SURFACE TENSION IN HYDROCARBON AND RELATED SYSTEMS

By

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AND RELATED SYSTEMS

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CHAPTER I

INTRODUCTION

Surface tension is an important parameter in many scientific and engineering areas, such as heat and mass transfer and reservoir engineering. Reliable experimental data or theoretical correlations are not common.

This study was undertaken to measure experimentally surface tension for binary and ternary mixtures of light hydrocarbons and related gases with heavier hydrocarbons. Once the experimental values were determined, the general applicability of the excess surface tension correlation was examined.

The method used for determining surface tension was the pendant drop procedure. The experimental apparatus consisted of a high-pressure pendant drop apparatus, temperature control, and an optical system.

Experimental measurements were made for methane-n-nonane, ethane-n-nonane, n-butane-n-decane, carbon dioxide-n-decane, and hydrogen sulfide-n-decane binary systems, and for ethane-n-butane-n-decane and methane-carbon dioxide-n-decane ternary systems.

CHAPTER II

LITERATURE REVIEW

Definitions and Experimental Methods

Surface tension is defined as the boundary tension between a liquid and a gas or vapor (2); it may also be defined as a measure of the specific free energy between two phases. Because it deals with equilibrium configurations, surface tension occupies a place in the general framework of thermodynamics - it deals with the macroscopic behavior of interfaces rather than with the details of their molecular structure (1). Surface tension may be referred to as a free energy per unit area or, equally well, as a force per unit length. Customary units, then, may either be ergs/cm^2 or dynes/cm; these are identical dimensionally.

Specifically, surface tension may be measured between two immiscible phases; e.g., two liquids, or between a liquid and a gas or a liquid and its own vapor. This last category, surface tension of a liquid in equilibrium with its own vapor, is the concern of this work.

There are several experimental methods for measuring surface tension. These include: capillary rise, drop weight, bubble pressure, and pendant drop methods. Adamson (1) presents a comprehensive analysis of the different methods.

Behavior of a liquid in a capillary tube is the basis for the capillary rise method. The height of rise of a liquid in the capillary tube determines its surface tension. While the method is valuable for some liquids, it nevertheless has some drawbacks. Comparison of different capillary tubes is not easy; and determination of tube diameter is an indirect procedure.

The drop weight method utilizes volumetric data from falling drops. The method is empirical and uses correction factors which is a disadvantage. The method is not absolute; it requires calibration of the apparatus with a liquid of known surface tension.

The bubble pressure method measures the pressure required to liberate bubbles from a capillary tube immersed in a liquid vertically. New bubbles carry away any impurities attaching to the capillary. Also contact angle is not important.

Pendant Drop Method

Surface tension is determined in the pendant drop method, used in this study, by measurements made on drops hanging from a tip. The method is absolute, requiring no calibration or correction factors, and has been subjected to complete mathematical analysis. As elaborate optical equipment has become available, the pendant drop technique has proven to be among the most reliable methods for determining surface tension, and, therefore, has been widely accepted.

The method is inherently usable under extreme conditions. High temperatures and pressures are handled without extra difficulty. Reactive materials, viscous liquids, and toxic gases require no special arrangements in this method.

The photograph of the drop serves as a permanent record, which is an important advantage over other methods. Also the results are independent of contact angle.

A drop, hanging from a tip, elongates as it grows larger because the variation in hydrostatic pressure eventually becomes appreciable in comparison with that given by the curvature at the apex. This is described by the Laplace and Young equation

$$P = \gamma \left(\frac{1}{R_1} + \frac{1}{R_2}\right). \qquad (1)$$

In other words the product of surface tension and the mean radius of curvature determines the pressure difference between two sides of a curved interface.

In the case of a figure of revolution, the two radii of curvature must be equal at the apex; e.g., at the bottom of a pendant drop. If this radius of curvature is denoted by b, and the elevation of a general point on the surface is denoted by z, then Equation (1) may be written as (1)

$$P = (d_L - d_V)\, g\, z + \frac{2\,\gamma}{b} \qquad (2)$$

But Equation (2) contains b, the measurement of which presents a difficulty. Andreas et al. (2) felt that the most conveniently measurable shape dependent quantity was

$$S = \frac{d_s}{d_e} \qquad (3)$$

As indicated by Figure 1, d_e is the equatorial diameter and d_s is the diameter measured a distance d_e up from the bottom of the drop. Define a new parameter β as

$$\beta = \frac{-\, g\, b^2\, (d_L - d_V)}{\gamma}. \qquad (4)$$

Again β cannot be measured precisely or quickly from a photograph of the drop. But the combination of S and β resolves this difficulty. A new shape dependent parameter, H, is defined as

$$H = -\,\beta\, \left(\frac{d_e}{b}\right)^2. \qquad (5)$$

Thus

$$\begin{aligned} \gamma &= \frac{-\,(d_L - d_V)\, g\, b^2}{\beta} \\ &= \frac{-\,(d_L - d_V)\, g\, d_e^{\,2}}{\beta\,(\, de/b)^2} \\ &= \frac{(d_L - d_V)\, g\, d_e^{\,2}}{H}\ . \end{aligned} \qquad (6)$$

The relationship between the shape dependent factor H and the experimentally measurable shape dependent quantity S

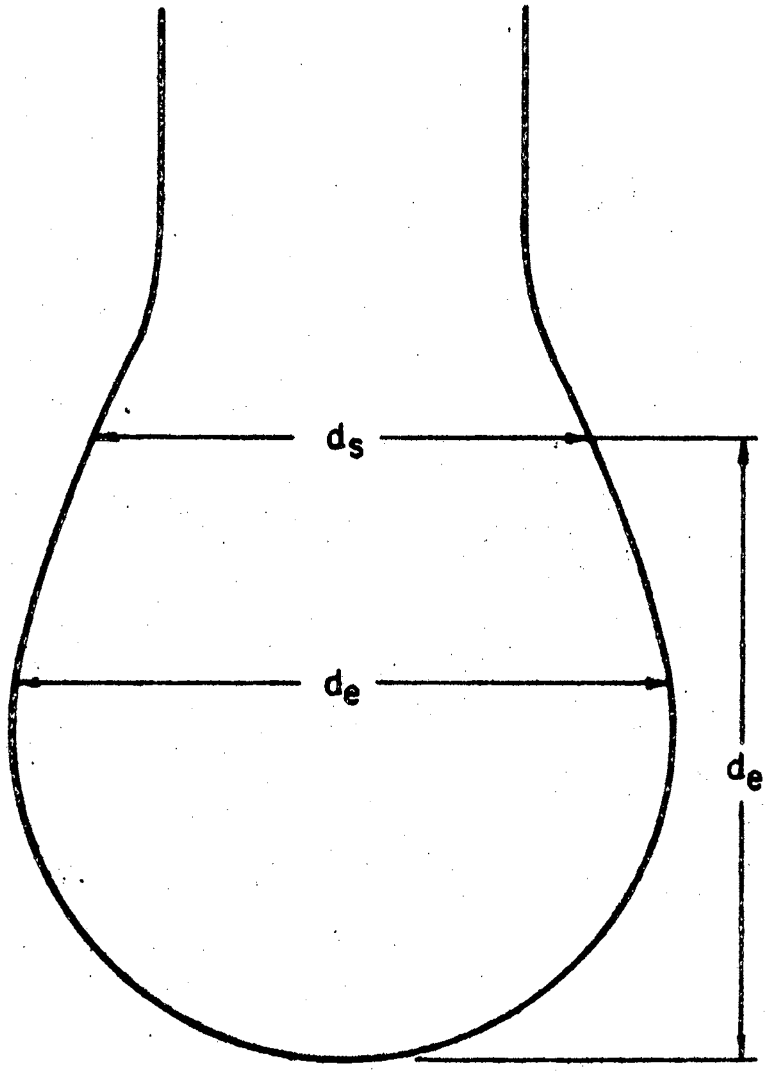

Figure 1. Experimentally Measured Parameters

is then determined and tabulated for ease of reference. Shape factors, H, for S values are obtained by solving profile equations by the Bashforth-Adams (4) integration formula. Niederhauser and Bartell (18) checked earlier tabulations by Forham (10) and found them rather accurate.

Deam (7) used a method of successive approximations for shape factor evaluation, and his results agreed well with the previous results derived by the Bashforth-Adams technique. This work used the results of Deam for evaluation of shape factors.

Theory and Correlations

Since the second half of the nineteenth century several workers have attempted to theoretically correlate surface tension or to, at least, empirically estimate it. These attempts, however, were only moderately successful.

Katayama (12) modified some earlier equations and took into account vapor and liquid densities as well as temperature

$$\gamma \left(\frac{M}{\Delta d}\right)^{2/3} = k \, (T_c - T) \qquad (7)$$

This equation applied to unassociated and nonpolar liquids. The constant k had a value of 2.12, but varied from 1.5 to 2.6 for different liquids.

Van der Waals (30) proposed to relate surface tension to reduced temperature as

$$\gamma = \gamma_o \left(1 - T_r \right)^n . \tag{8}$$

Van der Waals reported n to be 1.5; Ferguson (9), later, gave the exponent as 1.2.

Sugden (29) proposed that surface tension may be estimated by use of the parachor

$$\left[P \right] = \frac{M \gamma^{\frac{1}{4}}}{\Delta d} \tag{9}$$

where the parachor would be an additive property. Through statistical considerations of a liquid in contact with its own vapor, Fowler (11) deduced independently the parachor relationship.

Weinaug and Katz (31) extended the parachor treatment to better apply at high pressures

$$\gamma^{\frac{1}{4}} = \frac{d_L}{M_L} \Sigma x_i \left[P_i \right] - \frac{d_V}{M_V} \Sigma Y_i \left[P_i \right] . \tag{10}$$

They found that Equation (10) gave good agreement with their experimental values for the methane-propane system.

Different workers have recently measured surface tension. Most of them tried to apply the Weinaug and Katz relationship; some of them varied the exponent or the parachor values.

More recently, Deam (7) studied the methane-nonane and methane-butane-decane systems between -30 and 100°F. He reported that methane acted like a dissolved gas at such temperatures, which are above the methane critical temperature. He defined an excess surface tension function, to apply only above the critical temperature of the gas

$$\gamma^E = \gamma_m - \Sigma\ x_i\ \gamma_i. \qquad (11)$$

Pure component surface tension values are calculated by Equation (8). Pseudo-reduced temperatures of the mixtures are calculated by the Rackett technique and apply only to the super-critical components. Excess surface tension values were all negative, indicating a dissolved methane effect of lowering the surface tension of the mixture. Deam, further presented a diagram relating excess surface tension to methane concentration and pseudo-reduced temperature. Below the critical temperature of the gas, Deam proposed that Equation (8) applies, without any corrections for dissolved gas effects. Deam tested the applicability of his diagram to non-methane light components. He used experimental nitrogen-heptane and ethylene-heptane data and found that deviations were reasonable - considering that only methane data were used to construct his diagram. He also predicted that the same diagram could be used for other light components in heavier hydrocarbons.

This work was undertaken to attempt to apply the excess surface tension theory to some other hydrocarbon and related systems. Ethane-nonane, carbon dioxide-decane, and hydrogen sulfide-decane binaries, and ethane-butane-decane and methane-carbon dioxide-decane ternary systems were selected for this study.

CHAPTER III

EXPERIMENTAL APPARATUS AND PROCEDURE

Experimental Apparatus

The experimental apparatus consists of a high pressure cell, a drop forming unit, a gas cylinder, a temperature control system, and an optical system. The apparatus is schematically shown in Figure 2.

The cell is stainless steel with a rating of 1500 psi. It has a holding volume of fourteen cubic centimeters. Two viewports, with quartz lenses, expose the drop forming tip and the drop. Each lens, one inch in diameter, is held tightly in place between a silicon rubber seal, on the inside, and a teflon ring in front of the annular threaded plug, on the outside; the cell components are shown in Figure 3. The viewports have a 0.625 inch diameter opening. The capillary line, with the tip, screw into the cell from the top side. Gas inlet is provided for at the bottom face of the cell. The cell, capillary line, and liquid reservoir are contained in a temperature bath.

The capillary line, 0.087 inches in inside diameter and six inches long, connects the liquid reservoir to the cell. The tip is a Yale Number 15 stainless steel

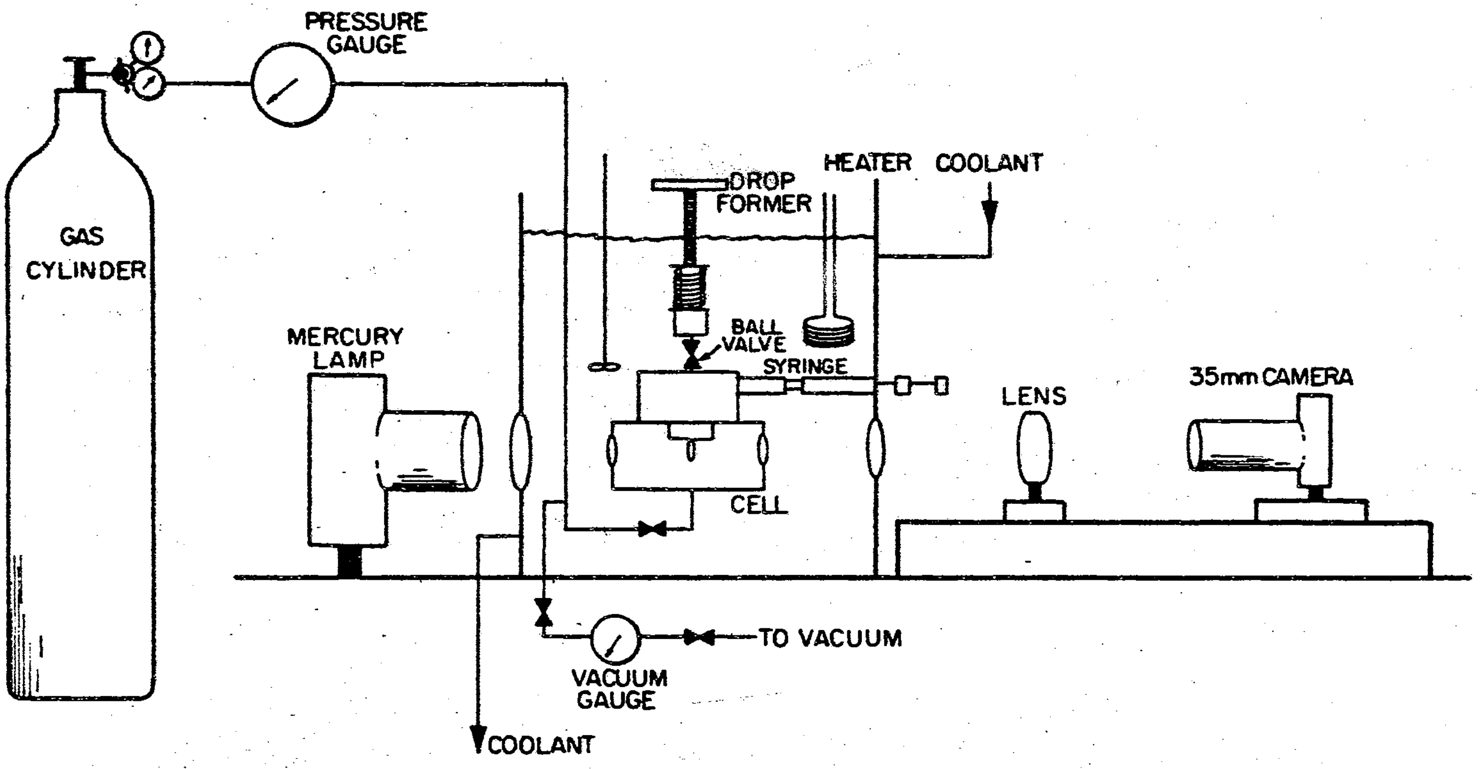
PRESSURE GAUGE
GAS CYLINDER
DROP FORMER
HEATER
COOLANT
BALL VALVE
SYRINGE
MERCURY LAMP
LENS
35mm CAMERA
CELL
TO VACUUM
VACUUM GAUGE
COOLANT

