10577	1.0607

Table I. Experimental viscosities and densities of mixture of
20% HNN + 80% XTIDCPD

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
50.00	0.02490	0.9492
30.15	0.03662	0.9640
14.88	0.05203	0.9759
0.00	0.07778	0.9870
-15.00	0.12451	0.9983
-30.00	0.22295	1.0099
-40.08	0.35547	1.0175
-50.00	0.61178	1.0251
-51.51	0.69513	1.0262

Table I. Experimental viscosities and densities of XTHDCPD

<u>T° (C)</u>	<u>η (P)</u>	<u>ρ (g/Cm³)</u>
49.99	0.01765	0.9126
30.05	0.02488	0.9273
15.38	0.03369	0.9392
0.00	0.04813	0.9508
-14.88	0.07401	0.9618
-30.07	0.11712	0.9735
-39.60	0.16979	0.9808
-50.20	0.27131	0.9892
-54.19	0.33199	0.9914

Table IIa. Parameters for density equations for ND, NBC, CD, DMO, NBB, and their mixtures

$$\rho \text{ (g/Cm}^3\text{)} = \rho(0) - bT(^{\circ}\text{C)}$$

<u>Molar Composition</u>	<u>T range($^{\circ}\text{C}$)</u>	<u>$\rho(0)$</u>	<u>$10^4 b$</u>	<u>Std. Dev. ρ</u>
ND (Run 1)	-31 to 50	0.7447	7.44	0.0001
(Run 2)	-31 to 50	0.7444	7.41	0.0002
NBC (Run 1)	-57 to 50	0.8135	7.35	0.0001
(Run 2)	-59 to 50	0.8139	7.37	0.0002
CD (Run 1)	-55 to 50	0.9114	7.59	0.0002
(Run 2)	-56 to 50	0.9096	7.54	0.0002
DMO	-56 to 50	0.7383	7.39	0.0002
NBB (Run 1)	-55 to 50	0.8752	7.77	0.0001
NBB (Run 2)	-54 to 50	0.8753	7.87	0.0001
0.5ND - 0.5NBC (Run 1)	-40 to 50	0.7780	7.36	0.0002
0.5ND - 0.5NBC (Run 2)	-40 to 50	0.7776	7.38	0.0001
0.5ND - 0.5CD	-40 to 50	0.8195	7.36	0.0002
0.5ND - 0.5DMO	-50 to 50	0.7417	7.35	0.0002
0.5ND - 0.5NBB	-45 to 50	0.8020	7.58	0.0002
0.5NBC - 0.5CD	-55 to 50	0.8598	7.39	0.0002
0.75NBC - 0.25DMO	-54 to 50	0.7922	7.53	0.0015
0.5NBC - 0.5DMO	-56 to 50	0.7743	7.23	0.0003
0.25NBC - 0.75DMO	-54 to 50	0.7562	7.35	0.0002
0.5NBC - 0.5NBB	-53 to 50	0.8423	7.60	0.0003
0.75CD - 0.25DMO	-54 to 50	0.8613	7.41	0.0002

0.5CD - 0.5DMO	-55 to 50	0.8158	7.24	0.0002
0.25CD - 0.75DMO	-53 to 50	0.7755	7.33	0.0002
0.5CD - 0.5NBB	-54 to 50	0.8920	7.67	0.0002
0.5DMO - 0.5NBB	-55 to 50	0.7991	7.61	0.0001
1/3ND-1/3NBC-1/3CD	-50 to 50	0.8176	7.35	0.0001
1/3ND-1/3NBC-1/3NBB	-55 to 50	0.8059	7.49	0.0002
1/3ND-1/3CD-1/3NBB	-51 to 50	0.8352	7.47	0.0002
1/3NBC-1/3CD-1/3NBB	-55 to 50	0.8641	7.52	0.0002
0.25ND - 0.25NBC - 0.25CD -0.25NBB	-55 to 50	0.8297	7.47	0.0002
0.2ND - 0.2NBC - 0.2CD -0.2DMO - 0.2NBB	-56 to 50	0.8094	7.41	0.0002

Table IIb. Parameters for density equations for HNN, XTHDCPD and their mixtures

$$\rho \text{ (g/Cm}^3\text{)} = \rho(0) - bT(^{\circ}\text{C)}$$

<u>Molar Composition</u>	<u>T range(°C)</u>	<u>$\rho(0)$</u>	<u>$10^4 b$</u>	<u>Std. Dev. ρ</u>
HNN	-50 to 50	1.1056	7.50	0.0005
0.8HNN - 0.2XTHDCPD	-55 to 50	1.0788	7.62	0.0005
0.6HNN - 0.4XTHDCPD	-54 to 50	1.05085	7.56	0.0004
0.4HNN - 0.6XTHDCPD	-54 to 50	1.0199	7.65	0.0004
0.2HNN - 0.8XTHDCPD	-51 to 50	0.9871	7.60	0.0001
XTHDCPD	-54 to 50	0.9506	7.61	0.0003

Table IIIa. Best fit VTF equation parameters for shear viscosity of ND, NBC, CD, DMO, NBB, and their mixtures

$$\ln \eta(P) = A + B/[T(K) - T_0]$$

Molar Composition	T range(°C)	A	B	T ₂	Std. Dev. ln η
ND (Run 1)	-31 to 50	-7.7764	614.11	94	0.001
(Run 2)	-31 to 50	-7.7442	605.03	95	0.002
NBC (Run 1)	-57 to 50	-7.3986	549.44	113	0.004
(Run 2)	-59 to 50	-7.3679	541.14	114	0.004
CD (Run 1)	-55 to 50	-7.6572	796.29	106	0.004
(Run 2)	-56 to 50	-7.6540	795.18	106	0.005
DMO	-56 to 50	-7.6158	534.10	104	0.005
NBB (Run 1)	-55 to 50	-7.2973	483.68	116	0.003
(Run 2)	-54 to 50	-7.1709	451.68	120	0.001
0.5ND - 0.5NBC (Run 1)	-40 to 50	-7.6013	584.93	102	0.002
0.5ND - 0.5NBC (Run 2)	-40 to 50	-7.6843	613.23	98	0.003
0.5ND - 0.5CD	-40 to 50	-7.6322	663.30	93	0.004
0.5ND - 0.5DMO	-50 to 50	-7.6243	547.89	103	0.004
0.5ND - 0.5NBB	-45 to 50	-7.5610	554.51	100	0.003
0.5NBC - 0.5CD	-55 to 50	-7.5108	658.00	108	0.003
0.75NBC - 0.25DMO	-54 to 50	-7.4807	550.85	110	0.005
0.5NBC - 0.5DMO	-56 to 50	-7.4730	529.26	110	0.005
0.25NBC - 0.75DMO	-55 to 50	-7.5039	517.64	109	0.004
0.5NBC - 0.5NBB	-53 to 50	-7.4491	549.18	106	0.004
0.75CD - 0.25DMO	-54 to 50	-7.5650	686.71	107	0.002

0.5CD - 0.5DMO	-55 to 50	-7.5067	604.51	108	0.003
0.25CD - 0.75DMO	-53 to 50	-7.5645	565.81	106	0.004
0.5CD - 0.5NBB	-54 to 50	-7.2042	543.55	114	0.003
0.5DMO - 0.5NBB	-55 to 50	-7.4655	511.02	106	0.004
1/3ND-1/3NBC-1/3CD	-50 to 50	-7.5518	621.52	104	0.003
1/3ND-1/3NBC-1/3NBB	-55 to 50	-7.5352	562.32	102	0.003
1/3ND-1/3CD-1/3NBB	-51 to 50	-7.4812	586.03	103	0.002
1/3NBC-1/3CD-1/3NBB	-55 to 50	-7.4028	584.63	108	0.004
0.25ND - 0.25NBC - 0.25CD - 0.25NBB	-55 to 50	-7.4598	577.65	105	0.003
0.2ND - 0.2NBC - 0.2CD -0.2DMO - 0.2NBB	-56 to 50	-7.4569	554.62	107	0.004

Table IIIb. Best fit VIF equation parameters for shear viscosity of HNN, XTHDCPD and their mixtures

$$\ln \eta(P) = A + B/[T(K) - T_0]$$

Molar Composition	T range(°C)	A	B	T ₀	Std. Dev. ln
HNN	-50 to 50	-7.1247	838.83	151	0.004
0.8HNN - 0.2XTHDCPD	-55 to 50	-7.1276	821.79	141	0.005
0.6HNN - 0.4XTHDCPD	-54 to 50	-7.1661	804.58	131	0.004
0.4HNN - 0.6XTHDCPD	-54 to 50	-7.3421	820.45	118	0.002
0.2HNN - 0.8XTHDCPD	-50 to 50	-7.5266	845.24	103	0.004
XTHDCPD	-54 to 50	-7.7096	864.80	88	0.006

Table IVa. Comparison of densities obtained in the present investigation with those reported in the literature.