Figure 2. Experimental Apparatus

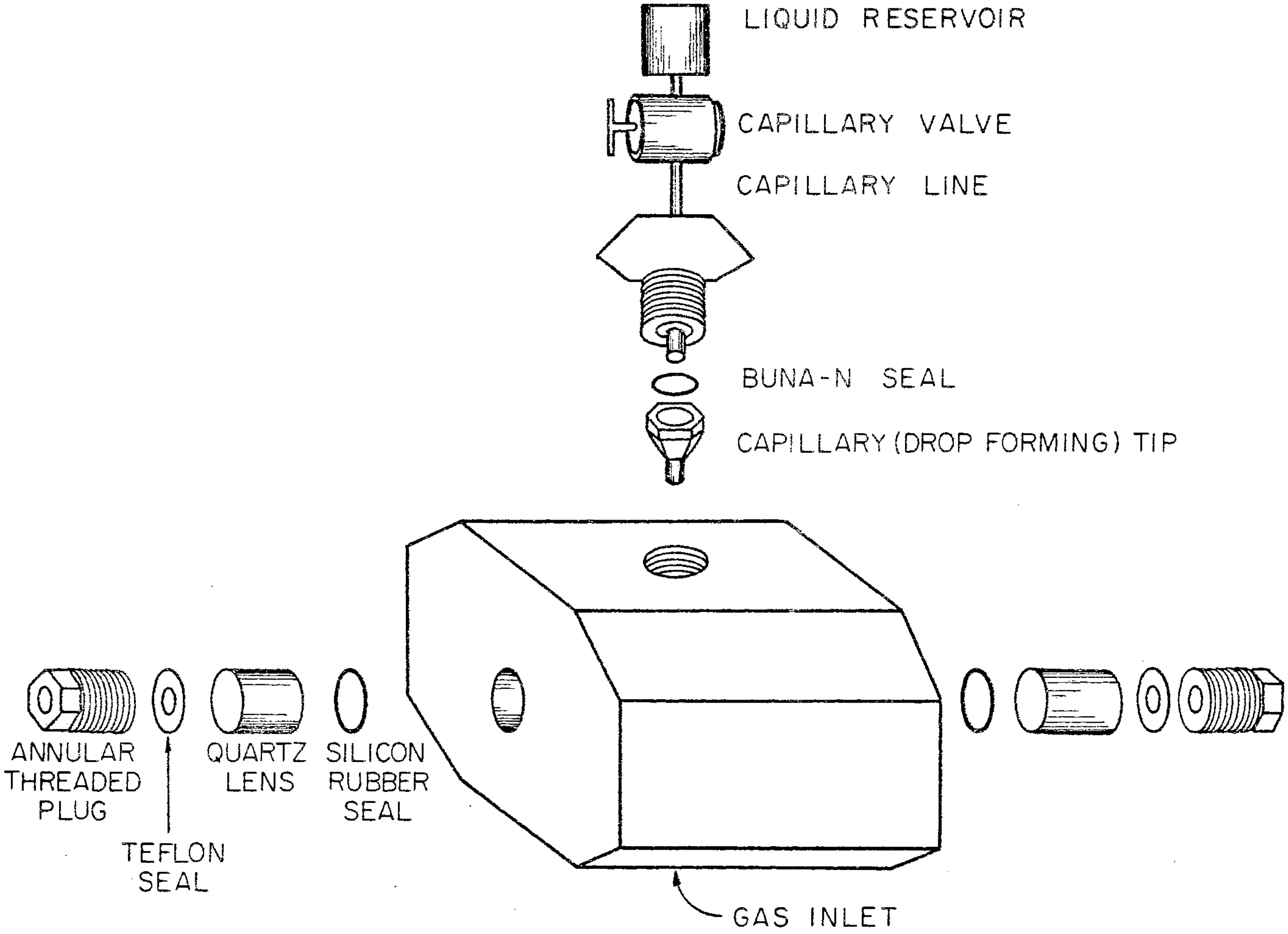
LIQUID RESERVOIR
CAPILLARY VALVE
CAPILLARY LINE
BUNA-N SEAL
CAPILLARY (DROP FORMING) TIP
ANNULAR THREADED PLUG
TEFLON SEAL
QUARTZ LENS
SILICON RUBBER SEAL
GAS INLET

Figure 3. Cell Components

capillary tip with an outside diameter of 0.072 inches; it is fitted on the bottom of the base of the capillary line. The base of the capillary line screws into the cell with a Buna N seal on its bottom to make the connection leak proof. A Circle Seal, ball, capillary valve is mounted in the middle of the capillary line and thus separates the gas in the cell from the liquid in the reservoir. The liquid reservoir has an inside diameter of 0.5 inches. A piston, with a Buna N or Viton seal, forces the liquid through the capillary line and generates the drops on the tip. A vernier screw drives the piston.

The gas cylinder is connected through a pressure regulator and a pressure gauge to the cell. This gas line also leads to a vacuum pump.

The temperature control system consists of a heating system and a refrigeration unit. The heating element is a 300-watt immersion heater, controlled by a Thermistemp Model 63 temperature controller capable of temperature control to 0.1°F. A compression-type refrigeration unit, charged with Freon 22 and capable of cooling to -40°F, is used for low temperatures. Liquid in the bath was methanol-water at temperatures below 32°F, water at temperatures between 32°F and 100°F, and ethylene glycol-water at temperatures above 100°F. The temperature bath is insulated with one inch of magnesia packing between the walls and one inch of fiberglass on the outside.

The optical system consists of a light source, a camera, and an optical comparator. A Cenco 100-watt high-pressure, mercury arc lamp provides light from one window of the temperature bath and through the cell. At the other window of the temperature bath, a lens is placed, followed by a Konica Model FS 35 mm single-lens reflex camera. A Kodak extreme resolution panchromatic film is used. Drops, recorded on film, are measured by projecting them on a Vangard Model C-11D motion analyzer of 30X magnification.

Hydrocarbons and other materials used in the study, obtained from Phillips Petroleum Company, have the following specifications:

	mole per cent	
research-grade n-nonane	99.68	
	0.32	iso-nonanes
research-grade n-decane	99.49	
	0.51	iso-decanes
instrument-grade n-butane	99.55	
	0.45	iso-butane
	0.05	propane
pure-grade ethane	99.	min.
instrument-grade methane	99.29	
	0.60	nitrogen
	20	ppm max. oxygen
	10	ppm max. water
bone-dry carbon dioxide	99.8	min.
pure-grade hydrogn sulfide	99.6	min.

Experimental Procedure

At a given temperature, an isotherm, an experiment consisted of surface tension measurements at several pressures. At each pressure about six photographs were taken, normally for more than one pendant drop as described later.

Prior to an experiment, the cell and drop tip were given a detergent wash in warm distilled water followed by three distilled water washes. Three absolute ethyl alcohol washes followed the water washes. All the washes were conducted in an ultrasonic cleaner and lasted 15 minutes each. After washing, the apparatus was drained and dried by passing filtered air through it. Thereafter, the apparatus was assembled, ready for a run.

Binary Procedure

Once the system was assembled, the temperature bath was filled with the appropriate liquid (depending on the temperature of the run) and the system was brought to thermal equilibrium. In the meanwhile it was ascertained that the system was leak proof by raising the system pressure to more than the highest pressure to be attained in the experiment.

When the system reached thermal equilibrium at the desired temperature and was leak free, the entire system was evacuated, to less than 29 inches of vacuum, then flushed with the gas to be used in the experiment, and evacuated again. After that the system pressure was increased to slightly above atmospheric pressure. The piston was withdrawn, and the desired liquid hydrocarbon was charged via a syringe. Minimal liquid was added, normally about ten drops. With the valve in the capillary line in the open position, the gas was allowed to bubble through the liquid for a few minutes to let the system approach mass equilibrium. Before the piston was reinserted, the valve was closed to prevent the liquid from going through the capillary line into the cell while inserting the piston. The cell pressure was increased to the desired first pressure.

A few shots of the bare tip were taken at this point before any liquid had covered it in order to use these pictures for determining the true measurements of the drops later on. The capillary valve was then opened. To enhance mass equilibrium, the vernier screw was used to force the liquid down the capillary line and onto the tip thus contacting the vapor in the cell, and then it was sucked back into the liquid reservoir to mix the liquid. This was repeated until mass equilibrium was achieved. This required, on the average, 45 minutes to one hour. Before mass equilibrium was reached, the drop was unstable when it formed on the tip; it would oscillate up and down. This oscillation phenomenon was also observed by Deam (7). When mass equilibrium was attained, the drop would form on the tip and exhibit a great deal of stability to the extent of staying pendant on the tip for thirty minutes to one hour if not disturbed. It should be remarked, though, that a truly pendant drop is in a state of unstable mechanical equilibrium--like a ball seated on the top of another ball; the least disturbance--even building traffic-would set it loose.

When mass equilibrium was achieved, the critical step of the pendant drop procedure was at hand, namely to obtain a truly pendant drop and record three to eight of its photographs on film. Oftentimes one drop would not stay that long and another drop is thus required. During the experiment it was, in general, difficult to ascertain that a drop was pendant unless a previous drop was lost from the same position when it was forced to go any farther. A distinguishing quiver was unmistakably exhibited, momentarily, by every new drop as soon as it reached the pendant stage.

A non-pendant drop, if used, would lead to eroneous surface tension values, and that is why it was so critical to have truly pendant drops. Figure 4 shows examples of both types of drops, the pendant and the non-pendant.

Ternary Procedure

The general procedure for a ternary resembles closely that of a binary. Liquid hydrocarbon was charged to the reservoir--while the capillary valve was closed. The

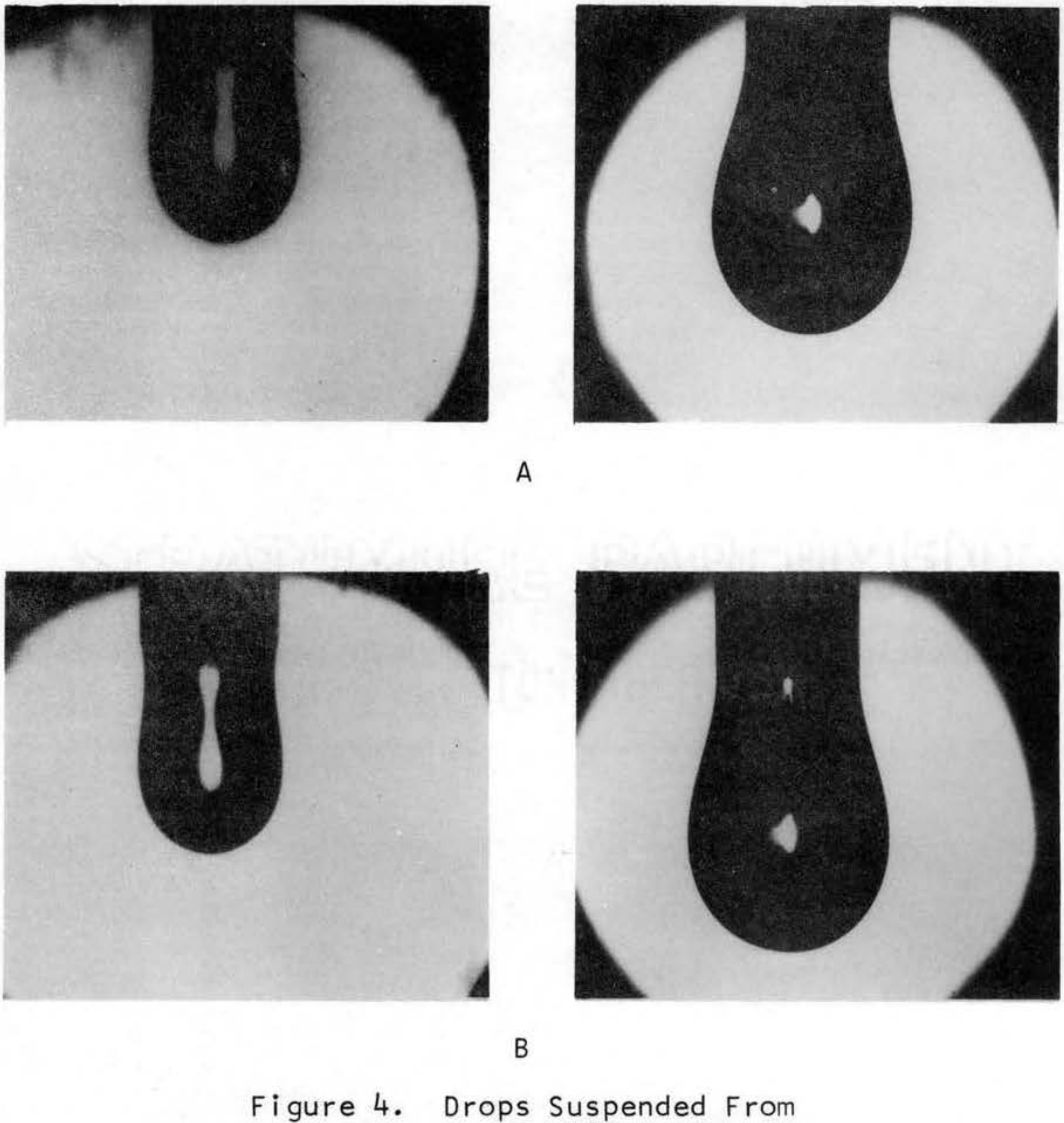

Figure 4. Drops Suspended From
a Capillary Tip

(A) Non-Pendant Drops

(B) Pendant Drops

cell was evacuated, as described before, and then n-butane pressure was adjusted to achieve the desired butane liquid composition. When mass equilibrium was reached by the binary, at the desired bubble point pressure, photographs were taken for drops of the binary.

The second gas was then connected to the system and its pressure was brought up to the first desired ternary pressure. Mass equilibrium was again achieved, by the procedure described before, for the ternary. Photographs were taken, and surface tension at consecutive pressures was determined in the same manner.

CHAPTER IV

EXPERIMENTAL RESULTS

One isotherm for the methane-nonane system was experimentally determined at 70°F; a pressure range from 50 psia to 1000 psia was investigated. Data for this system are presented in Table I and Figure 5. These data represent measurements on saturated liquid mixtures. Drop measurements and phase densities generate the surface tension values by use of Equation (6).

Experimental data for the ethane-nonane binary system are presented in Table II and Figure 6. The system was studied at 0°F, 32°F, 58°F, 130°F, and 170°F isotherms. The pressure range was 50 psia to 500 psia. Experimental measurements were taken after mass equilibrium was reached between the vapor and liquid phases for every temperature and pressure. Surface tension values were calculated from phase densities and experimental drop measurements combined with Equation (6).

Surface tension values for the carbon dioxide-decane binary system are shown in Table III and Figure 7. Isotherms studied were 0°F, 32°F, 40°F, 76°F, and 160°F. Several pressures from about 50 psia to about 800 psia were studied. Measurements were made on liquids saturated

TABLE I

EXPERIMENTAL SURFACE TENSION OF METHANE-NONANE SYSTEM

Temp. °F	Press. psia	Surface Tension dynes/cm	Ave. Value	Methane Mole Fraction
70	50	21.86	22.05	0.0183
		21.92		
		22.00		
		21.94		
		21.98		
		22.24		
		22.20		
	100	21.72	21.70	0.0363
		21.73		
		21.65		
		21.68		
		21.72		
	300	19.63	19.57	0.1042
		19.60		
		19.38		
		19.66		
	500	17.23	17.14	0.1662
		17.03		
		16.98		
		17.28		
		17.22		
		17.31		
		17.18		
		17.02		
		17.04		
	750	14.53	14.51	0.2360
		14.45		
		14.44		
		14.57		
		14.56		
		14.58		
		14.59		
	1000	12.52	12.51	0.2979
		12.49		
		12.54		
		12.51		
		12.49		

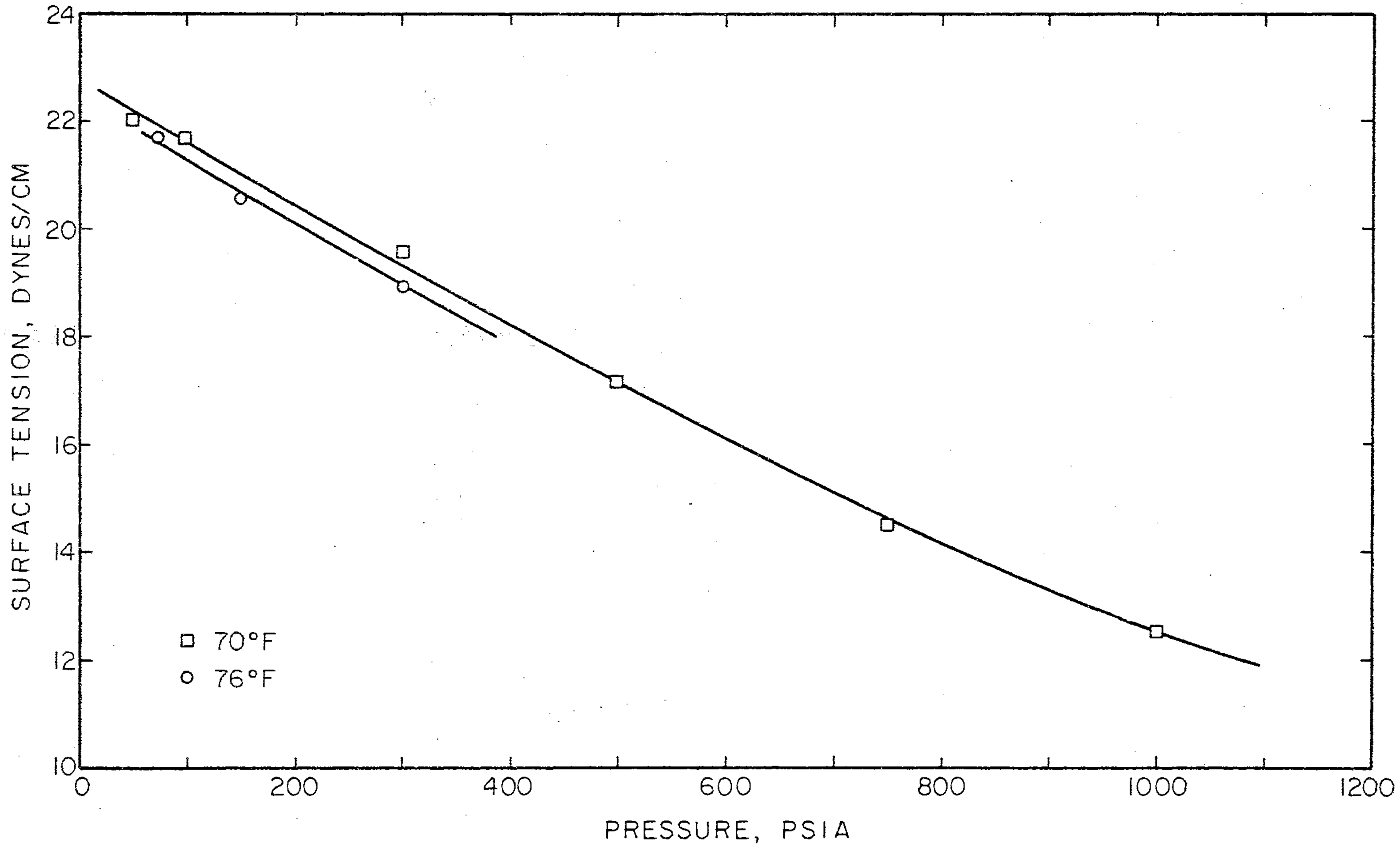

Figure 5. Isothermal Surface Tension of Methane-Nonane System

TABLE II

EXPERIMENTAL SURFACE TENSION OF ETHANE-NONANE SYSTEM

T = 0°F			
P psia	Surface Tension dynes/cm	Ave. Value	Ethane Mole Fraction
50	21.41	21.34	0.2197
	21.57		
	21.44		
	20.95		
100	16.83	16.47	0.4501
	16.29		
	16.28		
200	10.33	10.33	0.5000

T = 32°F			
P psia	Surface Tension dynes/cm	Ave. Value	Ethane Mole Fraction
50	22.14	22.24	0.1501
	22.18		
	22.15		
	22.29		
	22.19		
	22.50		
100	18.82	18.53	0.2998
	18.59		
	18.57		
	18.37		
	18.42		
	18.42		
200	12.64	12.71	0.5000
	12.54		
	13.01		
	12.59		
	12.83		
	12.60		
	12.54		
	13.03		
	12.79		
	12.52		
300	7.66	7.59	0.5000
	7.53		
	7.58		

T = 58°F			
P psia	Surface Tension dynes/cm	Ave. Value	Ethane Mole Fraction
320	8.33	8.12	0.7231
	8.02		
	7.87		
	8.26		
	8.23		
	8.27		
	7.90		
	8.33		
	8.01		
	8.36		
	8.04		
	7.95		
	7.91		
	8.01		
	8.15		
	8.20		
	8.38		
	8.16		
	8.02		

T = 130°F			
P psia	Surface Tension dynes/cm	Ave. Value	Ethane Mole Fraction
320	11.45	11.37	0.3887
	11.55		
	11.37		
	11.10		
	11.40		
	11.26		
	11.15		
	11.57		
	11.60		

T = 170°F			
P psia	Surface Tension dynes/cm	Ave. Value	Ethane Mole Fraction
75	16.12	15.86	0.0854
	15.80		
	15.87		
	15.75		
	15.81		
	16.03		
	15.75		
	15.73		
150	15.08	14.82	0.1425
	14.81		
	14.65		
	14.73		
	14.83		
	15.17		
	14.76		
	14.54		
	14.84		
300	12.22	12.28	0.2833
	12.29		
	12.26		
	12.21		
	12.41		
500	7.67	7.83	0.4669
	7.81		
	7.89		
	7.95		

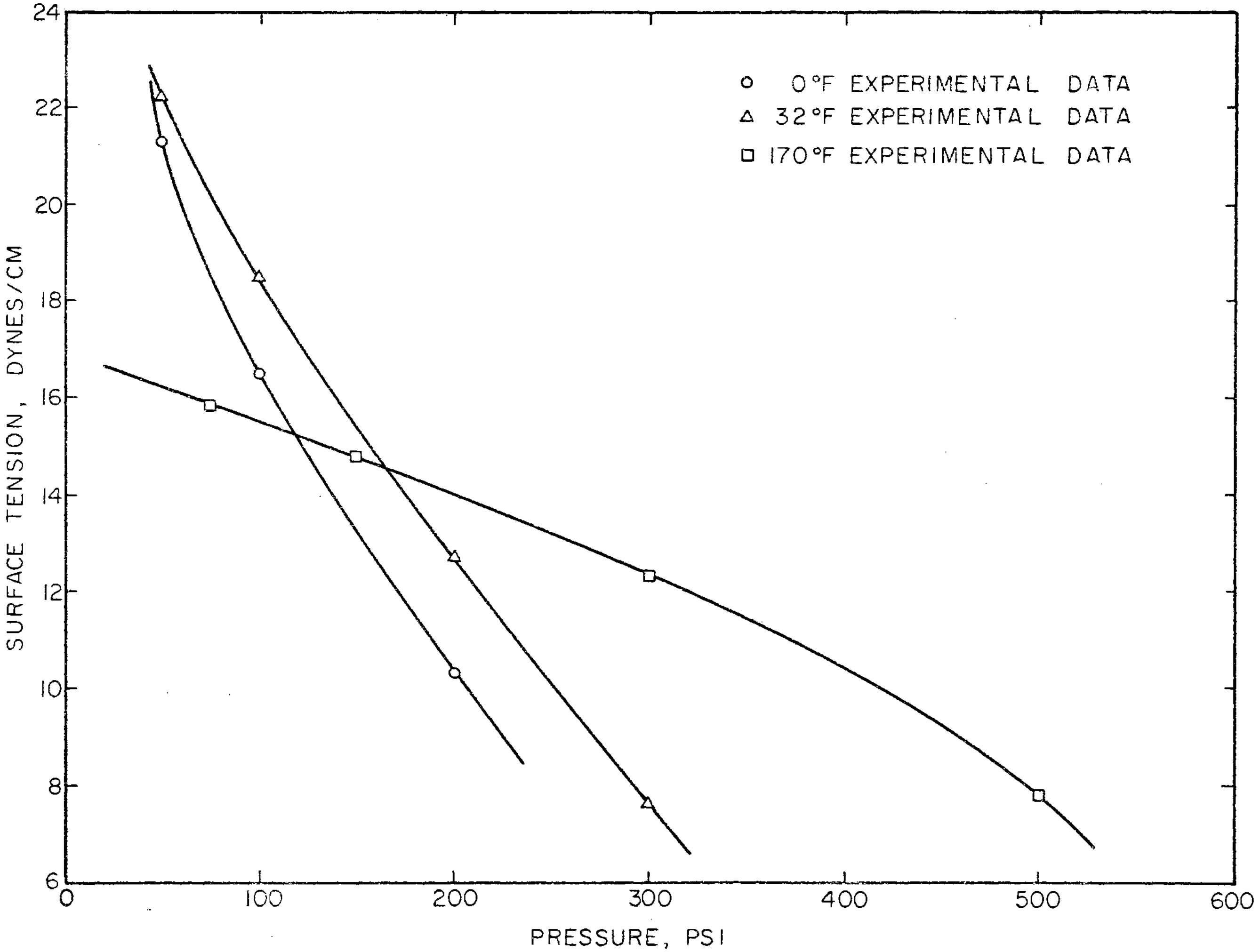

Figure 6. Isothermal Surface Tension of Ethane-Nonane System

TABLE III

EXPERIMENTAL SURFACE TENSION OF CARBON DIOXIDE-DECANE SYSTEM

T=0° F				T=40° F				T=160° F			
Press. psia	Surface Tension dynes/cm	Ave. Value	Carbon Dioxide Mole Fraction	Press. psia	Surface Tension dynes/cm	Ave. Value	Carbon Dioxide Mole Fraction	Press. psia	Surface Tension dynes/cm	Ave. Value	Carbon Dioxide Mole Fraction
50	27.38	27.31	0.1376	75	22.41	22.43	0.0843	200	16.05	15.77	0.1118
	27.28				22.32				15.46		
	27.28				22.30				15.63		
					22.73				15.44		
100	24.54	25.01	0.2709		22.41				16.06		
	25.05								15.61		
	25.14			150	20.62	20.73	0.1627		16.12		
	25.27				20.89						
	25.08				20.95			400	13.24	13.40	0.2127
	24.96				20.31				13.35		
					20.46				13.24		
300	10.01	9.89	0.5000		20.96				13.55		
	10.00				20.95				13.60		
	9.64										
	9.90			300	16.23	16.51	0.3355	600	11.79	11.82	0.3077
					16.47				11.93		
					16.44				11.65		
					16.63				11.86		
					16.78				11.88		
									11.81		
				500	8.96	9.04	0.7117				
					8.95			800	10.23	10.24	0.3988
					9.03				9.99		
					9.06				10.55		
					8.92				10.01		
					9.15				10.40		
					9.16						
					9.10						
					8.98						

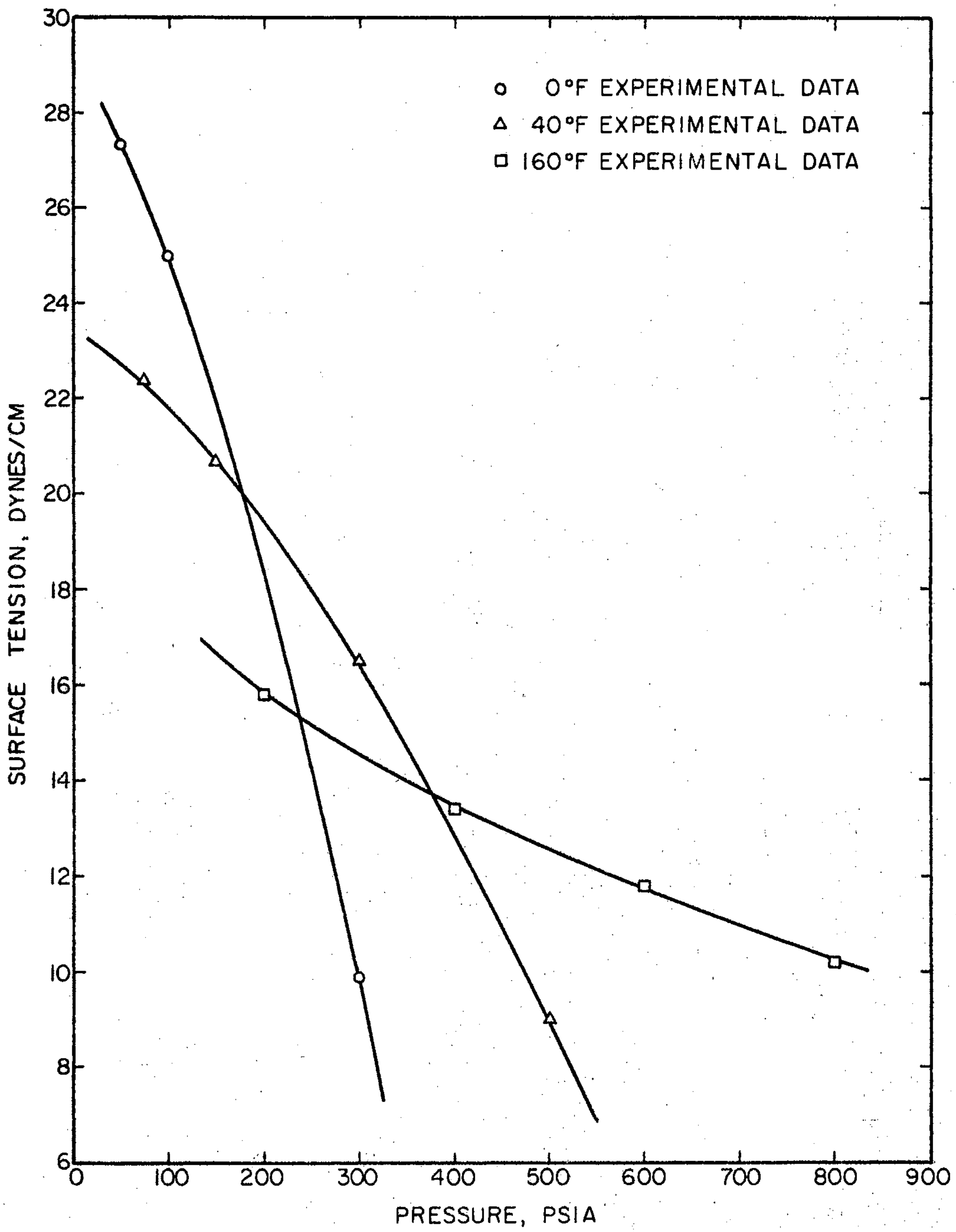

Figure 7. Isothermal Surface Tension of Carbon Dioxide-Decane System

and in contact with their vapors. Drop measurements and phase densities were used to calculate surface tension values via Equation (6). Data at 32 F and 76 F are shown with the methane-carbon dioxide-decane ternary system later.

For the hydrogen sulfide-decane system, temperatures studied were 40°F, 100°F, and 160°F. Pressures ranged from 30 psia to 115 psia. Equation (6) with experimental drop measurements and phase densities were used to calculate surface tension of the saturated mixtures. These data are shown in Table IV and Figure 8.

Surface tension values for the ethane-butane-decane ternary system were determined at 170°F and for pressure values from 50 psia to 500 psia. Data were taken for liquid in equilibrium with its own vapor. Phase densities and drop measurements were used with Equation (6) to find surface tension values. These are shown in Table V and Figure 9.