ρ (g/cm ³)			
<u>Compound</u>	<u>T(°C)</u>	<u>Present Results</u>	<u>Literature</u>
ND	0	0.744r, 0.7444	0.74487 ^a , 0.17448 ^b
	20	0.7298, 0.7296	0.72996 ^a , 0.7298 ^b , 0.73014 ^c , 0.72994 ^d , 0.72987 ^e
	30	0.7224, 0.7222	0.72238 ^a
NBC	20	0.7988, 0.7992	0.79918 ^f
	30	0.7915, 0.7918	0.79176 ^f
CD	0	0.9114	0.9120 ^g
	20	0.8962	0.8967 ^g , 0.89671 ^h
	30	0.8886	0.8892 ^g , 0.88911 ^h

^aBruylants (unp.). Timmermans.

^bChavanne and Tock (1932). Timmermans.

^cShepard, Henne and Midgley (1931). Timmermans.

^dBruun and Hicks - Brunn (1932). Timmermans.

^eMears, Fooksonetal (1950). Timm. Volume II.

^fForziati and Rossini (1949). Timm. Volume II.

^gSeyer and Davenport (1941). Timmermans.

^hCamín and Rossini (1955). Timm. Volume II.

DMO	0	0.7383	0.73785 ⁱ
	15	0.7272	0.72640 ⁱ
	30	0.7161	0.71494 ⁱ
NBB	-45.2	0.9103, 0.9103	0.91210 ^j
	-22.9	0.8930, 0.8933	0.89440 ^j
	0	0.8752, 0.8753	0.87608 ^j , 0.87609 ^m
	20	0.8597, 0.8596	0.86013 ^f , 0.86013 ^k , 0.8603 ^l
	40	0.8442, 0.8438	0.84389 ^k

ⁱTimmermans and Hennaut - Roland (1929). Timmermans.

^jMassart (1936). Timmermans

^kDonaldson and Quayle (1950). Timm. Volume II.

^lBirch, Deanetal (1949).

^mB. E. (unp.). Timmermans.

Table IVb. Comparison of viscosities obtained in present investigation with those reported in the literature.

Compound	T(°C)	η (CP)	
		Present Results	Literature
ND	25	0.850, 0.851	0.8527 ^a
NBC	-55	11.38, 11.39	11.7 ^b
	-25	3.57, 3.56	3.58 ^b
	0	1.891, 1.892	1.90 ^b
	25	1.189, 1.189	1.205 ^b
	50	8.360, 8.356	8.561 ^b
CD	0	5.54, 5.52	5.620 ^c
	10	4.23, 4.22	4.300 ^c
	20	3.33, 3.32	3.381 ^c
	30	2.683, 2.677	2.723 ^c
DMO	15	0.896	0.889 ^d
	30	0.720	0.703 ^d

^aShepard, Henne and Midgley (1931). Timmermans.

^bCarpenter Davies and Matheson, J. Chem. Phys., 46, 2451 (1966).

^cSeyer and Leslie (1942). Timmermans.

^dTimmermans and Hennaut - Roland (1929). Timmermans.

NBB	-50.7	6.37, 6.32	6.39 ^e
	-30.5	3.09, 3.06	3.12 ^e
	-10.3	1.825, 1.815	1.82 ^e
	9.8	1.228, 1.229	1.23 ^e
	30.0	0.898, 0.905	0.897 ^e

^eBarlow, Lamb and Matheson, Proc. Roy. Soc., A292, 322 (1966)

Table Va. Test of isothermal additivity rules for shear viscosities of mixtures of C_{10} hydrocarbons. η_{ex} is the experimental mixture of viscosity.

Molar Composition	$(\sum X_i/\eta_i)^{-1}/\eta_{ex}$		$\exp(\sum X_i \ln \eta_i)/\eta_{ex}$	
	50°C	-30°C	50°C	-30°C
0.5ND - 0.5NBC	1.00	1.01	1.02	1.04
0.5ND - 0.5CD	1.00	0.95	1.15	1.36
0.5ND - 0.5DMO	1.00	0.99	1.00	1.00
0.5ND - 0.5NBB	1.05	1.11	1.05	1.12
0.5NBC - 0.5CD	0.99	0.93	1.07	1.14
0.75NBC - 0.25DMO	0.99	0.98	1.01	1.02
0.5NBC - 0.5DMO	0.99	0.98	1.01	1.02
0.25NBC - 0.75DMO	0.99	0.99	1.01	1.02
0.5NBC - 0.5NBB	1.04	1.10	1.05	1.12
0.75CD - 0.25DMO	0.95	0.79	1.11	1.21
0.5CD - 0.5DMO	0.95	0.83	1.12	1.25
0.25CD - 0.75DMO	0.97	0.91	1.08	1.15
0.5CD - 0.5NBB	1.02	1.02	1.14	1.38
0.5DMO - 0.5NBB	1.04	1.10	1.04	1.11
1/3ND-1/3NBC-1/3CD	0.99	0.95	1.10	1.21
1/3ND-1/3NBC-1/3NBB	1.04	1.09	1.05	1.11
1/3ND-1/3CD-1/3NBB	1.04	1.04	1.15	1.34
1/3NBC-1/3CD-1/3NBB	1.03	1.04	1.12	1.26
0.25ND - 0.25NBC - 0.25CD - 0.25NBB	1.03	1.04	1.11	1.26
0.2ND - 0.2NBC - 0.2CD - 0.2DMO - 0.2NBB	1.02	1.01	1.10	1.21

Table Vb. Tests of isothermal additivity rules for shear viscosities of mixtures of HNN and XTHDCPD. η_{ex} is the experimental mixture viscosity.

<u>Molar Composition</u>	$(\sum X_i/\eta_i)^{-1}/\eta_{ex}$		$\exp(\sum X_i \ln \eta_i)/\eta_{ex}$	
	<u>50°C</u>	<u>-30°C</u>	<u>50°C</u>	<u>-30°C</u>
0.8HNN - 0.2XTHDCPD	0.72	0.22	1.01	1.27
0.6HNN - 0.4XTHDCPD	0.69	0.29	1.01	1.38
0.4HNN - 0.6XTHDCPD	0.75	0.87	1.02	1.34
0.2HNN - 0.8XTHDCPD	0.85	0.66	1.01	1.20

Table VI. Test of the Katti - Chaudhri model for the NBC-DMO binary system

$T/^{\circ}\text{C}$	Mole Fraction	-W/R from η_V	-W/R from η
50 $^{\circ}\text{C}$	0.25NBC - 0.75DMO	11 $\pm$ 7	11 $\pm$ 7
	0.50NBC - 0.50DMO	11 $\pm$ 7	11 $\pm$ 7
	0.75NBC - 0.25DMO	18 $\pm$ 7	24 $\pm$ 7
0 $^{\circ}\text{C}$	0.25NBC - 0.75DMO	21 $\pm$ 7	21 $\pm$ 7
	0.50NBC - 0.50DMO	16 $\pm$ 7	17 $\pm$ 7
	0.75NBC - 0.25DMO	17 $\pm$ 7	20 $\pm$ 7
-50 $^{\circ}\text{C}$	0.25NBC - 0.75DMO	19 $\pm$ 7	20 $\pm$ 7
	0.50NBC - 0.50DMO	20 $\pm$ 7	21 $\pm$ 7
	0.75NBC - 0.25DMO	24 $\pm$ 7	25 $\pm$ 7