The ternary system of methane-carbon dioxide-decane and the carbon dioxide-decane binary system were studied at 32 F and 76°F. Table VI shows experimental results for the binary system. Table VII and Figures 10, 11, and 12 present the data for the ternary system. At every temperature a series of pressures were investigated ranging from about 30 psia to 1000 psia. Liquid mixtures were saturated and in contact with their vapors. Phase densities and experimental drop measurements generated

TABLE IV

EXPERIMENTAL SURFACE TENSION OF HYDROGEN SULFIDE-DECANE SYSTEM

T = 40°F				T = 100°F				T = 160°F			
Press. psia	Surface Tension	Ave. Value	Hydrogen Sulfide Mole Fraction	Press psia	Surface Tension	Ave. Value	Hydrogen Sulfide Mole Fraction	Press psia	Surface Tension	Ave. Value	Hydrogen Sulfide Mole Fraction
50	21.45	21.60	0.195	30	20.18	20.40	0.0707	50	17.93	18.06	0.0796
	21.44				20.79				18.05		
	21.95				20.48				17.91		
	21.55				20.41				18.33		
					20.13						
100	20.55	20.07	0.446					75	17.59	17.61	0.1192
	19.59			50	19.92	19.87	0.1153		17.87		
					20.05				17.65		
115	19.56	19.56	0.546		20.17				17.50		
					19.96				17.46		
					19.52						
					19.66			100	17.35	17.21	0.1585
					19.79				17.06		
									17.23		
				100	18.77	18.82	0.2332				
					18.73						
					18.89						
					18.92						
					18.89						
					18.88						
					18.76						
					18.75						
				115	18.62	18.49	0.2703				
					18.37						
					18.48						
					18.33						
					18.53						
					18.58						
					18.36						
					18.65						

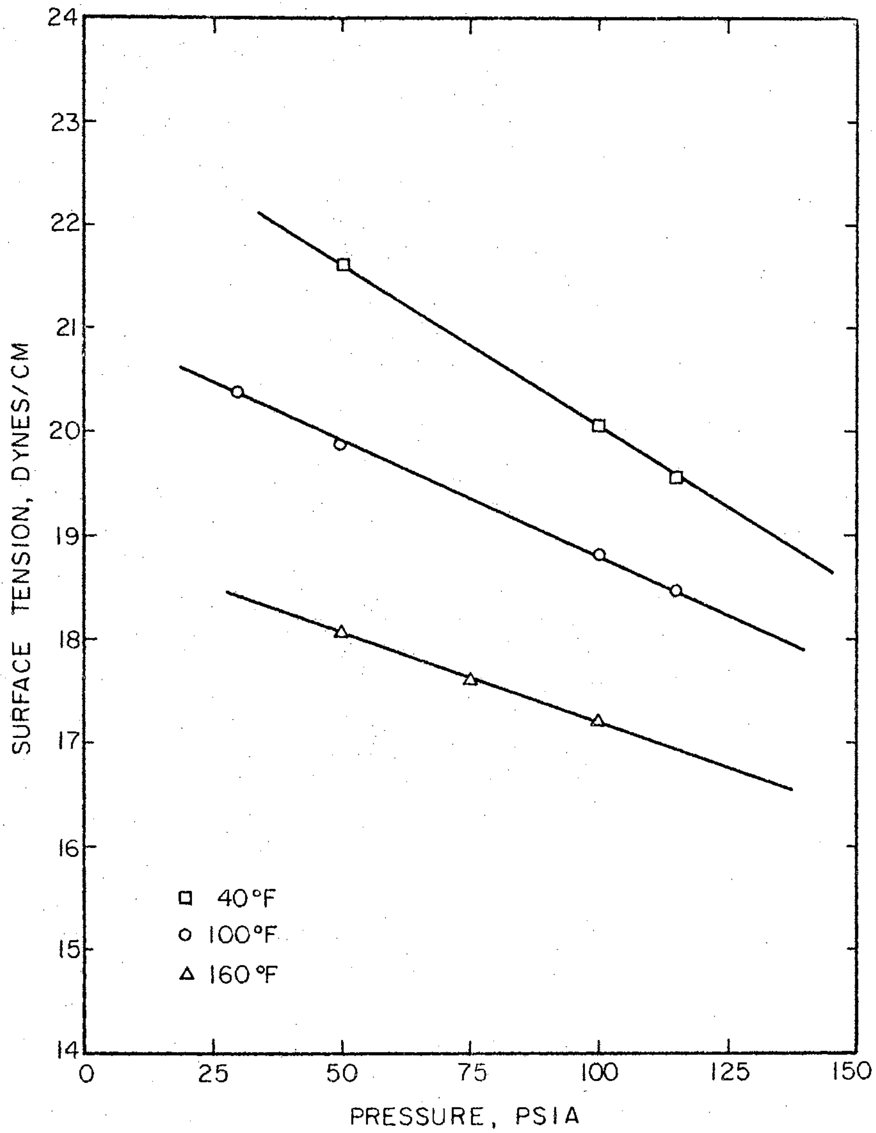

Figure 8. Isothermal Surface Tension Hydrogen Sulfide-Decane System

TABLE V

EXPERIMENTAL SURFACE TENSION OF ETHANE-BUTANE-DECANE SYSTEM

Temp. °F	Press. psia	Surface Tension dynes/cm	Ave. Value	Mole Fraction Ethane	Mole Fraction Butane
170	50	13.88	14.10	0.0	0.3873
		14.24			
		14.00			
		14.26			
		14.14			
	100	13.16	13.22	0.0619	0.3605
		13.24			
		13.26			
	200	12.18	11.99	0.1618	0.3221
		11.93			
		12.16			
		11.95			
		11.90			
		11.96			
		11.84			
	300	10.20	10.15	0.2578	0.2856
		10.04			
		10.00			
		10.39			
	400	8.34	8.65	0.3520	0.2493
		8.34			
		8.90			
		8.63			
		8.92			
		8.54			
		8.94			
		8.57			
	500	7.10	7.21	0.4448	0.2133
		7.36			
		7.10			
		7.28			
		7.32			
		7.12			
		7.20			

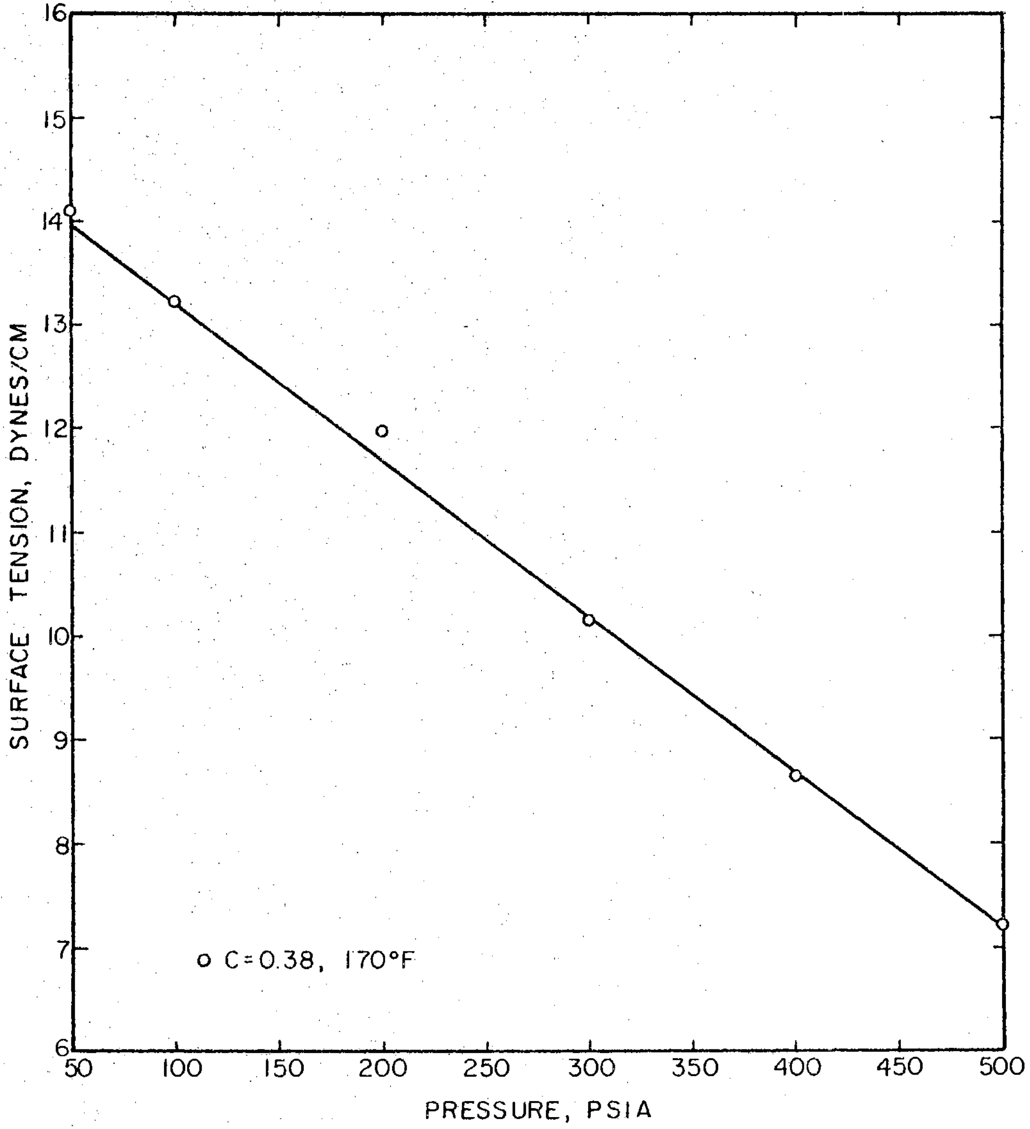

Figure 9. Isothermal Surface Tension of Ethane-Butane-Decane System

TABLE VI

EXPERIMENTAL SURFACE TENSION OF CARBON DIOXIDE-DECANE SYSTEM

Composition Parameter	Temp. °F	Press. psia	Surface Tension	Ave. Value	Carbon Dioxide Mole Fraction
0.40	76	34	21.64	21.78	0.0384
			21.91		
			21.63		
			21.85		
			21.89		
			21.74		
			21.65		
			21.85		
			21.86		
0.35	32	34	23.37	23.50	0.0625
			23.57		
			23.45		
			23.61		
0.52	32	50	22.25	22.73	0.0917
			22.97		
			22.97		
0.68	32	100	21.13	21.34	0.1817
			21.16		
			21.55		
			21.30		
			21.58		

TABLE VII

EXPERIMENTAL SURFACE TENSION OF METHANE-CARBON DIOXIDE-DECANE SYSTEM

Composition Parameter	T °F	P psia	Exp. Value	Ave. Value	Mole Fraction	
					Methane	Carbon Dioxide
0.40	76	100	21.29 21.06 21.33 21.10 21.40 21.23 21.30 21.04 20.96 21.19 21.28	21.20	0.0216	0.0425
		200	20.25 20.22 20.09 20.03 20.00 20.18 20.43	20.17	0.0554	0.0415
		300	19.06 18.68 19.09 18.68 18.74 18.74	18.83	0.1055	0.0406
		500	16.36 16.23 16.34 16.10	16.26	0.2012	0.0386
		750	14.30 14.19 14.29	14.26	0.2620	0.0356
		1000	12.22	12.22	0.3442	0.0323
0.35	32	100	22.61 22.55 22.66 22.50 22.49 22.72	22.59	0.0284	0.0551
0.35	32	200	21.41 21.25 21.10 21.63 21.26	21.33	0.0676	0.0529
		300	20.07 20.61 20.26 20.08 20.53 20.56	20.35	0.1047	0.0507
		500	17.34 17.43 17.51 17.43 17.39 17.50 17.45 17.63 17.62 17.79 17.68	17.52	0.1943	0.0462
		750	15.26 15.20 15.13 15.35 15.04 15.01 15.47 15.11	15.20	0.2688	0.0407
		1000	12.85 12.66 12.71 12.88 12.76 12.91 12.69 12.88 12.70 12.54 12.79 12.83 12.82 12.62 12.51	12.74	0.3471	0.0349

TABLE VII (Continued)

Composition Parameter	T °F	P psia	Exp. Value	Ave. Value	Mole Fraction	
					Methane	Carbon Dioxide
0.52	32	200	20.97	20.76	0.0625	0.0759
			20.67			
			20.64			
		500	17.72	17.57	0.1690	0.0663
			17.50			
			17.48			
		750	14.27	14.32	0.2551	0.0583
			14.38			
		1000	12.40	12.60	0.3459	0.0499
			12.67			
			12.74			

Composition Parameter	T °F	P psia	Exp. Value	Ave. Value	Mole Fraction	
					Methane	Carbon Dioxide
0.68	32	200	20.26	20.16	0.0454	0.1529
			19.94			
			20.31			
			19.96			
			20.35			
		300	19.32	19.28	0.0840	0.1456
			19.10			
			19.17			
			19.28			
			19.13			
			19.38			
			19.40			
			19.42			
			19.33			
			19.28			
		500	17.49	17.26	0.1648	0.1321
			17.59			
			17.36			
			17.00			
			17.04			
			17.10			
		750	14.56	14.52	0.2738	0.1157
			14.60			
			14.41			

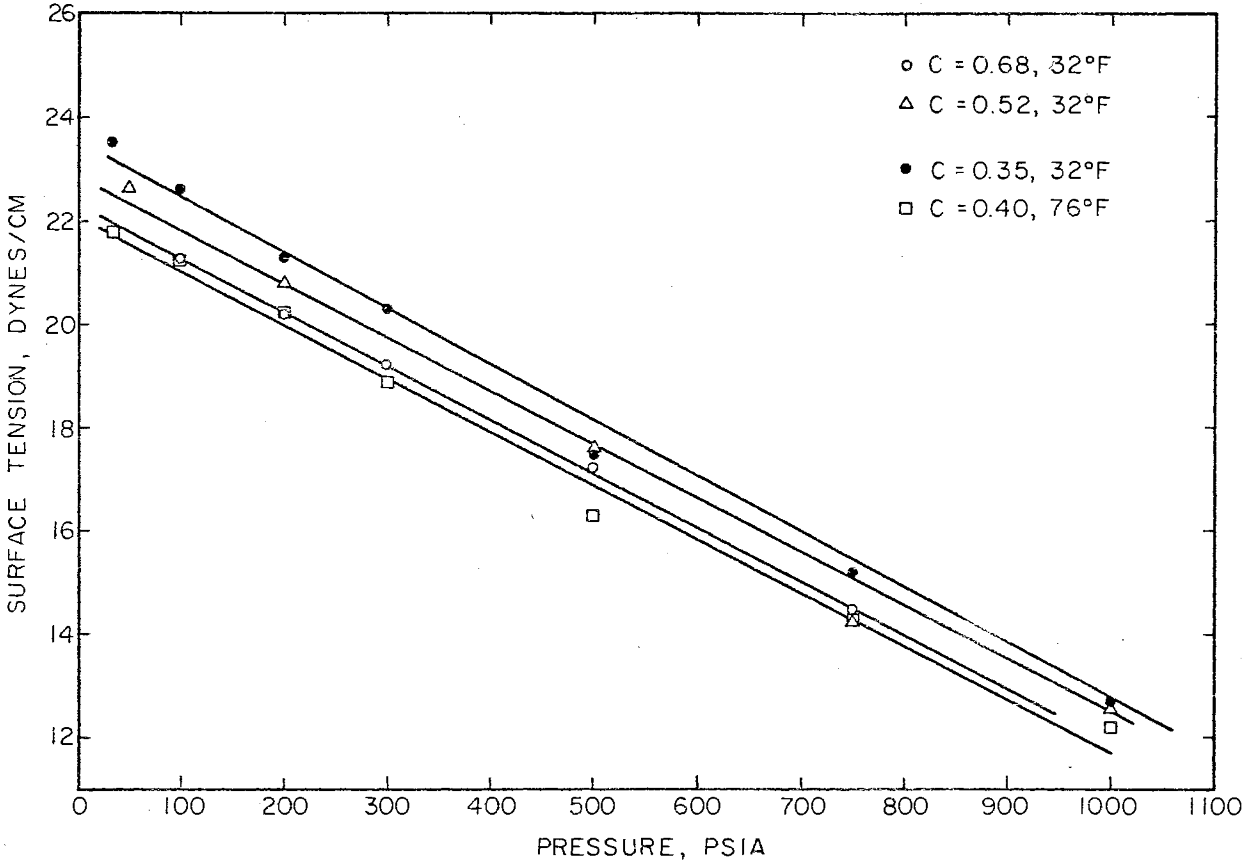

Figure 10. Isothermal Surface Tension of Methane-Carbon Dioxide-Decane System

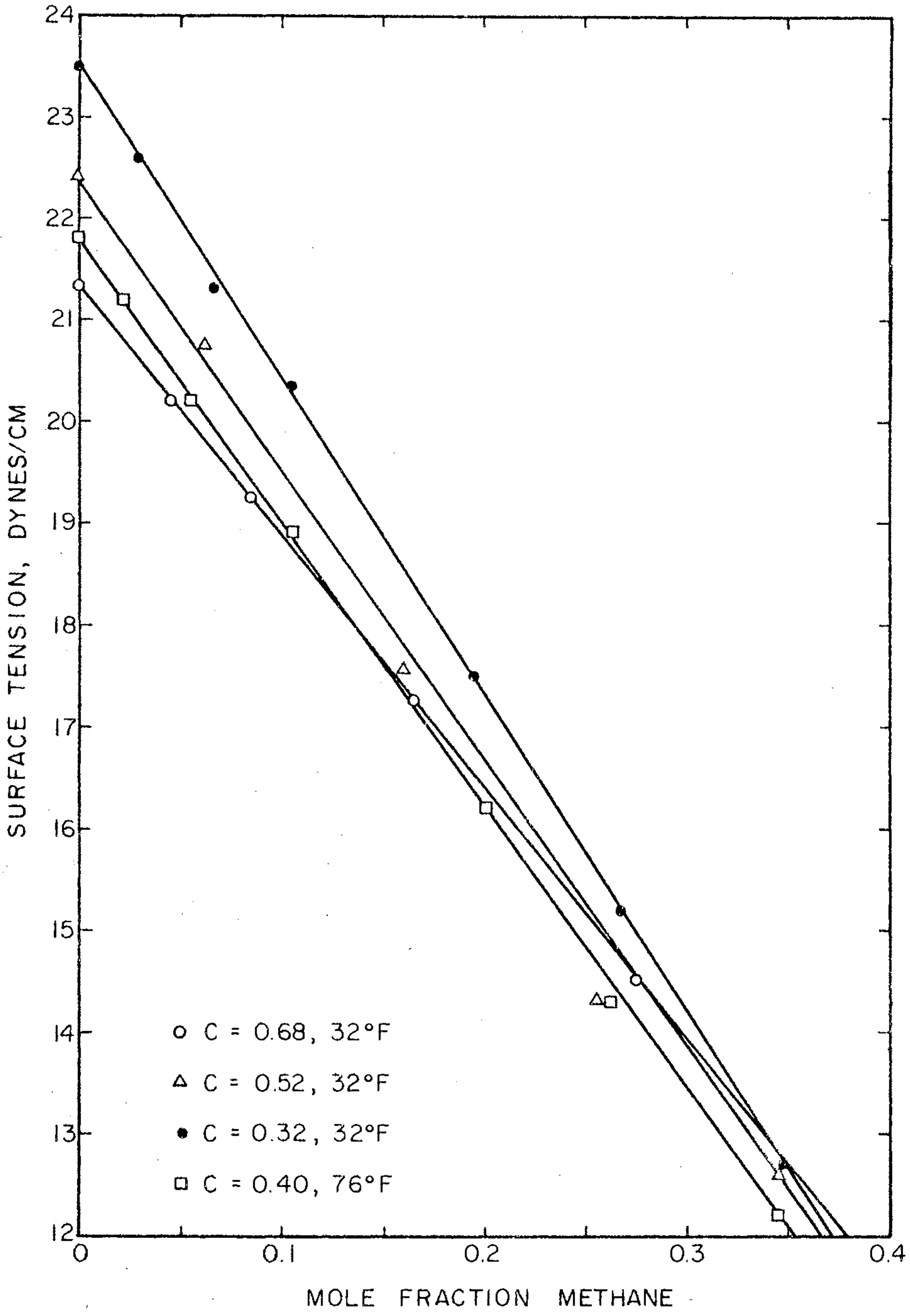

Figure 11. Surface Tension of Methane-Carbon Dioxide-Decane System Versus Methane Concentration

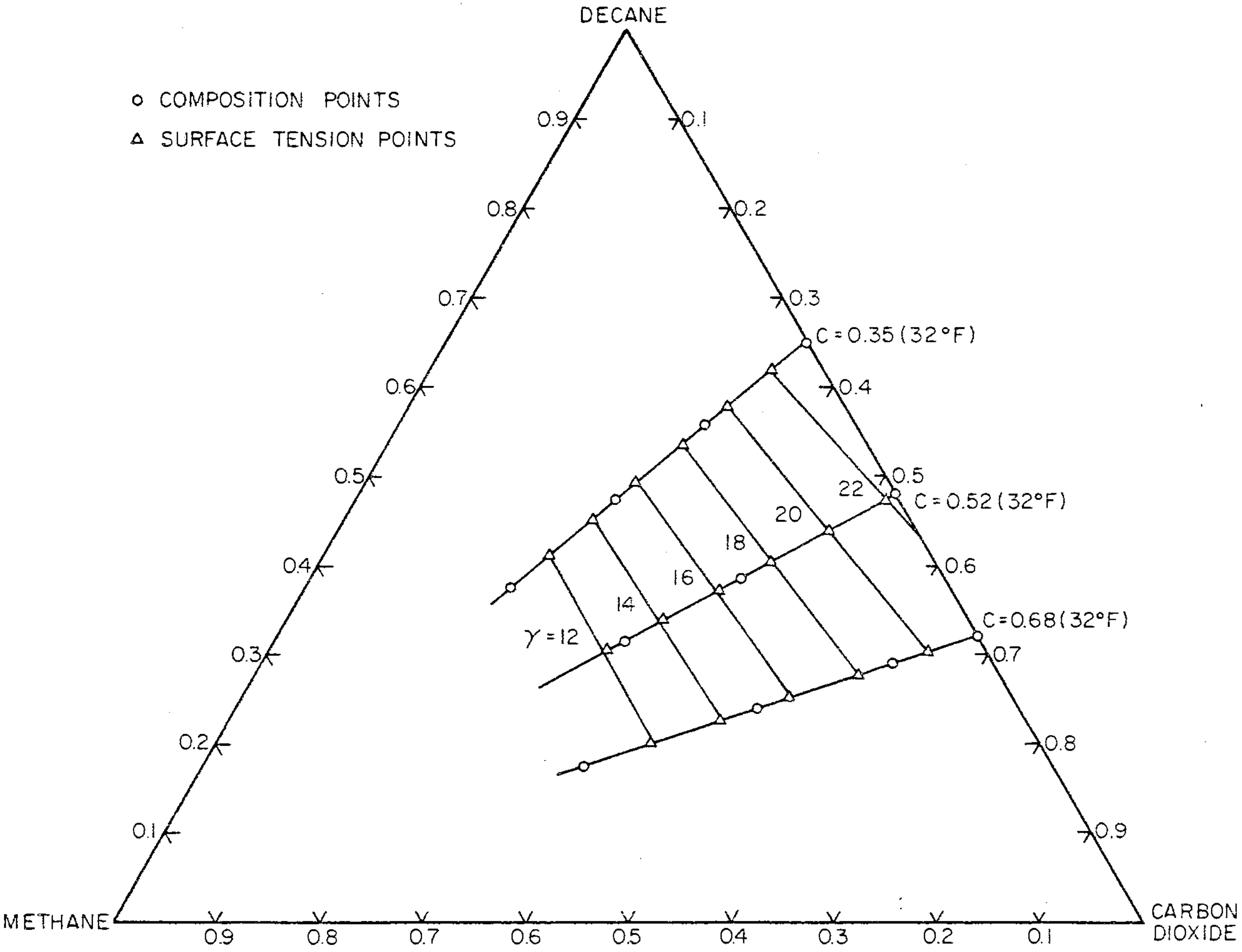

Figure 12. Phase Diagram for Methane-Carbon Dioxide-Decane Surface Tension

surface tension values by use of Equation (6). Three composition parameters were studied at 32°F, .35, .52, and 68; and one composition parameter was investigated at 76°F, 0.40.

CHAPTER V

DISCUSSION OF EXPERIMENTAL RESULTS

Reliability of Results

Initial experimental work, after the familiarization stage, was concerned with verifying the results of this work. Therefore, surface tension of the methane-nonane binary system at 76°F was determined in order to compare these results with values reported by Deam (7) for the same system. The experimental data of this work are compared to Deam's data in Table VIII. The spread around the average value of this work is approximately ± 1 percent. Agreement of the data of this work with Deam's is rather good. Deviation of the averages of this work from Deam's averages is less than ± 1 percent.

A complete error analysis of experimental surface tension values is presented in Appendix A. The limiting accuracy of the experimental data seems to be about 2 percent. This takes into consideration the probable errors in measuring the two selected diameters, d_e and d_s, which are the main contributors to experimental error. Using the Vanguard motion analyzer, the images of the drops were measured to 0.001 inches. However any measurement errors are magnified about five fold, mainly through the

TABLE VIII

RELIABILITY OF EXPERIMENTAL DATA

Methane-Nonane System at 76°F

Press. psia	Surface Tension, dynes/cm.		
	This Work	Ave.Value	Deam
75	21.62	21.79	21.77
	21.64		
	21.82		
	21.99		
	21.88		
150	20.56	20.52	20.58
	20.55		
	20.52		
	20.38		
	20.41		
	20.81		
	20.40		
	20.75		
	20.53		
	20.36		
	20.49		

relationship of 1/H to S.

Phase Rule Interpretation

Surface tension is an intensive property of a system; i.e., it is independent of the mass of the system, but is fully dependent on the thermodynamic state of the system. Therefore, it is important to analyze and keep in perspective the number of independent degrees of freedom, or variables, that must be specified for a system under consideration to be fixed. The phase rule states this as follows

$$F + \phi = N + 2 \qquad (12)$$

where F is the number of degrees of freedom, or independent variables, ϕ is the number of phases, and N is the number of components.

Thus for a binary system, in order for the state of equilibrium to be completely defined, two variables, normally temperature and pressure, must be fixed. This fixes all intensive properties of the system; e.g., equilibrium composition of the two phases and the surface tension. Therefore, interpolation between data points in the two-phase region is feasible.

For a ternary system, on the other hand, the situation is very different. The phase rule requires, for a ternary system in the two-phase region, that three variables be specified in order for the system to be fixed. In other words, specifying the temperature and pressure alone is no longer sufficient; a third variable must be specified in order to define the state of equilibrium and to allow correlating the data. Reamer et al. (24) defined a composition parameter expressed as

$$C = \frac{n_2}{n_2 + n_3} \tag{13}$$

where n_2 and n_3 refer to the mole fraction of the intermediate and heavy components, respectively, in the mixture as a whole. This facilitates considerably the graphical presentation of ternary data. Direct interpolation of surface tension with respect to the light component concentration is also possible.

For a ternary system, a measured amount of liquid, n_3, was charged to the system; and with the capillary valve in the closed position, the cell was pressured with the intermediate component to a predetermined pressure. With thermal equilibrium established, this allowed calculation of n_2. To charge the light component, the capillary valve was closed and the cell was pressured. This allowed calculation of n_1 once thermal equilibrium was achieved.

Methane-Nonane Experimental Results

Vapor-liquid equilibrium data for the methane-nonane system were obtained from a combination of correlations. The NGPA K and H computer program was used to obtain phase compositions. The program uses the Chao-Seader (6) technique for the calculation. The vapor density was also obtained from the program; for this calculation the program uses the compressibility factor computed by the Redlich-Kwong equation of state. The liquid phase density was calculated by the Rackett technique. This method is discussed in detail by Deam (7) and predicts saturated liquid phase densities to ± 2 per cent.

Figure 5 shows that surface tension of the system decreases with increasing pressure, which is equivalent to increasing the methane concentration. Experimental data of Deam at 76°F are shown in Figure 5 for comparison purposes.

Ethane-Nonane Experimental Results

Literature data on phase compositions and densities of the ethane-nonane system were not available. Phase compositions and vapor phase densities were calculated by the NGPA program. Liquid phase densities were calculated by the Rackett procedure.

Figure 6 shows that surface tension decreases as the pressure increases. Furthermore, surface tension of the system decreases with increasing temperatures - below the critical temperature of ethane. However, the effect of pressure on surface tension diminishes above the critical temperature of ethane.

Carbon Dioxide-Decane Experimental Results

Vapor-liquid equilibrium data of Reamer et al. (22)

for the carbon dioxide-decane system was utilized at 40°F and 160°F. Some interpolation was necessary at 40°F and 75 psia. No experimental data was reported at 0°F; the NGPA program was, therefore, used for phase compositions and vapor phase densities. Liquid densities were calculated by the Rackett Method.

As the pressure increases, surface tension is seen in Figure 7 to decrease. Crossovers are observed in the diagram; this is due in part to the pressure effect and in part to the fact that the solubility of carbon dioxide increases at lower temperatures, tending to lower the surface tension values. Like the ethane-nonane system, the carbon dioxide-decane system is less sensitive to pressure changes above the critical temperature of carbon dioxide than below it.

Hydrogen Sulfide-Decane Experimental Data

Equilibrium phase data of Sage and coworkers (26) was used. Interpolation was required in some cases. Vapor phase densities are not given in the range investigated in this work; these were determined, therefore, by the NGPA program. The same program was used at 160°F and 50 and 75 psia, which could not be interpolated for phase compositions. Liquid phase densities for these two points were determined by the Rackett technique.

Surface tension is seen to decrease as the pressure increases. Also, surface tension decreases as the temperature increases.

Ethane-Butane-Decane Experimental Results

No experimental phase composition data were available for the ethane-butane-decane ternary system in the literature. The NGPA program was used to determine phase compositions and vapor phase densities. Liquid phase density was then determined by the Rackett equation.

One isotherm was determined for this system. This was at 170°F, above the critical temperature of ethane. The effect of pressure on surface tension is pronounced; as pressure is increased, surface tension decreased - linearly.

Methane-Carbon Dioxide-Decane Experimental Results

Equilibrium vapor-liquid data for the methane-carbon dioxide-decane ternary system were not available. In order to determine phase compositions, the NGPA program was used. Liquid densities were determined by the Rackett procedure.

Surface tension was measured along three composition parameters, 0.35, 0.52, and 0.68, at 32°F, and along one composition parameter, 0.40, at 76°F. Pressure range covered was from about 30 psia to approximately 1000 psia.

As seen from Figures 10 and 11 surface tension decreased as methane pressure was increased. Also, surface tension decreased as the carbon dioxide fraction, composition parameter, was increased. The four isothermal lines are seen to be linear and approximately parallel.

Figure 12 may be used to estimate surface tension for this ternary in the outlined area.

CHAPTER VI

EXCESS SURFACE TENSION AND PARACHOR APPLICABILITY

Correlation of experimental data was attempted by two independent techniques. The excess surface tension function proposed by Deam (7) applies a correction to pure component surface tension, calculated by the Ferguson equation, when the mixture temperature is above the critical temperature of the gas; this is shown in Equation (11). The Katz method, Equation (10), employs the parachor, a rather constant quantity for every component, to predict surface tension of a mixture. Both methods were applied to the different systems investigated in this study. Values of the constants used in the correlations are presented in Appendix B.

The Katz relationship was used to correlate the experimental methane-nonane surface tension data. Katz' predictions show a positive bias with a maximum error of 12.87 percent and an average error of 8.44 percent. Deam's predictions show a maximum positive error of 7.27 percent, a maximum negative error of 3.20 percent, and an average absolute deviation of 3.08 percent. Table IX shows a comparison of the experimental data with values calculated

TABLE IX

COMPARISON OF PARACHOR AND EXCESS SURFACE TENSION CORRELATIONS WITH EXPERIMENTAL METHANE-NONANE DATA AT 70°F

Temp. ^{o}F	Press. psia	Surface Tension Ave.Exp.	Surface Tension Katz	Surface Tension Deam	Per Cent Deviation Katz	Per Cent Deviation Deam
70	50	22.05	24.89	22.05	12.87	-0.01
	100	21.70	24.11	21.05	11.10	-3.01
	300	19.57	21.19	18.94	8.29	-3.20
	500	17.14	18.54	17.14	8.15	-0.02
	750	14.51	15.54	15.23	7.11	4.96
	1000	12.51	12.90	13.42	3.13	7.27

by the parachor and excess surface tension methods.

When applied to the ethane-nonane system, the Katz equation produced a positive bias, with a maximum deviation of 129.57 percent. The average deviation over the entire temperature and pressure range was 45.18 percent; above the critical temperature of ethane, however, the average error was 14.27 percent. Deam's equation gave a maximum positive error 81.86 percent, a maximum negative error of 4.64 percent. The overall, average absolute deviation for Deam's correlation was 13.29 percent; but above the ethane critical temperature the average absolute deviation was only 2.70 percent. A comparison of experimental and calculated values is presented in Table X.

The carbon dioxide-decane system is probably most nonideal, and this is reflected in higher average deviations as shown in Table XI. The parachor correlation gave a maximum positive error of 113.67 percent, a maximum negative error of 10.25 percent, and an average absolute error of 28.10 percent - over the temperature and pressure range investigated. Above the critical temperature of carbon dioxide, the average error was 7.79 percent. Deam's correlation produced a maximum positive error of 76.84 percent, a maximum negative error of 12.71 percent and an average absolute error of 18.54 percent; but the average error was 19.21 percent above the critical temperature of carbon dioxide. Correlation of this system exhibits quite significant deviations from experimental

TABLE X

COMPARISON OF PARACHOR AND EXCESS SURFACE TENSION CORRELATIONS WITH EXPERIMENTAL ETHANE-NONANE DATA

Temp. °F	Press. psia	Surface Tension			Per Cent Deviation	
		Ave.Exp.	Katz	Deam	Katz	Deam
0	50	21.34	25.80	21.60	20.89	0.78
	100	16.47	21.57	16.95	30.97	1.73
	200	10.33	20.23	15.94	95.88	52.23
32	50	22.24	24.90	21.25	11.97	-4.64
	100	18.53	22.05	18.12	19.00	-2.64
	200	12.71	18.00	13.94	41.60	8.60
	300	7.59	17.42	13.94	129.57	81.86
58	320	8.12	11.13	7.68	37.09	-6.74
130	320	11.39	13.59	11.45	19.29	-3.10
170	75	15.86	18.08	16.17	14.02	1.47
	150	14.82	16.52	14.87	11.45	-0.69
	300	12.28	13.48	12.40	9.74	-1.37
	500	7.83	9.54	8.78	21.88	6.89

TABLE XI

COMPARISON OF PARACHOR AND EXCESS SURFACE TENSION CORRELATIONS WITH EXPERIMENTAL CARBON DIOXIDE-DECANE DATA

Temp. °F	Press. psia	Surface Tension			Per Cent Deviation	
		Ave.Exp.	Katz	Deam	Katz	Deam
0	50	27.31	48.55	25.36	77.78	-7.15
	100	25.01	37.26	23.07	48.98	-7.75
	300	9.89	21.13	18.10	113.67	82.99
32	34	23.50	23.76	24.07	1.11	2.41
	50	22.73	22.56	23.46	-0.75	3.22
	100	21.34	19.01	21.58	-10.93	1.12
40	75	22.43	24.68	23.49	10.02	4.72
	150	20.73	22.24	22.50	7.31	8.55
	300	16.51	18.16	18.75	9.96	13.57
	500	9.04	8.11	15.90	-10.25	75.91
76	34	21.78	22.27	22.33	2.27	2.54
160	200	15.77	16.96	14.91	7.54	-5.46
	400	13.40	14.99	11.01	11.85	-17.84
	600	11.82	12.83	7.76	8.50	-34.31
	800	10.24	10.58	5.20	3.28	-49.25
170	50	14.10	15.04	13.92	6.65	-1.30

values. The untolerable errors are observed at higher pressures of every isotherm where carbon dioxide concentration in the liquid phase exceeds about 40 percent. For the 0°F isotherm, critical conditions were exhibited as soon as the pressure was raised beyond 300 psia; i.e., the liquid phase was lost. Critical conditions were observed around 550 psia for the 40°F isotherm. Therefore the large errors found at the three extreme pressures for this system have critical contributions as well as contributions due to the high concentration of carbon dioxide.