Table VII. Test of the Katti - Chaudhri model for the CD-DMO binary system

<u>T°C</u>	<u>Mole Fraction</u>	<u>-W/R from η_V</u>	<u>-W/R from η</u>
50°C	0.25CD - 0.75DMO	128 $\pm$ 6	133 $\pm$ 6
	0.50CD - 0.50DMO	140 $\pm$ 6	145 $\pm$ 6
	0.75CD - 0.25DMO	168 $\pm$ 6	173 $\pm$ 6
0°C	0.25CD - 0.75DMO	159 $\pm$ 6	164 $\pm$ 6
	0.50CD - 0.50DMO	180 $\pm$ 6	186 $\pm$ 6
	0.75CD - 0.25DMO	209 $\pm$ 6	215 $\pm$ 6
-50°C	0.25CD - 0.75DMO	197 $\pm$ 6	202 $\pm$ 6
	0.50CD - 0.50DMO	226 $\pm$ 6	232 $\pm$ 6
	0.75CD - 0.25DMO	261 $\pm$ 6	267 $\pm$ 6

Table VIII. Test of the Katti - Chaudhri model for the HNN-XTHDCPD binary system

$T, ^\circ\text{C}$	Mole Fraction	-W/R from nV	-W/R from n
50 $^\circ\text{C}$	0.20HNN - 0.80XTHDCPD	22 $\pm$ 5	23 $\pm$ 5
	0.40HNN - 0.60XTHDCPD	28 $\pm$ 5	31 $\pm$ 5
	0.60HNN - 0.40XTHDCPD	18 $\pm$ 5	20 $\pm$ 5
	0.80HNN - 0.20XTHDCPD	12 $\pm$ 5	16 $\pm$ 5
0 $^\circ\text{C}$	0.20HNN - 0.80XTHDCPD	129 $\pm$ 5	130 $\pm$ 5
	0.40HNN - 0.60XTHDCPD	143 $\pm$ 5	145 $\pm$ 5
	0.60HNN - 0.40XTHDCPD	152 $\pm$ 5	155 $\pm$ 5
	0.80HNN - 0.20XTHDCPD	159 $\pm$ 5	162 $\pm$ 5
-50 $^\circ\text{C}$	0.20HNN - 0.80XTHDCPD	476 $\pm$ 5	478 $\pm$ 5
	0.40HNN - 0.60XTHDCPD	512 $\pm$ 5	514 $\pm$ 5
	0.60HNN - 0.40XTHDCPD	566 $\pm$ 5	568 $\pm$ 5
	0.80HNN - 0.20XTHDCPD	643 $\pm$ 5	645 $\pm$ 5

Table IXa. VTF Eq. parameters for shear viscosity of ND, NBC, CD, NBB, DMO and their mixtures for constant B parameter.

$$\ln \eta(P) = A + B/[T(K) - T_0]$$

<u>Molar Composition</u>	<u>T range(°C)</u>	<u>A</u>	<u>B</u>	<u>T₀</u>	<u>Std. Dev. ln η</u>
ND	-31 to 50	-7.6864	591.26	98.9	0.002
NBC	-57 to 50	-7.5057	"	109.0	0.006
CD	-55 to 50	-6.9070	"	127.2	0.033
DMO	-56 to 50	-7.7613	"	97.5	0.007
NBB	-55 to 50	-7.6269	"	102.7	0.014
0.5ND - 0.5NBC	-40 to 50	-7.5905	"	102.5	0.002
0.5ND - 0.5CD	-40 to 50	-7.3976	"	108.1	0.007
0.5ND - 0.5DMO	-50 to 50	-7.7245	"	98.3	0.005
0.5ND - 0.5NBB	-45 to 50	-7.6391	"	96.1	0.003
0.5NBC - 0.5CD	-55 to 50	-7.2497	"	116.6	0.012
0.75NBC - 0.25DMO	-54 to 50	-7.5786	"	106.0	0.007
0.5NBC - 0.5DMO	-56 to 50	-7.6397	"	103.1	0.008
0.25NBC - 0.75DMO	-55 to 50	-7.7048	"	100.3	0.008
0.5NBC - 0.5NBB	-53 to 50	-7.5507	"	101.7	0.005
0.75CD - 0.25DMO	-54 to 50	-7.2126	"	118.8	0.014
0.5CD - 0.5DMO	-55 to 50	-7.4307	"	110.8	0.006
0.25CD - 0.75DMO	-53 to 50	-7.6139	"	104.0	0.004
0.5CD - 0.5NBB	-54 to 50	-7.3304	"	109.2	0.006
0.5NBB - 0.5DMO	-55 to 50	-7.6821	"	96.1	0.008
1/3ND-1/3NBC-1/3CD	-50 to 50	-7.4243	"	109.1	0.005

1/3ND-1/3NBC-1/3NBB	-55 to 50	-7.5935	581.26	99.4	0.004
1/3ND-1/3CD-1/3NBB	-51 to 50	-7.4667	"	103.6	0.002
1/3NBC-1/3CD-1/3NBB	-55 to 50	-7.3916	"	108.4	0.004
0.25ND - 0.25NBC - 0.25CD - 0.25NBB	-55 to 50	-7.4712	"	104.5	0.003
0.2ND - 0.2NBC - 0.2 CD - 0.2NBB - 0.2DMO	-56 to 50	-7.5415	"	103.5	0.005

Table IXb. VTF Eq. parameters for shear viscosity of NBC-DMO binary system for constant B parameter

$$\ln \eta (P) = A + B/[T(K) - T_0]$$

<u>Molar Composition</u>	<u>T range(°C)</u>	<u>A</u>	<u>B</u>	<u>T₀</u>	<u>Std. Dev. ln η</u>
NBC	-57 to 50	-7.3533	536.26	114.7	0.005
0.75NBC - 0.25DMO	-54 to 50	-7.4328	"	111.9	0.005
0.5NBC - 0.5DMO	-56 to 50	-7.4958	"	109.1	0.005
0.25NBC - 0.75DMO	-55 to 50	-7.5639	"	106.4	0.004
DMO	-56 to 50	-7.6226	"	103.7	0.005

Table IXc. VTF Eq. parameters for shear viscosity of CD-DMO binary system for constant B parameter.

$$\ln \eta(P) = A + B/[T(K) - T_0]$$

<u>Molar Composition</u>	<u>T range(°C)</u>	<u>A</u>	<u>B</u>	<u>T₀</u>	<u>Std. Dev. ln η</u>
CD	-55 to 50	-7.1142	637.48	121.4	0.023
0.75CD - 0.25DMO	-54 to 50	-7.4041	"	112.4	0.006
0.5CD - 0.5DMO	-55 to 50	-7.6120	"	104.1	0.006
0.25CD - 0.75DMO	-53 to 50	-7.7881	"	96.9	0.008
DMO	-56 to 50	-7.9271	"	90.0	0.011

Table IXd. VTF Eq. parameters for shear viscosity of HNN-XTHDCPD binary system for constant B parameter.

$$\ln \eta (P) = A + B/[T(K) - T_0]$$

<u>Molar Composition</u>	<u>T range(°C)</u>	<u>A</u>	<u>B</u>	<u>T₀</u>	<u>Std. Dev. $\ln \eta$</u>
HNN	-50 to 50	-7.0959	832.45	151.4	0.006
0.8HNN - 0.2XTHDCPD	-55 to 50	-7.1727	"	140.3	0.006
0.6HNN - 0.4XTHDCPD	-54 to 50	-7.2734	"	128.9	0.006
0.4HNN - 0.6XTHDCPD	-54 to 50	-7.3838	"	117.0	0.003
0.2HNN - 0.8XTHDCPD	-50 to 50	-7.4869	"	104.2	0.004
XTHDCPD	-54 to 50	-7.6161	"	91.2	0.006

Table X. Results of fits to master viscosity Eqs. (43) and (44).