Table XII compares the predictions of the parachor correlation to experimental hydrogen sulfide-decane data. A positive bias is exhibited by the data. Maximum deviation is 17.66 percent; and average deviation is 6.70 percent. Deam's equation was not applied to this set of data. Ferguson equation constants for hydrogen sulfide are not available, and pure hydrogen sulfide surface tension data is not available to determine these constants. These data, however, are sub-critical; and therefore, provide no test for the excess surface tension function proposed by Deam.

The comparison of results for the ethane-butane-decane ternary system is presented in Table XIII. This is well above the critical temperature of ethane. The parachor equation again shows a positive bias with a maximum deviation of 17.77 percent and an average deviation of 14.01

TABLE XII

COMPARISON OF PARACHOR PREDICTIONS WITH HYDROGEN SULFIDE-DECANE DATA

Temp. °F	Press. psia	Surface Tension Ave. Exp.	Surface Tension Katz	Per Cent Deviation
40	50	21.60	24.13	11.71
	100	20.07	21.53	7.28
	115	19.56	20.66	5.64
100	30	20.40	20.85	2.18
	50	19.87	21.39	7.67
	100	18.82	20.22	7.44
	115	18.49	21.76	17.66
160	50	18.06	18.34	1.53
	75	17.61	17.81	1.14
	100	17.21	18.02	4.73

TABLE XIII

COMPARISON OF PARACHOR AND EXCESS SURFACE TENSION CORRELATIONS WITH ETHANE-BUTANE-DECANE DATA, C=0.38

Temp. °F	Press. psia	Surface Tension			Per Cent Deviation	
		Ave.Exp.	Katz	Deam	Katz	Deam
170	50	14.10	15.04	13.92	6.65	-1.30
	100	13.22	14.96	14.19	13.13	6.82
	200	11.99	13.48	12.96	12.46	6.63
	300	10.15	11.83	11.29	16.51	8.67
	400	8.65	10.17	9.65	17.54	7.70
	500	7.21	8.49	8.70	17.77	15.29

percent. The excess surface tension function gave an average absolute deviation of 7.74 percent, a maximum positive deviation of 15.29 percent, and a maximum negative deviation of 1.30 percent.

For the methane-carbon dioxide-decane system, the two correlations are compared to experimental data in Table XIV. For the four composition parameters studied, the parachor equation yielded an average absolute error of only 4.37 percent, which is quite good, a maximum positive error of 3.08 percent, and a maximum negative error of 12.72 percent. The Deam equation averaged a mere 2.93 percent deviation, with a maximum positive error of 11.74 percent, and a maximum negative error of 4.03 percent Deam's predictions gave, on the average, positive bias.

TABLE XIV

COMPARISON OF PARACHOR AND EXCESS SURFACE TENSION CORRELATIONS WITH EXPERIMENTAL METHANE-CARBON DIOXIDE-DECANE DATA

Composition	Temp.	Press.	Surface Tension			Per Cent Deviation	
Parameter	°F	psia	Ave. Exp.	Katz	Deam	Katz	Deam
.40	76	100	21.20	21.27	21.30	0.32	0.45
		200	20.17	19.99	20.49	-0.89	1.60
		300	18.86	18.75	19.29	-0.56	2.28
		500	16.24	16.46	17.49	1.32	7.70
		750	14.26	13.84	15.11	-2.93	5.99
		1000	12.22	11.56	13.65	-5.43	11.74
.35	32	100	22.59	23.29	22.82	3.08	1.00
		200	21.33	21.47	21.87	0.65	2.52
		300	20.35	20.05	19.53	-1.49	-4.03
		500	17.52	17.38	17.87	-0.82	1.99
		750	15.20	14.36	15.22	-5.55	0.14
		1000	12.74	11.67	13.02	-8.37	2.16
.52	32	200	20.76	20.80	21.20	0.18	2.12
		500	17.57	16.88	17.82	-3.93	1.44
		750	14.32	13.69	15.11	-2.54	5.52
		1000	12.60	11.36	13.03	-9.82	3.44
.68	32	200	20.16	18.51	20.46	-8.17	1.48
		300	19.25	17.38	19.76	-9.69	2.65
		500	17.11	15.21	17.37	-11.13	1.55
		750	14.52	12.67	14.76	-12.72	1.63

CHAPTER VII

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

This project set out to experimentally measure surface tension, by the pendant drop method, of a variety of hydrocarbon and related systems. Surface tension was determined for methane-nonane, ethane-nonane, carbon dioxide-decane, and hydrogen sulfide-decane binary systems, and for ethane-butane-decane and methane-carbon dioxide-decane ternary systems. In general, a range of temperature and pressure was investigated for each system.

Applicability of the Katz parachor technique was tested. Predictions of the parachor procedure were compared to experimental results for all the systems investigated. This method proved only moderately successful in correlating surface tension of the mixtures studied here.

The excess surface tension concept proved to be efficient in correlating the experimental data. This function accounts for the behavior of the super-critial, volatile component in the mixture. The light component behaves like a dissolved gas in the liquid when used above its critical temperature. A diagram developed by Deam (7) to estimate surface tension of methane containing mixtures, on the basis of the dissolved gas behavior of methane, was successfull in predicting surface tension of other mixtures studied in this work.

Deam's diagram, discussed above, is reproduced in Figure 13. The data of this work are superimposed on the diagram. The agreement is quite good. This supports and reinforces the excess surface tension proposal as being generally applicable to a variety of systems over a wide range of conditions as long as they are reasonably removed from their critical region. Only the carbon dioxide-decane system diverges considerably on the diagram.

Recommendations

There is a range of temperature and pressure over which the unpredictable critical effects of the light component in a mixture are exhibited. At the present there is no method of estimating this range of conditions such that one feels assured he is operating outside this region. A systematic method of estimating this critical range should be a welcome contribution.

A reliable method for obtaining experimental liquid densities is needed and deserves attention in future studies. While the Rackett technique is powerful, it nevertheless, requires accurate phase composition information.

So far the excess surface tension function has been

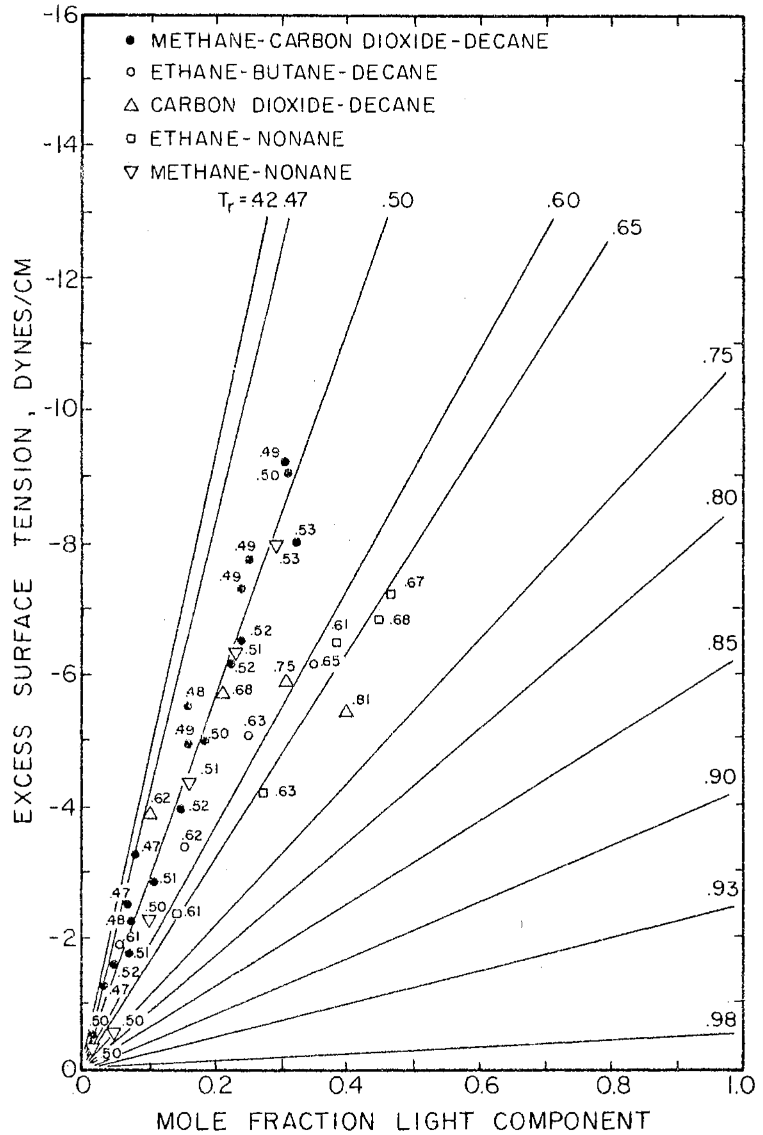

Figure 13. Deam's Excess Surface Tension as a Function of Light Component Concentration and Reduced Temperature

applied to ternary systems with only one component being super-critical. A more severe test would be a ternary mixture with two super-critical components; such systems could include, for example, methane-nitrogen-decane, methane-ethane-decane, and methane-carbon dioxide-decane mixtures.

NOMENCLATURE

English Letters

b = radius of curvature for a drop at its apex

C = composition parameter

d = density

d_e = equatorial diameter of a drop

d_n = magnified diameter of tip

d_s = selected diameter

g = acceleration of gravity

H = shape dependent parameter

M = molecular weight

n = exponent in van der Waals and Ferguson equations

P = pressure

pressure difference across a curved interface

p = parachor

R_1 and R_2 = radii of curvature for a drop

S = experimentally measureable shapte factor

T = absolute temperature

V = volume

x = mole fraction in liquid phase

Y = mole fraction in vapor phase

z = vertical co-ordinate measured from bottom of drop

NOMENCLATURE (Continued)

z = compressibility factor

Greek Letters

β = drop shape factor

γ = surface tension

γ^E = excess surface tension

γ_o = van der Waals equation constant

d = density difference between liquid and vapor phases

Subscripts

c = critical property

i = component number

L = liquid phase

m = mixture property

r = reduced property

V = vapor phase

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APPENDIX A

ERROR ANALYSIS

ERROR ANALYSIS

Deam (7) presented a detailed error analysis for surface tension values determined by the pendant drop method. Calculus is utilized to evaluate the error resulting rom the different, individual quantities used in the calculation. In the equation

$$\gamma = \frac{g\ (\Delta d)\ d_e^{\ 2}}{H} \tag{14}$$

error in surface tension values can result from errors in the gravitational constant, density difference, equatorial diameter, and the selected plane diameter.

The error generated by error in the gravitational constant is

$$\gamma_g = \left(\frac{\partial\gamma}{\partial g}\right)\ \delta g = \left(\frac{\gamma}{g}\right)\ \delta g \tag{15}$$

where δg is the error in the gravitational constant.

Errors in the density values used affect the calculated surface tension in the following manner

$$\gamma_{\ \Delta d} = \left(\frac{\partial\gamma}{\partial\Delta d}\right)\ \delta\ (\Delta d) = \left(\frac{\gamma}{\Delta d}\right)\ \delta\ (\Delta d). \tag{16}$$

The equatorial diameter, d_e, appears in two terms

$$\gamma_{d_e} = \left(\frac{\partial\gamma}{\partial d_e}\right)\ \delta\ (d_e) = \left[\ \frac{g(\Delta d)}{H}\quad 2\ d_e + g(\Delta d)\ d_e^{\ 2}\ \frac{\partial\ 1/H}{\partial\ d_e}\right]\ \delta\ (d_e) \tag{17}$$

but

$$1/H = f\ (S)\ = f\ (d_s/d_e). \tag{18}$$

Therefore,

$$\frac{\partial 1/H}{\partial d_e} = - \frac{d_s}{d_e^{\,2}} \quad \frac{\partial 1/H}{\partial S} \tag{19}$$

Stegemeier (6) reported that

$$\frac{\partial 1/H}{\partial S} = - 2.6444 \ (1/H) \ \frac{1}{S} \tag{20}$$

Thus, from Equations (17), (19), and (20)

$$\gamma_{d_e} = 4.6444 \ (\gamma / \ d_e) \ \ \delta d_e . \tag{21}$$

The effect of error in the selected plane diameter is

$$\gamma_{d_s} = g \ (\Delta d) \ d_e^{\,2} \quad \frac{\partial 1/H}{\partial d_s} . \tag{22}$$

Considering Equation (20), the error in surface tension as a result of error in d_s is found to be

$$\gamma_{d_s} = - \ 2.6444 \ \ (\frac{\gamma}{d_s}) \ \ \delta d_s . \tag{23}$$

Then, the expected error in surface tension may be written as

$$\delta \gamma = \sqrt{\gamma_g^{\,2} + \gamma_{\Delta d}^{\,2} + \gamma_{d_e}^{\,2} + \gamma_{d_s}^{\,2}} . \tag{24}$$

Typical data for the ethane-nonane system are as follows

d_e = 0.218 cm.

d_s = 0.192 cm

Δd = 0.6526 gm/ml.

γ = 12.54 dynes/cm

$\delta g = 0.$

$\delta \Delta d = 0.005$ gm/ml

$\delta d_e = 0.001$ cm.

$\delta d_s = 0.001$ cm.

From Equation (24) the probable error in the reported surface tension values is 0.33 dynes/cm., or

$\gamma = 12.54 \pm 0.33$ dynes/cm.

APPENDIX B

DATA USED WITH CORRELATIONS

TABLE XV

CRITICAL CONSTANTS (20)

Component	Z_c	T_c, °R	V_c, ml/g mole
Methane	.289	344.0	99.0
Ethane	.279	554.4	146.3
propane	.276	665.3	199.6
i-Butane	.275	734.6	256.0
n-Butane	.273	765.4	254.0
i-Pentane	.270	829.6	310.0
n-Pentane	.268	846.5	311.0
n-Hexane	.264	914.2	368.0
Cyclohexane	.272	995.8	309.0
n-Heptane	.261	972.4	428.0
n-Octane	.257	1024.9	488.0
n-Nonane	.254	1071.0	552.0
n-Decane	.251	1114.2	614.0
Ethylene	.282	508.9	131.0
Propylene	.278	654.7	182.2
1-Butene	.274	755.3	238.0
2-Butene	.276	770.4	236.0
i-Butene	.276	752.4	240.0
1,3-Butadiene	.271	765.0	221.0
Nitrogen	.289	227.2	89.5
Carbon Dioxide	.274	546.3	93.2
Hydrogen Sulfide	.285	672.5	89.3

TABLE XVI

PARACHOR VALUES USED (17)

Component	Parachor
Methane	71.0
Ethane	111.0
n-Butane	191.0
n-Nonane	391.0
n-Decane	431.0
Carbon Dioxide	48.6
Hydrogen Sulfide	80.1

TABLE XVII

FERGUSON EQUATION CONSTANTS

Component	γ_o	n	Source of Data
Methane	39.05	1.221	(7)
Ethane	50.095	1.26	*
Propane	49.90	1.20	(7)
Butane	52.50	1.22	(7)
Pentane	52.90	1.22	(3)
Heptane	47.27	1.099	(7)
Nonane	51.60	1.22	(3)
Decane	51.60	1.22	(3)
Ethylene	51.80	1.25	(7)
Nitrogen	28.42	1.232	(7)
Carbon Dioxide	79.621	1.248	**

* Pure ethane surface tension from API Data Book (3) were used to determine Ferguson equation constants for ethane.

** Carbon dioxide surface tension data reported by Quinn(19) were used to evaluate Ferguson equation constants for carbon dioxide.