Units of A_i , a_i Bare so chosen to shear viscosity in units of poise.

Mixture	com- ponent	$T_r(K)$	A_i values from fit to viscosity Eq. (43)	Std. Dev. of $\ln \eta$ from Eq. (43)	a_i values from fit to fluidity Eq. (44)	average frac- tional Std. Dev. from Eq. (44)
C ₁₀ Mixtures	ND	96.63	-7.715		2255.02	
	NBC	107.83	-7.506		1841.37	
	CD	124.50	-6.990	0.066	982.36	0.050
	NBB	97.60	-7.638		2086.22	
	DMO	97.53	-7.786		2412.83	
		B =	581.75	B =	586.	
NBC-DMO	NBC	114.65	-7.359		1562.81	
	DMO	103.65	-7.626	0.007	2043.95	0.005
		B =	538.91	B =	536.	
CD-DMO	CD	120.63	-7.184		1218.74	
	DMO	89.26	-7.985	0.064	2807.66	0.029
		B =	639.57	B =	637.	
HNN- XTHDCPD	HNN	152.27	-7.081		1224.54	
	XTH DCPD	92.06	-7.572	0.056	1932.18	0.063
		B =	830.34		834.5	

Table XI. Comparison between the experimental viscosity, η_{exp} , and viscosity, η_{calc} , obtained from Eq. (43) using the parameters of Table X.

100 ($\ln \eta_{\text{exp}} - \ln \eta_{\text{calc}}$)			
C_{10} Mixtures	50°C	0°C	-50°C
ND	5.0	7.1	9.6
NBC	2.0	1.9	5.1
CD	7.1	18.3	23.3
DMO	2.9	1.5	2.2
NBB	11.1	10.2	21.2
0.5ND - 0.5NBC	2.1	2.3	2.6
0.5ND - 0.5CD	- 6.4	- 5.3	-10.8
0.5ND - 0.5DMO	4.2	4.2	7.2
0.5ND - 0.5NBB	2.6	1.3	0.2
0.5NBC - 0.5CD	- 1.5	1.6	1.4
0.75NBC - 0.25DMO	- 2.2	- 2.1	- 3.1
0.5NBC - 0.5DMO	1.7	0.4	2.2
0.25NBC - 0.75DMO	- 1.4	- 2.3	- 2.6
0.5NBC - 0.5NBB	1.3	- 0.5	- 2.0
0.75CD - 0.25DMO	- 4.4	- 3.6	- 6.4
0.5CD - 0.5DMO	- 5.1	- 4.6	- 5.7
0.25CD - 0.75DMO	- 0.3	- 2.8	- 2.4
0.5CD - 0.5NBB	- 3.4	- 6.4	-10.0
0.5DMO - 0.5NBB	1.1	- 0.9	- 2.4

1/3 ND-1/3 NBC-1/3 CD	- 3.7	- 3.2	- 5.7
1/3 ND-1/3 NBC-1/3 NBB	1.2	- 0.3	- 2.4
1/3 ND-1/3 CD-1/3 NBB	- 5.4	- 7.5	-13.2
1/3 NBC-1/3 CD-1/3 NBB	- 3.7	- 5.0	- 8.8
0.25ND - 0.25NBC - 0.25CD - 0.25NBB	- 3.7	- 5.6	-10.2
0.2ND - 0.2NBC - 0.2CD - 0.2DMO - 0.2NBB	- 2.8	- 4.8	- 7.1

<u>NBC-DMO</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
NBC	- 1.0	- 0.9	- 1.8
0.75NBC - 0.25DMO	- 2.2	- 2.1	- 3.1
0.5NBC - 0.5DMO	- 1.6	- 2.3	- 3.0
0.25NBC - 0.75DMO	- 1.4	- 2.3	- 2.6
DMO	- 0.8	- 1.2	- 1.7

<u>CD-DMO</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
CD	3.6	9.7	8.6
0.75CD - 0.25DMO	- 4.4	- 3.6	- 6.4
0.5CD - 0.5DMO	- 4.4	- 6.4	- 8.3
0.25CD - 0.75DMO	- 0.3	- 2.8	- 2.4
DMO	7.2	4.9	7.5

<u>HNN-XTHDCPD</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
HNN	- 3.0	- 4.6	-13.3
0.8HNN - 0.2XTHDCPD	2.4	2.3	4.1
0.6HNN - 0.4XTHDCPD	4.0	4.3	9.8
0.4HNN - 0.6XTHDCPD	2.2	3.3	7.6
0.2HNN - 0.8XTHDCPD	- 0.1	0.0	2.5
XTHDCPD	- 5.3	- 5.2	- 7.3

Table XII. Comparison between the experimental viscosity, η_{exp} , and viscosity, η_{calc} , calculated via Eq. (44) using the parameters of Table X.

100 ($\ln \eta_{\text{exp}} - \ln \eta_{\text{calc}}$)			
<u>C₁₀ Mixtures</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
ND	3.8	5.3	6.8
NBC	1.3	0.6	2.6
CD	- 5.0	5.4	9.0
DMO	1.3	- 0.6	- 1.0
NBB	9.8	8.3	18.3
0.5ND - 0.5NBC	1.6	1.3	0.5
0.5ND - 0.5CD	- 4.8	- 4.2	-10.9
0.5ND - 0.5DMO	2.8	2.3	4.3
0.5ND - 0.5NBB	1.4	- 0.5	- 2.5
0.5NBC - 0.5CD	- 3.1	- 0.8	- 2.1
0.75NBC - 0.25DMO	- 0.5	0.0	- 0.2
0.5NBC - 0.5DMO	1.5	- 0.4	0.3
0.25NBC - 0.75DMO	0.2	- 0.3	0.2
0.5NBC - 0.5NBB	0.4	- 1.9	- 4.4
0.75CD - 0.25DMO	- 2.8	- 1.6	- 3.7
0.5CD - 0.5DMO	- 2.2	- 2.4	- 4.6
0.25CD - 0.75DMO	1.1	- 1.0	- 0.1
0.5CD - 0.5NBB	- 3.2	- 6.8	-11.6
0.5DMO - 0.5NBB	0.9	- 2.7	- 5.1

1/3 ND-1/3 NBC-1/3 CD	- 2.6	- 2.7	- 6.4
1/3 ND-1/3 NBC-1/3 NBB	0.4	- 1.7	- 4.8
1/3 ND-1/3 CD-1/3 NBB	- 4.0	- 6.7	-13.5
1/3 NBC-1/3 CD-1/3 NBB	- 3.4	- 5.4	-10.3
0.25ND - 0.25NBC - 0.25CD - 0.25NBB	- 2.8	- 5.3	-11.0
0.2ND - 0.2NBC - 0.2CD - 0.2DMO - 0.2NBB	- 1.7	- 4.2	- 7.6

<u>NBC-DMO</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
NBC	- 0.1	0.5	0.4
0.75NBC - 0.25DMO	- 0.5	0.0	- 0.2
0.5NBC - 0.5DMO	0.3	0.0	0.0
0.25NBC - 0.75DMO	0.2	- 0.3	0.2
DMO	0.2	0.2	0.4

<u>CD-DMO</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
CD	- 3.0	3.6	3.2
0.75CD - 0.25DMO	- 2.8	- 1.6	- 3.7
0.5CD - 0.5DMO	- 0.9	- 2.6	- 3.8
0.25CD - 0.75DMO	1.1	- 1.0	- 0.1
DMO	3.8	1.8	4.9

<u>HNN-XTHDCPD</u>	<u>50°C</u>	<u>0°C</u>	<u>-50°C</u>
HNN	- 2.5	- 5.1	-16.1
0.8HNN - 0.2XTHDCPD	4.2	3.3	3.2
0.6HNN - 0.4XTHDCPD	5.9	5.6	9.6
0.4HNN - 0.6XTHDCPD	3.4	3.9	7.0
0.2HNN - 0.8XTHDCPD	-22.5	- 0.8	-78.2
XTHDCPD	- 7.7	- 8.1	-11.0

Table XIII. Comparison of molecular diameters of C_{10} hydrocarbons calculated from viscosity with those estimated from density and molecular models

Molecule	a_{0j} ($\text{\AA}^0$)	$(M/\rho_0 Na)^{1/3}$ ($\text{\AA}^0$)	L, W, H ($\text{\AA}^0$) from models	a_{0j}/L
ND	9.5	6.8	12.8, 5.9, 5.6	0.75
NBC	8.5	6.6	11.9, 6.3, 5.1	0.71
CD	6.5	6.3	9.8, 7.5, 5.9	0.67
DMO	9.9	6.8	13.4, 5.9, 5.1	0.74
NBB	9.0	6.3	12.1, 7.0, 5.3	0.74
HNN	7.1	6.5	9.3, 6.8, 6.8	0.76
XTHDCPD	9.0	6.2	8.4, 6.9, 6.3	1.07

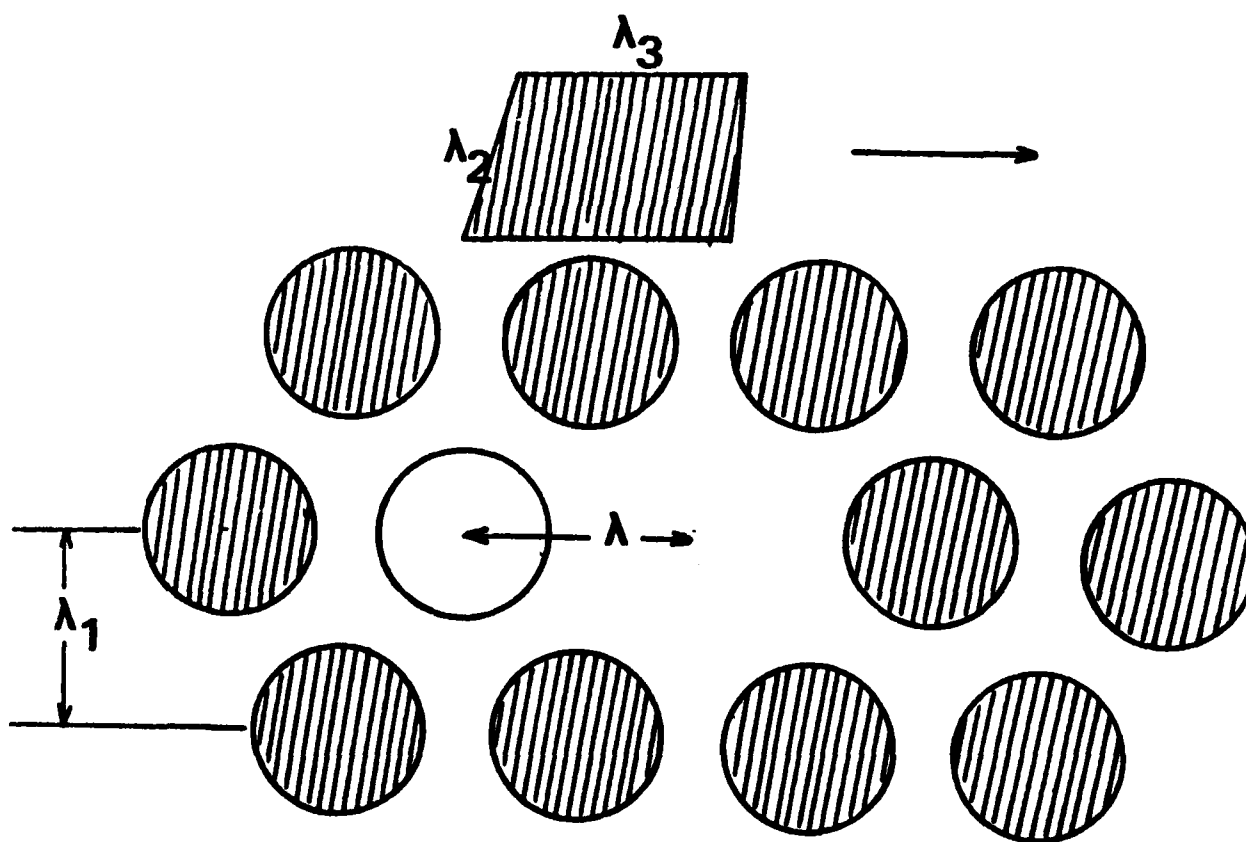