APPENDIX C

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS OF EXPERIMENTAL RUNS

TABLE XVIII

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS FOR METHANE-NONANE RUNS

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
70	100	22.483	4.770	5.789	6.747	0.7305	0.0046
70	100	22.659	4.770	5.787	6.757	0.7305	0.0046
70	100	22.752	4.770	5.763	6.747	0.7305	0.0046
70	100	22.581	4.770	5.784	6.750	0.7305	0.0046
70	100	22.653	4.770	5.777	6.750	0.7305	0.0046
70	100	22.622	4.770	5.777	6.748	0.7305	0.0046
70	100	22.252	4.770	5.792	6.734	0.7305	0.0046
70	300	20.307	4.770	5.638	6.545	0.7183	0.0142
70	300	20.342	4.770	5.648	6.554	0.7183	0.0142
70	300	20.302	4.770	5.631	6.540	0.7183	0.0142
70	500	17.643	4.770	5.509	6.309	0.7067	0.0243
70	500	18.113	4.770	5.494	6.335	0.7067	0.0243
70	500	17.729	4.770	5.505	6.313	0.7067	0.0243
70	500	17.918	4.770	5.515	6.334	0.7067	0.0243
70	500	17.737	4.770	5.524	6.326	0.7067	0.0243
70	500	17.601	4.770	5.494	6.296	0.7067	0.0243
70	500	17.747	4.770	5.506	6.315	0.7067	0.0243
70	500	17.440	4.770	5.507	6.292	0.7067	0.0243
70	500	17.459	4.770	5.514	6.298	0.7067	0.0243
70	750	14.808	4.770	5.318	6.007	0.6928	0.0376
70	750	14.798	4.770	5.349	6.026	0.6928	0.0376
70	750	14.979	4.770	5.309	6.016	0.6928	0.0376
70	750	15.000	4.770	5.331	6.032	0.6928	0.0376
70	750	14.931	4.770	5.320	6.019	0.6928	0.0376
70	750	14.943	4.770	5.337	6.031	0.6928	0.0376
70	750	14.977	4.770	5.303	6.012	0.6928	0.0376
70	750	14.926	4.770	5.344	6.034	0.6928	0.0376
70	750	14.939	4.770	5.336	6.030	0.6928	0.0376
70	750	14.943	4.770	5.320	6.020	0.6928	0.0376
70	750	14.918	4.770	5.342	6.032	0.6928	0.0376
70	750	14.938	4.770	5.344	6.035	0.6928	0.0376
70	750	14.949	4.770	5.313	6.016	0.6928	0.0376
70	750	14.972	4.770	5.327	6.027	0.6928	0.0376
70	1000	12.902	4.770	5.164	5.787	0.6798	0.0517
70	1000	12.869	4.770	5.191	5.801	0.6798	0.0517
70	1000	13.041	4.770	5.151	5.792	0.6798	0.0517
70	1000	12.998	4.770	5.174	5.793	0.6798	0.0517
70	1000	12.974	4.770	5.186	5.808	0.6798	0.0517
76	75	21.620	4.886	5.862	6.850	0.7091	0.0035
76	75	21.640	4.886	5.890	6.870	0.7091	0.0035
76	75	21.821	4.886	5.837	6.847	0.7091	0.0035
76	75	21.989	4.886	5.847	6.865	0.7091	0.0035
76	75	21.876	4.886	5.827	6.844	0.7091	0.0035
76	150	20.562	4.886	5.703	6.683	0.7065	0.0067
76	150	20.548	4.886	5.831	6.767	0.7065	0.0067
76	150	20.524	4.886	5.725	6.695	0.7065	0.0067
76	150	20.383	4.886	5.632	6.623	0.7065	0.0067
76	150	20.413	4.980	5.747	6.757	0.7065	0.0067
76	150	20.811	4.980	5.714	6.763	0.7065	0.0067

TABLE XVIII (Continued)

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
76	150	20.402	4.980	5.778	6.777	0.7065	0.0067
76	150	20.750	4.980	5.710	6.756	0.7065	0.0067
76	150	20.531	4.980	5.727	6.752	0.7065	0.0067
76	150	20.356	4.980	5.756	6.759	0.7065	0.0067
76	150	20.492	4.980	5.881	6.852	0.7065	0.0067

TABLE XIX

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS FOR ETHANE-NONANE RUNS

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
0	50	21.412	4.429	5.223	6.090	0.7358	0.0051
0	50	21.570	4.429	5.207	6.089	0.7358	0.0051
0	50	21.440	4.429	5.240	6.103	0.7358	0.0051
0	50	20.948	4.429	5.215	6.056	0.7358	0.0051
0	100	16.828	4.429	4.952	5.687	0.6962	0.0107
0	100	16.290	4.429	4.890	5.607	0.6962	0.0107
0	100	16.281	4.429	4.865	5.590	0.6962	0.0107
0	200	10.327	4.469	4.428	4.844	0.6860	0.0147
32	50	22.145	4.291	5.030	5.928	0.7324	0.0048
32	50	22.178	4.291	5.045	5.940	0.7324	0.0048
32	50	22.150	4.291	5.040	5.935	0.7324	0.0048
32	50	22.292	4.291	5.010	5.923	0.7324	0.0048
32	50	22.190	4.291	5.044	5.940	0.7324	0.0048
32	50	22.500	4.291	5.025	5.945	0.7324	0.0048
32	100	18.819	4.291	4.899	5.687	0.7090	0.0098
32	100	18.595	4.291	4.941	5.700	0.7090	0.0098
32	100	18.565	4.291	4.947	5.702	0.7090	0.0098
32	100	18.370	4.291	4.886	5.649	0.7090	0.0098
32	100	18.417	4.291	4.845	5.625	0.7090	0.0098
32	100	18.419	4.291	4.869	5.641	0.7090	0.0098
32	200	12.643	4.291	4.632	5.132	0.6710	0.0184
32	200	12.543	4.291	4.638	5.127	0.6710	0.0184
32	200	13.008	4.291	4.617	5.154	0.6710	0.0184
32	200	12.592	4.291	4.620	5.120	0.6710	0.0184
32	200	12.831	4.291	4.611	5.135	0.6710	0.0184
32	200	12.603	4.291	4.491	5.039	0.6710	0.0184
32	200	12.541	4.291	4.504	5.042	0.6710	0.0184
32	200	13.031	4.291	4.492	5.076	0.6710	0.0184
32	200	12.788	4.291	4.510	5.067	0.6710	0.0184
32	300	12.520	4.291	4.467	5.024	0.6710	0.0229
32	300	7.662	4.291	3.858	4.158	0.6710	0.0229
32	300	7.529	4.291	4.128	4.305	0.6710	0.0229
32	300	7.584	4.291	4.038	4.258	0.6710	0.0229
58	320	8.329	4.762	4.879	5.225	0.5945	0.0355
58	320	8.017	4.762	4.839	5.158	0.5945	0.0355
58	320	7.872	4.762	4.856	5.148	0.5945	0.0355
58	320	8.259	5.065	5.003	5.433	0.5945	0.0355
58	320	8.228	5.065	5.033	5.447	0.5945	0.0355
58	320	8.267	5.065	4.836	5.330	0.5945	0.0355
58	320	7.896	5.065	4.940	5.342	0.5945	0.0355
58	320	8.326	5.065	4.777	5.301	0.5945	0.0355
58	320	8.015	5.065	4.872	5.317	0.5945	0.0355
58	320	8.362	5.065	4.858	5.357	0.5945	0.0355
58	320	8.037	5.065	4.731	5.232	0.5945	0.0355
58	320	7.951	5.065	4.832	5.283	0.5945	0.0355
58	320	7.905	5.065	4.788	5.249	0.5945	0.0355
58	320	8.010	5.065	4.928	5.351	0.5945	0.0355
58	320	8.146	5.065	4.772	5.273	0.5945	0.0355

TABLE XIX (Continued)

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
58	320	8.204	5.065	4.866	5.340	0.5945	0.0355
58	320	8.382	5.065	4.876	5.371	0.5945	0.0355
58	320	8.156	5.065	4.800	5.292	0.5945	0.0355
58	320	8.022	5.065	4.755	5.245	0.5945	0.0355
130	320	11.449	4.840	5.229	5.736	0.6465	0.0285
130	320	11.554	4.840	5.076	5.651	0.6465	0.0285
130	320	11.374	4.840	5.065	5.625	0.6465	0.0285
130	320	11.102	4.840	5.075	5.602	0.6465	0.0285
130	320	11.404	4.840	5.060	5.625	0.6465	0.0285
130	320	11.257	4.840	5.039	5.596	0.6465	0.0285
130	320	11.154	4.840	5.205	5.689	0.6465	0.0285
130	320	11.574	4.840	5.057	5.641	0.6465	0.0285
130	320	11.604	4.840	5.112	5.679	0.6465	0.0285
170	75	16.124	5.279	5.906	6.741	0.6808	0.0058
170	75	15.798	5.279	5.836	6.666	0.6808	0.0058
170	75	15.868	5.279	5.811	6.656	0.6808	0.0058
170	75	15.752	5.279	5.801	6.639	0.6808	0.0058
170	75	15.814	5.279	5.748	6.610	0.6808	0.0058
170	75	16.032	5.279	5.819	6.676	0.6808	0.0058
170	75	15.748	5.279	5.754	6.608	0.6808	0.0058
170	75	15.731	5.279	5.787	6.628	0.6808	0.0058
170	150	15.082	5.279	5.697	6.545	0.6700	0.0117
170	150	14.809	5.279	5.715	6.531	0.6700	0.0117
170	150	14.648	5.279	5.717	6.517	0.6700	0.0117
170	150	14.732	5.279	5.697	6.512	0.6700	0.0117
170	150	14.831	5.279	5.724	6.539	0.6700	0.0117
170	150	15.167	5.279	5.694	6.551	0.6700	0.0117
170	150	14.763	5.279	5.737	6.541	0.6700	0.0117
170	150	14.535	5.279	5.726	6.512	0.6700	0.0117
170	150	14.843	5.279	5.730	6.544	0.6700	0.0117
170	300	12.223	5.279	5.547	6.237	0.6464	0.0245
170	300	12.288	5.279	5.614	6.287	0.6464	0.0245
170	300	12.258	5.279	5.530	6.230	0.6464	0.0245
170	300	12.208	5.279	5.484	6.195	0.6464	0.0245
170	300	12.410	5.279	5.509	6.233	0.6464	0.0245
170	500	7.671	5.279	5.136	5.515	0.6083	0.0445
170	500	7.809	5.279	5.214	5.584	0.6083	0.0445
170	500	7.886	5.279	5.221	5.600	0.6083	0.0445
170	500	7.947	5.279	5.113	5.543	0.6083	0.0445

TABLE XX

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS FOR CARBON DIOXIDE-DECANE RUNS

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
0	50	27.378	4.650	5.597	6.531	0.9010	0.0074
0	50	27.277	4.650	5.632	6.549	0.9010	0.0074
0	50	27.281	4.650	5.671	6.575	0.9010	0.0074
0	100	24.536	4.650	5.433	6.318	0.8786	0.0150
0	100	25.051	4.650	5.410	6.331	0.8786	0.0150
0	100	25.143	4.650	5.434	6.352	0.8786	0.0150
0	100	25.272	4.650	5.452	6.371	0.8786	0.0150
0	100	25.078	4.650	5.477	6.377	0.8786	0.0150
0	100	24.956	4.650	5.478	6.371	0.8786	0.0150
0	300	10.013	4.650	4.419	4.692	0.8478	0.0350
0	300	9.996	4.650	4.520	4.751	0.8478	0.0350
0	300	9.641	4.650	4.491	4.697	0.8478	0.0350
0	300	9.898	4.650	4.510	4.735	0.8478	0.0350
40	75	22.413	4.824	5.771	6.740	0.7478	0.0102
40	75	22.320	4.824	5.771	6.734	0.7478	0.0102
40	75	22.299	4.824	5.716	6.696	0.7478	0.0102
40	75	22.729	4.824	5.757	6.751	0.7478	0.0102
40	75	22.408	4.824	5.767	6.737	0.7478	0.0102
40	150	20.619	4.824	5.666	6.565	0.7518	0.0214
40	150	20.892	4.824	5.662	6.581	0.7518	0.0214
40	150	20.951	4.824	5.665	6.587	0.7518	0.0214
40	150	20.307	4.824	5.644	6.529	0.7518	0.0214
40	150	20.458	4.824	5.695	6.573	0.7518	0.0214
40	150	20.961	4.824	5.661	6.585	0.7518	0.0214
40	150	20.947	4.824	5.676	6.594	0.7518	0.0214
40	300	16.227	4.824	5.373	6.073	0.7641	0.0469
40	300	16.469	4.824	5.391	6.104	0.7641	0.0469
40	300	16.437	4.824	5.333	6.064	0.7641	0.0469
40	300	16.627	4.824	5.336	6.061	0.7641	0.0469
40	300	16.776	4.824	5.266	6.047	0.7641	0.0469
40	500	8.963	4.824	4.810	5.009	0.8146	0.0914
40	500	8.951	4.824	4.804	5.004	0.8146	0.0914
40	500	9.032	4.824	4.719	4.963	0.8146	0.0914
40	500	9.058	4.824	4.729	4.972	0.8146	0.0914
40	500	8.918	4.824	4.782	4.987	0.8146	0.0914
40	500	9.153	4.824	4.742	4.991	0.8146	0.0914
40	500	9.156	4.824	4.566	4.885	0.8146	0.0914
40	500	9.103	4.824	4.632	4.919	0.8146	0.0914
40	500	8.979	4.824	4.575	4.870	0.8146	0.0914
160	200	16.046	4.639	5.214	5.941	0.6957	0.0248
160	200	15.462	4.639	5.157	5.857	0.6957	0.0248
160	200	15.632	4.639	5.105	5.837	0.6957	0.0248
160	200	15.438	4.639	5.063	5.794	0.6957	0.0248
160	200	16.055	4.639	5.156	5.904	0.6957	0.0248
160	200	15.612	4.639	5.109	5.838	0.6957	0.0248
160	200	16.115	4.639	5.161	5.912	0.6957	0.0248
160	400	13.243	4.639	4.928	5.553	0.7023	0.0496
160	400	13.354	4.639	4.928	5.563	0.7023	0.0496

TABLE XX (Continued)

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
160	400	13.239	4.639	4.885	5.525	0.7023	0.0496
160	400	13.547	4.639	4.937	5.586	0.7023	0.0496
160	400	13.605	4.639	5.104	5.698	0.7023	0.0496
160	600	11.787	4.639	4.923	5.450	0.7065	0.0745
160	600	11.927	4.639	4.838	5.410	0.7065	0.0745
160	600	11.651	4.639	4.870	5.403	0.7065	0.0745
160	600	11.363	4.639	4.807	5.334	0.7065	0.0745
160	600	11.379	4.639	4.803	5.383	0.7065	0.0745
160	600	11.809	4.639	4.795	5.371	0.7065	0.0745
160	800	10.229	4.639	4.571	5.115	0.7116	0.1064
160	800	9.989	4.639	4.628	5.125	0.7116	0.1064
160	800	10.549	4.639	4.560	5.142	0.7116	0.1064
160	800	10.011	4.639	4.550	5.078	0.7116	0.1064
160	800	10.405	4.639	4.433	5.045	0.7116	0.1064

TABLE XXI

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS FOR HYDROGEN SULFIDE-DECANE RUNS

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
40	50	21.449	4.586	5.386	6.277	0.7449	0.0053
40	50	21.435	4.586	5.419	6.298	0.7449	0.0053
40	50	21.953	4.586	5.468	6.363	0.7449	0.0053
40	50	21.549	4.586	5.354	6.262	0.7449	0.0053
40	100	20.549	4.586	5.352	6.204	0.7466	0.0109
40	100	19.593	4.586	5.226	6.058	0.7466	0.0109
40	115	19.558	4.586	5.226	6.050	0.7501	0.0112
100	30	20.180	4.073	4.811	5.573	0.7103	0.0027
100	30	20.792	4.073	4.802	5.603	0.7103	0.0027
100	30	20.480	4.073	4.828	5.602	0.7103	0.0027
100	30	20.411	4.073	4.816	5.590	0.7103	0.0027
100	30	20.132	4.073	4.829	5.582	0.7103	0.0027
100	50	19.921	4.073	4.823	5.555	0.7184	0.0046
100	50	20.054	4.073	4.817	5.559	0.7184	0.0046
100	50	20.166	4.073	4.819	5.567	0.7184	0.0046
100	50	19.957	4.073	4.835	5.565	0.7184	0.0046
100	50	19.521	4.073	4.834	5.538	0.7184	0.0046
100	50	19.664	4.073	4.807	5.529	0.7184	0.0046
100	50	19.792	4.073	4.812	5.540	0.7184	0.0046
100	100	18.766	4.073	4.735	5.435	0.7184	0.0096
100	100	18.727	4.073	4.748	5.441	0.7184	0.0096
100	100	18.886	4.073	4.745	5.449	0.7184	0.0096
100	100	18.917	4.073	4.762	5.462	0.7184	0.0096
100	100	18.892	4.073	4.772	5.467	0.7184	0.0096
100	100	18.880	4.073	4.764	5.461	0.7184	0.0096
100	100	18.756	4.073	4.762	5.452	0.7184	0.0096
100	100	18.753	4.073	4.770	5.457	0.7184	0.0096
100	115	18.619	4.073	4.770	5.424	0.7349	0.0110
100	115	18.368	4.073	4.776	5.412	0.7349	0.0110
100	115	18.480	4.073	4.782	5.423	0.7349	0.0110
100	115	18.326	4.073	4.791	5.419	0.7349	0.0110
100	115	18.530	4.073	4.788	5.430	0.7349	0.0110
100	115	18.581	4.073	4.766	5.419	0.7349	0.0110
100	115	18.357	4.073	4.774	5.410	0.7349	0.0110
100	115	18.650	4.073	4.798	5.444	0.7349	0.0110
160	50	17.934	4.134	4.782	5.487	0.6896	0.0043
160	50	18.048	4.134	4.766	5.484	0.6896	0.0043
160	50	17.911	4.134	4.769	5.477	0.6896	0.0043
160	50	18.328	4.134	4.752	5.493	0.6896	0.0043
160	75	17.591	4.134	4.752	5.451	0.6880	0.0064
160	75	17.874	4.134	4.728	5.454	0.6880	0.0064
160	75	17.649	4.134	4.766	5.464	0.6880	0.0064
160	75	17.501	4.134	4.749	5.443	0.6880	0.0064
160	75	17.456	4.134	4.752	5.442	0.6880	0.0064
160	100	17.346	4.134	4.633	5.351	0.6936	0.0086
160	100	17.056	4.134	4.741	5.402	0.6936	0.0086
160	100	17.233	4.134	4.698	5.386	0.6936	0.0086