Fig.1. Transition State Model of Molecular Transport in a Liquid

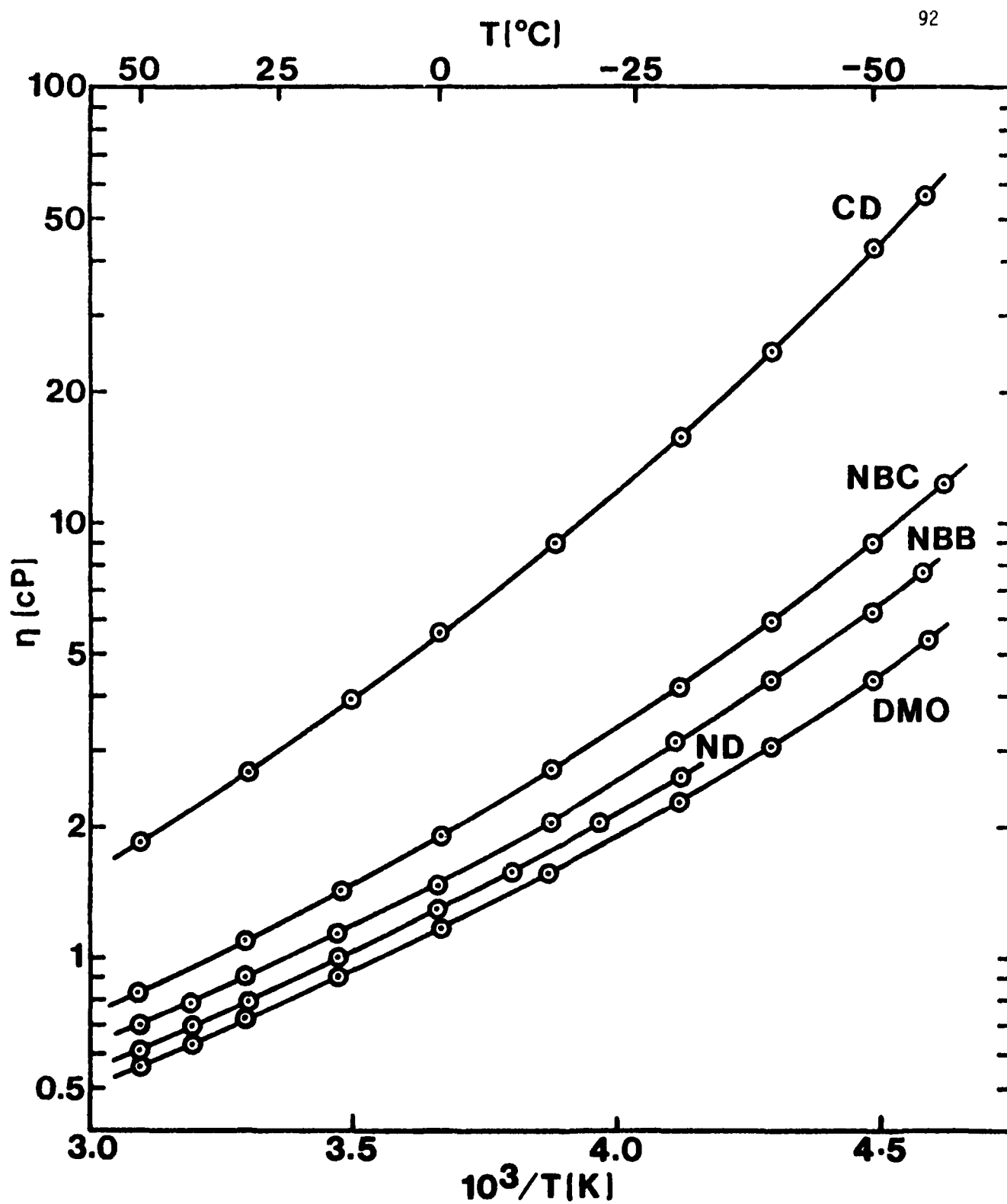


Fig.2

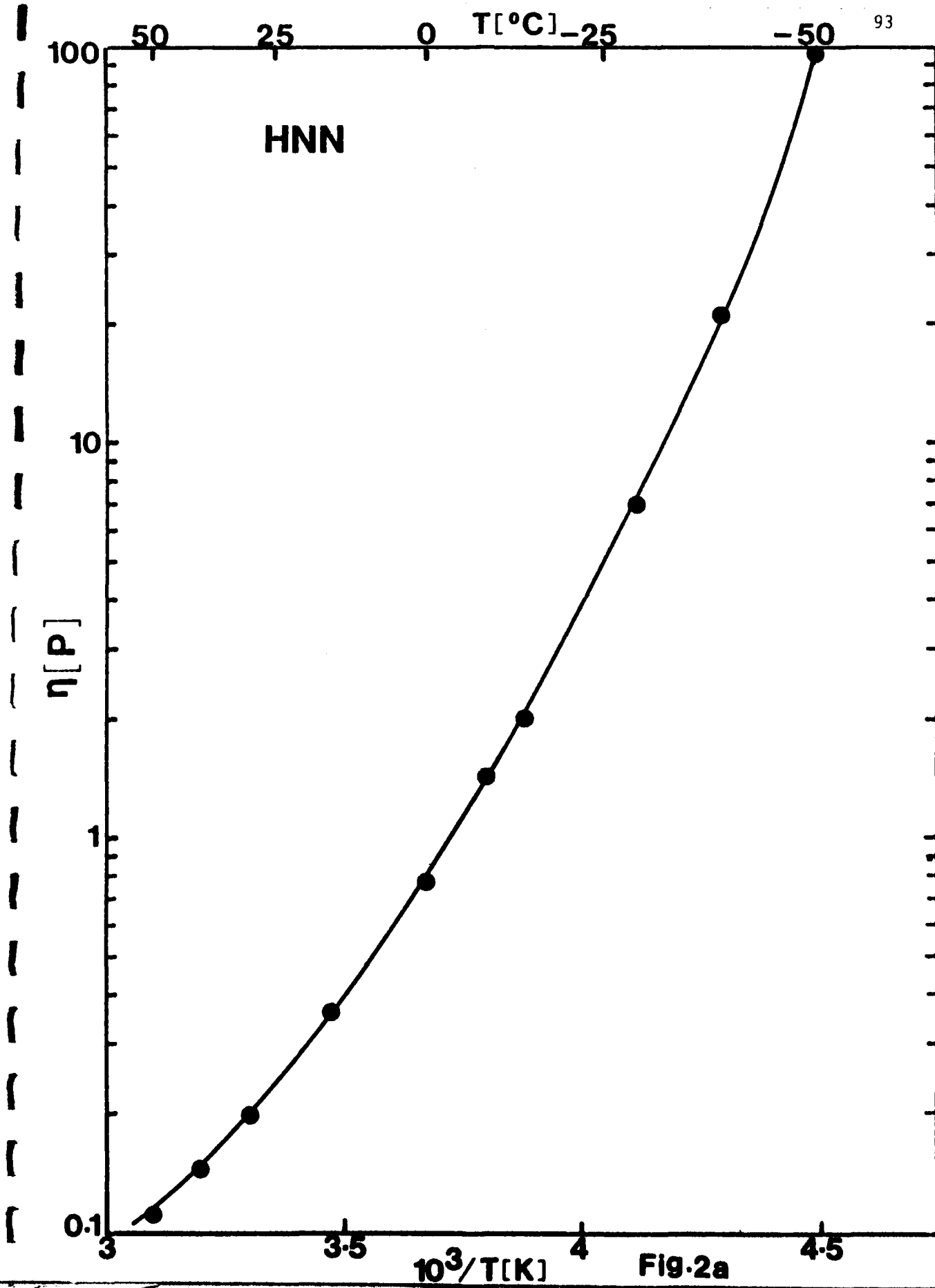


Fig. 2a

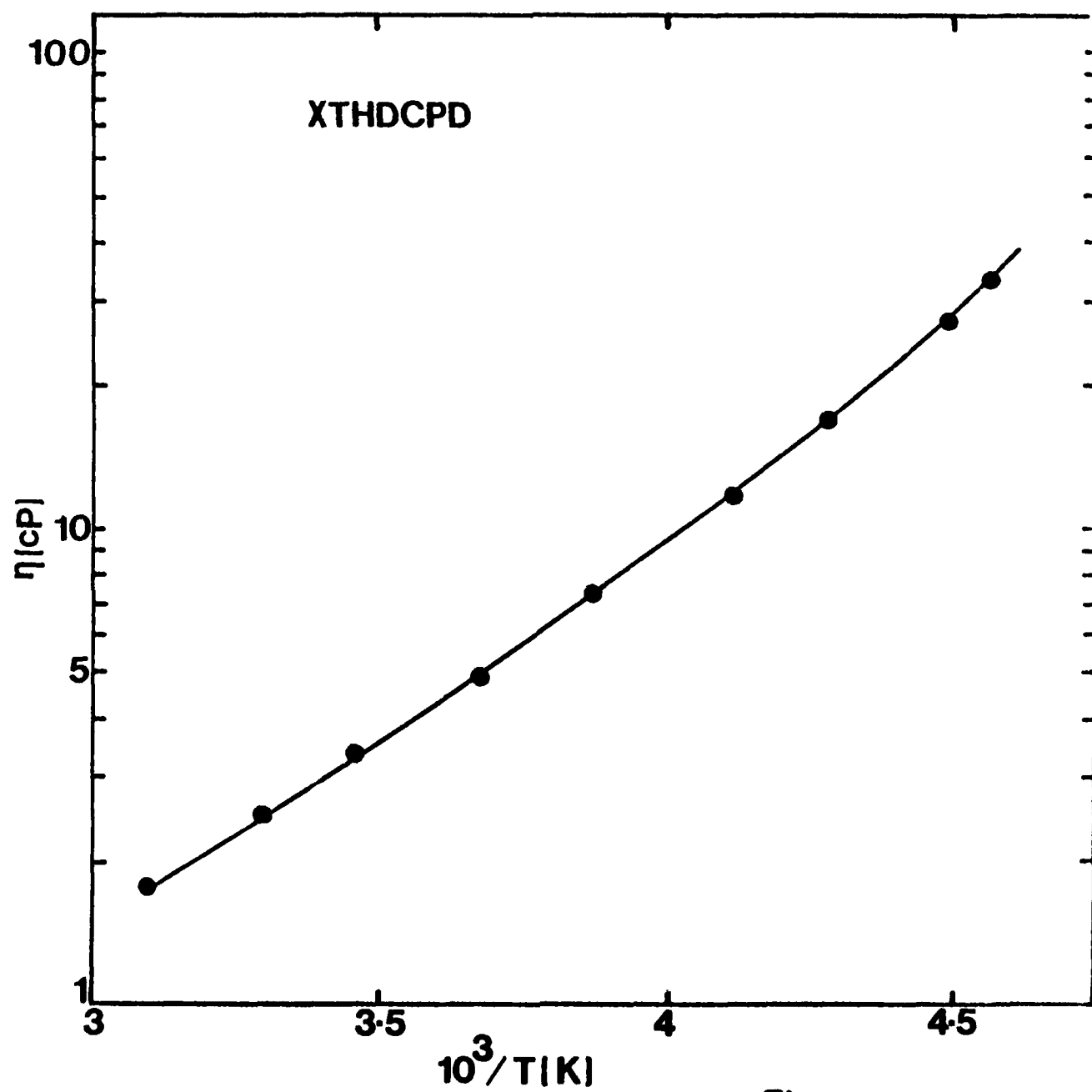


Fig.2b

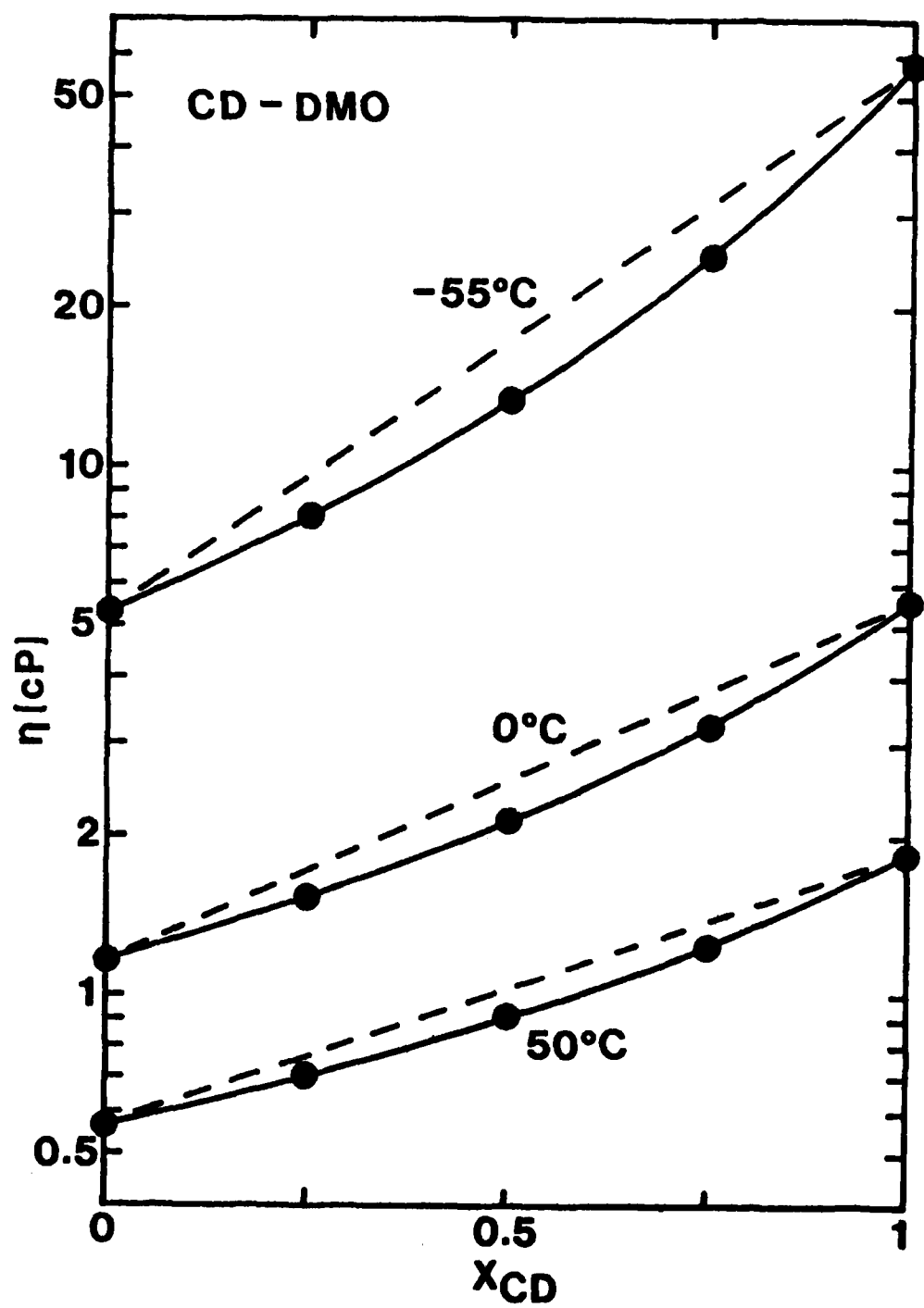
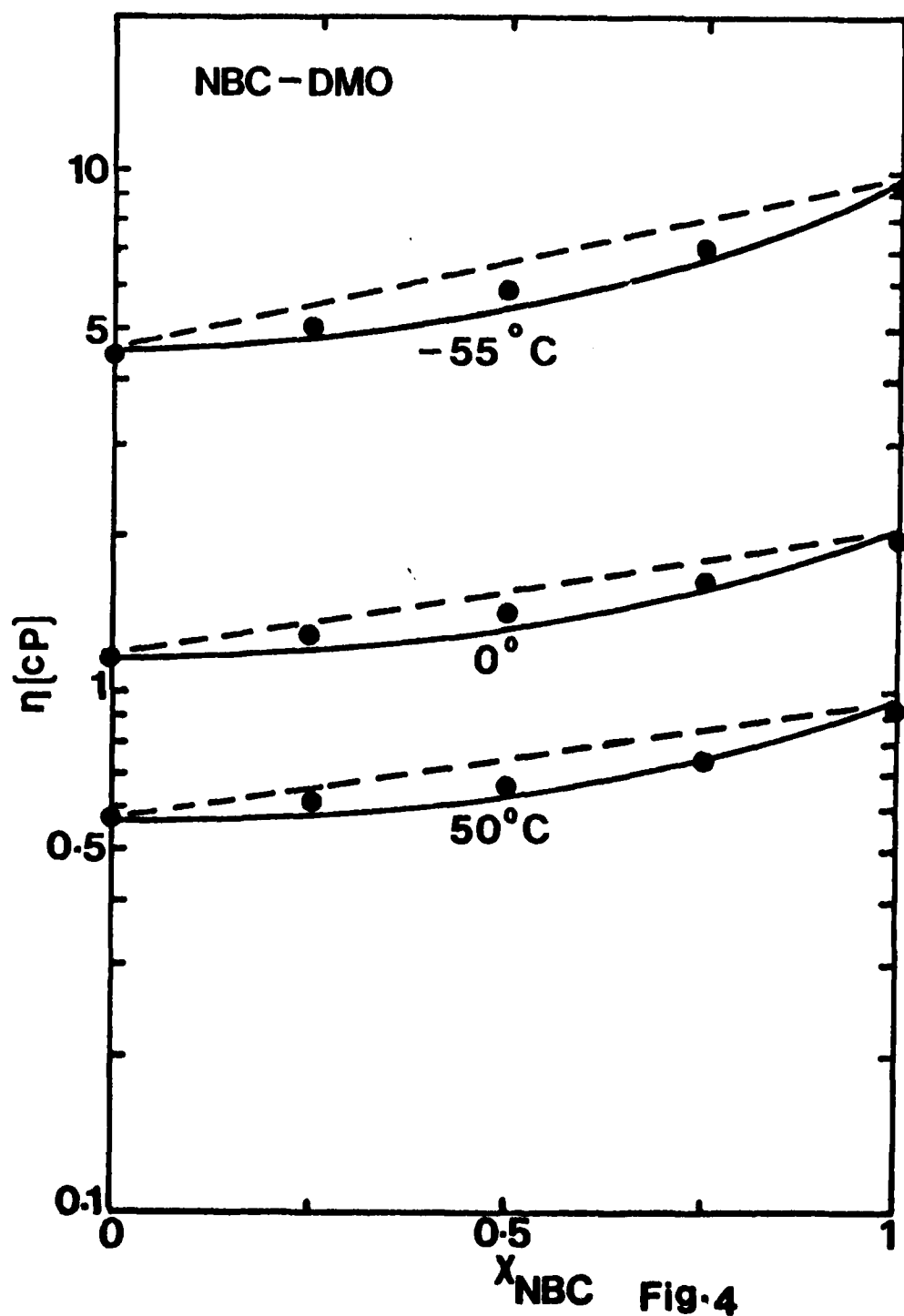