TABLE XXII

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS FOR ETHANE-BUTANE-DECANE RUNS

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
170	50	13.884	5.469	5.887	6.693	0.6466	0.0072
170	50	14.242	5.469	5.829	6.692	0.6466	0.0072
170	50	13.996	5.469	5.843	6.676	0.6466	0.0072
170	50	14.256	5.469	5.922	6.754	0.6466	0.0072
170	50	14.138	5.469	5.879	6.714	0.6466	0.0072
170	100	13.158	5.469	5.649	6.460	0.6509	0.0104
170	100	13.239	5.469	5.759	6.540	0.6509	0.0104
170	100	13.262	5.469	5.828	6.587	0.6509	0.0104
170	200	12.179	5.469	5.810	6.495	0.6410	0.0182
170	200	11.930	5.469	5.620	6.345	0.6410	0.0182
170	200	12.162	5.469	5.490	6.287	0.6410	0.0182
170	200	11.950	5.469	5.817	6.473	0.6410	0.0182
170	200	11.904	5.469	5.794	6.453	0.6410	0.0182
170	200	11.956	5.469	5.720	6.412	0.6410	0.0182
170	200	11.842	5.469	5.689	6.379	0.6410	0.0182
170	300	10.197	5.469	5.562	6.146	0.6277	0.0272
170	300	10.042	5.469	5.620	6.162	0.6277	0.0272
170	300	9.975	5.469	5.647	6.170	0.6277	0.0272
170	300	10.390	5.469	5.452	6.101	0.6277	0.0272
170	400	8.336	5.469	5.481	5.890	0.6122	0.0368
170	400	8.339	5.469	5.546	5.930	0.6122	0.0368
170	400	8.899	5.469	5.222	5.811	0.6122	0.0368
170	400	8.633	5.469	5.477	5.932	0.6122	0.0368
170	400	8.916	5.469	5.264	5.840	0.6122	0.0368
170	400	6.539	5.469	5.354	5.842	0.6122	0.0368
170	400	8.937	5.469	5.283	5.855	0.6122	0.0368
170	400	8.573	5.469	5.333	5.834	0.6122	0.0368
170	500	7.356	5.469	5.174	5.610	0.5941	0.0475
170	500	7.102	5.469	5.189	5.577	0.5941	0.0475
170	500	7.104	5.469	5.133	5.543	0.5941	0.0475
170	500	7.284	5.469	5.080	5.540	0.5941	0.0475
170	500	7.316	5.469	5.123	5.572	0.5941	0.0475
170	500	7.121	5.469	5.130	5.544	0.5941	0.0475
170	500	7.189	5.469	5.052	5.507	0.5941	0.0475

TABLE XXIII

SURFACE TENSION, DENSITY, AND DROP MEASUREMENTS FOR METHANE-CARBON DIOXIDE-DECANE RUNS

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
76	34	21.645	4.584	5.551	6.434	0.7243	0.0042
76	34	21.906	4.584	5.538	6.442	0.7243	0.0042
76	34	21.634	4.584	5.543	6.428	0.7243	0.0042
76	34	21.849	4.584	5.545	6.443	0.7243	0.0042
76	34	21.692	4.584	5.565	6.459	0.7243	0.0042
76	34	21.744	4.584	5.549	6.439	0.7243	0.0042
76	34	21.651	4.584	5.552	6.435	0.7243	0.0042
76	34	21.850	4.584	5.560	6.453	0.7243	0.0042
76	34	21.859	4.584	5.541	6.441	0.7243	0.0042
76	100	21.287	4.584	5.490	6.385	0.7202	0.0075
76	100	21.057	4.584	5.502	6.378	0.7202	0.0075
76	100	21.333	4.584	5.484	6.384	0.7202	0.0075
76	100	21.102	4.584	5.496	6.377	0.7202	0.0075
76	100	21.397	4.584	5.508	6.404	0.7202	0.0075
76	100	21.230	4.584	5.494	6.384	0.7202	0.0075
76	100	21.297	4.584	5.486	6.383	0.7202	0.0075
76	100	21.040	4.584	5.490	6.369	0.7202	0.0075
76	100	20.963	4.584	5.487	6.362	0.7202	0.0075
76	100	21.193	4.584	5.469	6.365	0.7202	0.0075
76	100	21.276	4.584	5.488	6.383	0.7202	0.0075
76	200	20.251	4.584	5.417	6.288	0.7148	0.0122
76	200	20.221	4.584	5.420	6.288	0.7148	0.0122
76	200	20.094	4.584	5.433	6.288	0.7148	0.0122
76	200	20.030	4.584	5.438	6.287	0.7148	0.0122
76	200	19.991	4.584	5.445	6.289	0.7148	0.0122
76	200	20.185	4.584	5.404	6.275	0.7148	0.0122
76	200	20.431	4.584	5.417	6.300	0.7148	0.0122
76	300	19.055	4.584	5.317	6.160	0.7094	0.0170
76	300	18.677	4.584	5.333	6.144	0.7094	0.0170
76	300	19.089	4.584	5.321	6.165	0.7094	0.0170
76	300	18.682	4.584	5.258	6.095	0.7094	0.0170
76	300	18.737	4.584	5.287	6.118	0.7094	0.0170
76	300	18.740	4.584	5.264	6.103	0.7094	0.0170
76	500	16.364	4.584	5.155	5.895	0.6991	0.0270
76	500	16.234	4.584	5.172	5.896	0.6991	0.0270
76	500	16.343	4.584	5.116	5.868	0.6991	0.0270
76	500	16.102	4.584	5.168	5.883	0.6991	0.0270
76	750	14.304	4.584	4.984	5.665	0.6866	0.0402
76	750	14.187	4.584	4.998	5.664	0.6866	0.0402
76	750	14.290	4.584	5.006	5.678	0.6866	0.0402
76	1000	12.217	4.584	4.923	5.485	0.6748	0.0539
32	34	23.374	4.414	5.400	6.307	0.7400	0.0046
32	34	23.570	4.414	5.401	6.319	0.7400	0.0046
32	34	23.454	4.414	5.402	6.313	0.7400	0.0046
32	34	23.610	4.414	5.384	6.310	0.7400	0.0046
32	100	22.610	4.414	5.350	6.236	0.7392	0.0077
32	100	22.554	4.414	5.349	6.232	0.7392	0.0077
32	100	22.661	4.414	5.353	6.241	0.7392	0.0077
32	100	22.497	4.414	5.318	6.208	0.7392	0.0077

TABLE XXIII (Continued)

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
32	100	22.486	4.414	5.328	6.214	0.7392	0.0077
32	100	22.717	4.414	5.342	6.237	0.7392	0.0077
32	200	21.407	4.414	5.237	6.114	0.7303	0.0128
32	200	21.248	4.414	5.282	6.134	0.7303	0.0128
32	200	21.105	4.414	5.223	6.086	0.7303	0.0128
32	200	21.631	4.414	5.227	6.121	0.7303	0.0128
32	200	21.262	4.414	5.240	6.107	0.7303	0.0128
32	300	20.073	4.414	5.186	6.017	0.7242	0.0181
32	300	20.610	4.414	5.163	6.036	0.7242	0.0181
32	300	20.261	4.414	5.210	6.045	0.7242	0.0181
32	300	20.076	4.414	5.163	6.002	0.7242	0.0181
32	300	20.530	4.414	5.121	6.003	0.7242	0.0181
32	300	20.563	4.414	5.187	6.049	0.7242	0.0181
32	500	17.342	4.414	5.013	5.760	0.7125	0.0294
32	500	17.433	4.414	5.026	5.775	0.7125	0.0294
32	500	17.513	4.414	5.080	5.816	0.7125	0.0294
32	500	17.429	4.414	5.054	5.793	0.7125	0.0294
32	500	17.389	4.414	5.043	5.783	0.7125	0.0294
32	500	17.499	4.414	5.054	5.798	0.7125	0.0294
32	500	17.449	4.414	5.035	5.782	0.7125	0.0294
32	500	17.626	4.414	5.031	5.792	0.7125	0.0294
32	500	17.625	4.414	5.077	5.822	0.7125	0.0294
32	500	17.786	4.414	5.058	5.821	0.7125	0.0294
32	500	17.682	4.414	5.028	5.794	0.7125	0.0294
32	750	15.263	4.414	4.916	5.594	0.6986	0.0446
32	750	15.197	4.414	4.938	5.603	0.6986	0.0446
32	750	15.127	4.414	4.942	5.600	0.6986	0.0446
32	750	15.349	4.414	4.921	5.604	0.6986	0.0446
32	750	15.045	4.414	4.898	5.565	0.6986	0.0446
32	750	15.013	4.414	4.922	5.578	0.6986	0.0446
32	750	15.474	4.414	4.886	5.591	0.6986	0.0446
32	750	15.107	4.414	4.912	5.579	0.6986	0.0446
32	1000	12.851	4.414	4.761	5.346	0.6856	0.0611
32	1000	12.662	4.414	4.758	5.327	0.6856	0.0611
32	1000	12.715	4.414	4.752	5.328	0.6856	0.0611
32	1000	12.885	4.414	4.786	5.365	0.6856	0.0611
32	1000	12.758	4.414	4.782	5.351	0.6856	0.0611
32	1000	12.905	4.414	4.791	5.370	0.6856	0.0611
32	1000	12.689	4.414	4.740	5.318	0.6856	0.0611
32	1000	12.877	4.414	4.748	5.340	0.6856	0.0611
32	1000	12.703	4.414	4.749	5.325	0.6856	0.0611
32	1000	12.537	4.414	4.735	5.301	0.6856	0.0611
32	1000	12.788	4.414	4.762	5.341	0.6856	0.0611
32	1000	12.826	4.414	4.763	5.345	0.6856	0.0611
32	1000	12.818	4.414	4.761	5.343	0.6856	0.0611
32	1000	12.616	4.414	4.766	5.328	0.6856	0.0611
32	1000	12.513	4.414	4.740	5.302	0.6856	0.0611
32	50	22.249	4.364	5.274	6.138	0.7361	0.0069
32	50	22.966	4.364	5.247	6.162	0.7361	0.0069

TABLE XXIII (Continued)

TEMP. °F	PRESS. PSIA	GAMMA DYNES/CM	d_n	d_s	d_e	LIQUID DENSITY	VAPOR DENSITY
32	50	22.966	4.364	5.253	6.166	0.7361	0.0069
32	200	20.971	4.364	5.121	5.986	0.7283	0.0139
32	200	20.675	4.364	5.132	5.975	0.7283	0.0139
32	200	20.637	4.364	5.122	5.966	0.7283	0.0139
32	500	17.721	4.364	4.923	5.704	0.7111	0.0306
32	500	17.500	4.364	4.919	5.686	0.7111	0.0306
32	500	17.484	4.364	4.901	5.673	0.7111	0.0306
32	750	14.269	4.364	4.746	5.382	0.6974	0.0459
32	750	14.380	4.364	4.777	5.411	0.6974	0.0459
32	1000	12.396	4.364	4.679	5.231	0.6847	0.0626
32	1000	12.668	4.364	4.647	5.235	0.6847	0.0626
32	1000	12.739	4.364	4.600	5.211	0.6847	0.0626
32	100	21.128	4.322	5.200	6.031	0.7239	0.0139
32	100	21.160	4.322	5.197	6.031	0.7239	0.0139
32	100	21.547	4.322	5.136	6.014	0.7239	0.0139
32	100	21.301	4.322	5.181	6.029	0.7239	0.0139
32	100	21.580	4.322	5.148	6.024	0.7239	0.0139
32	200	20.257	4.322	5.080	5.910	0.7212	0.0181
32	200	19.940	4.322	5.121	5.917	0.7212	0.0181
32	200	20.314	4.322	5.055	5.897	0.7212	0.0181
32	200	19.955	4.322	5.068	5.883	0.7212	0.0181
32	200	20.348	4.322	5.091	5.923	0.7212	0.0181
32	300	19.319	4.322	4.991	5.810	0.7161	0.0234
32	300	19.095	4.322	5.048	5.833	0.7161	0.0234
32	300	19.171	4.322	5.045	5.836	0.7161	0.0234
32	300	19.285	4.322	5.008	5.819	0.7161	0.0234
32	300	19.132	4.322	5.017	5.815	0.7161	0.0234
32	300	19.384	4.322	5.033	5.842	0.7161	0.0234
32	300	19.401	4.322	5.045	5.851	0.7161	0.0234
32	300	19.422	4.322	5.055	5.859	0.7161	0.0234
32	300	19.326	4.322	5.010	5.823	0.7161	0.0234
32	300	19.276	4.322	5.068	5.858	0.7161	0.0234
32	500	17.489	4.322	4.850	5.633	0.7059	0.0347
32	500	17.591	4.322	4.853	5.642	0.7059	0.0347
32	500	17.363	4.322	4.883	5.646	0.7059	0.0347
32	500	16.999	4.322	4.933	5.653	0.7059	0.0347
32	500	17.036	4.322	4.839	5.594	0.7059	0.0347
32	500	17.104	4.322	4.809	5.579	0.7059	0.0347
32	750	14.564	4.322	4.721	5.382	0.6934	0.0502
32	750	14.602	4.322	4.724	5.387	0.6934	0.0502
32	750	14.410	4.322	4.726	5.373	0.6934	0.0502

APPENDIX D

TERNARY SYSTEMS COMPOSITIONS

TABLE XXIV

SYSTEM COMPOSITION FOR ETHANE-BUTANE-DECANE RUNS

Temp. °F	Composition Parameter	Press. psia	No. of Moles in System*		
			Ethane	Butane	Decane
170	0.38	50	0.0	1.96	3.10
		100	1.45	1.96	3.10
		200	5.11	1.96	3.10
		300	9.09	1.96	3.10
		400	13.42	1.96	3.10
		500	18.20	1.96	3.10

* Number of moles in the experimental system were multiplied by 1000 to arrive at the numbers presented here.

TABLE XXV

SYSTEM COMPOSITION FOR METHANE-CARBON DIOXIDE-DECANE RUNS

Temp. °F	Composition Parameter	Press. psia	No. of Moles in System *		
			Methane	Carbon Dioxide	Decane
76	0.40	34	0.0	1.40	2.06
		100	2.21	1.40	2.06
		200	5.90	1.40	2.06
		300	9.67	1.40	2.06
		500	17.52	1.40	2.06
		750	27.80	1.40	2.06
		1000	38.50	1.40	2.06
32	0.35	34	0.0	1.40	2.58
		100	2.95	1.40	2.58
		200	7.37	1.40	2.58
		300	11.98	1.40	2.58
		500	21.70	1.40	2.58
		750	34.70	1.40	2.58
		1000	49.00	1.40	2.58
	0.52	50	0.0	1.96	1.80
		200	6.81	1.96	1.80
		500	21.14	1.96	1.80
		750	34.14	1.96	1.80
		1000	48.44	1.96	1.80
	0.68	100	0.0	3.92	1.85
		200	4.85	3.92	1.85
		300	9.46	3.92	1.85
		500	19.18	3.92	1.85
		750	32.18	3.92	1.85

* Number of moles in the experimental system were multiplied by 1000 to arrive at the numbers presented here.

VITA

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