Fig.3



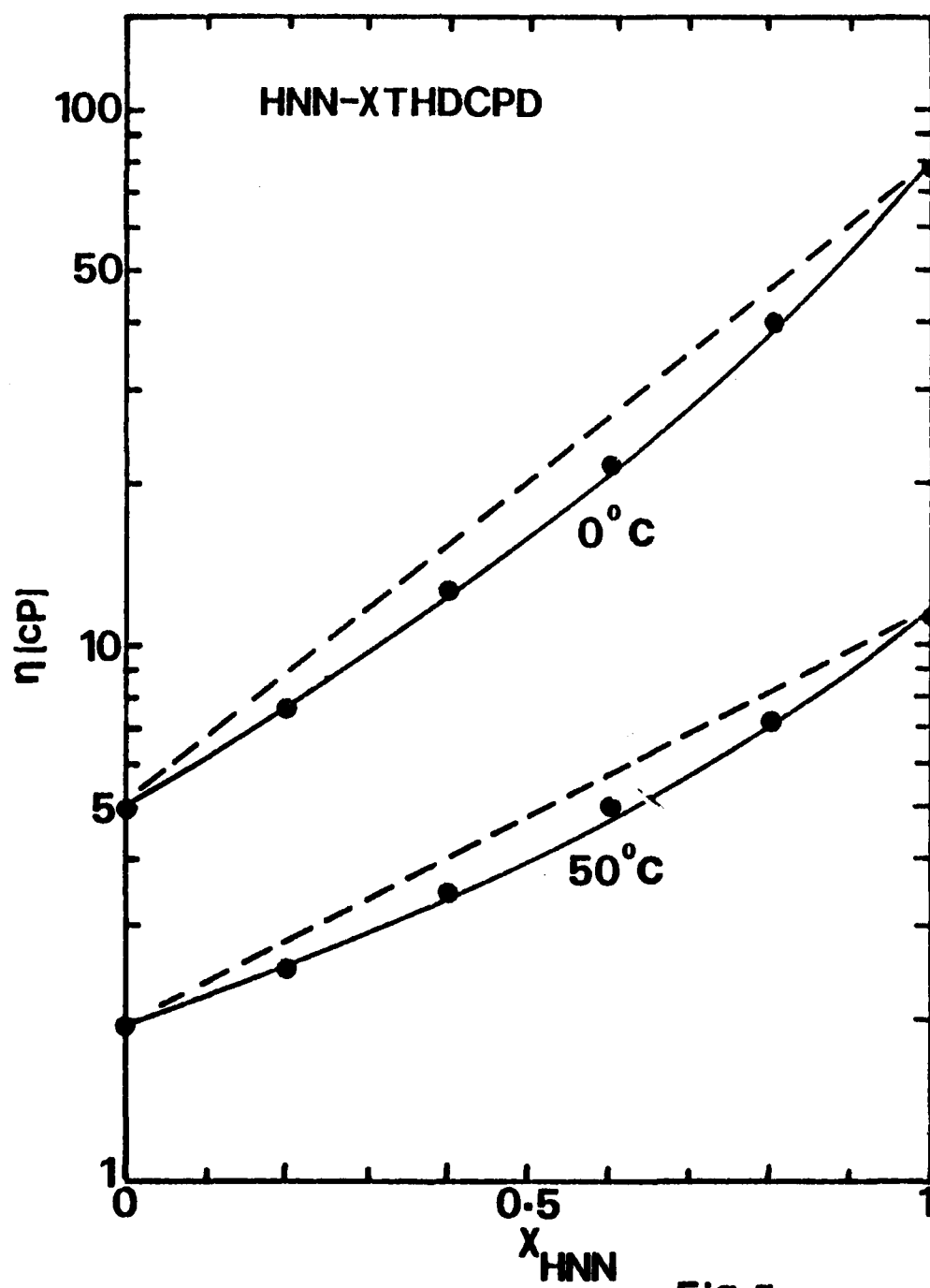


Fig.5

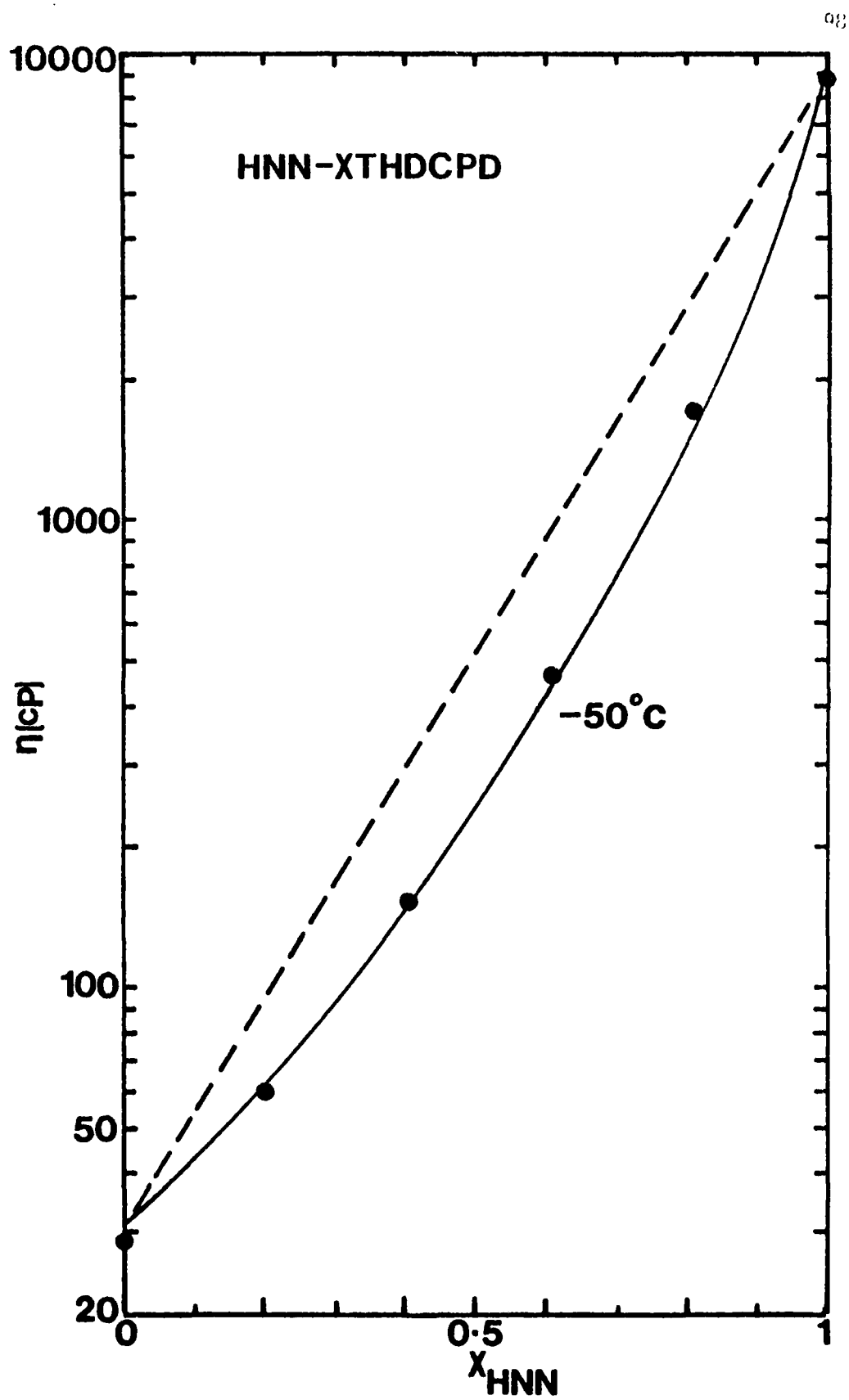


Fig.5a

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temperature range covered in this study. Two simple semi-empirical equations have been developed and applied to the description of the combined composition-temperature dependence of the viscosity of both the hydrocarbons studied under the present contract and two polycyclic hydrocarbons studied under a parallel Air Force Contract. The equations assume additivity of component free volumes in the mixtures and contain no more adjustable parameters than are required to account for the viscosities of the pure hydrocarbon components.

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