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APPLICATION OF SHERESHEFSKY'S THEORY OF BINARY SOLUTION SURFACE TENSION

By

Donald J. Cotton

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Application of Shereshefsky's Theory of Binary Solution Surface Tension

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Naval Ship Research and Development Center
Washington, D.C. 20007

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APPLICATION OF SHERESHEFSKY'S THEORY OF BINARY SOLUTION SURFACE TENSION

By
Donald J. Cotton

February 1969 MATLAB 266 Report 2851

ABSTRACT

Shereshefsky's equation describing the surface tension of binary systems has been applied to cryogenic liquid, metal alloy, organic, and molten salt binary systems. It has been demonstrated that the equation successfully predicts the behavior of these systems in terms of the physical properties of their components. It has been shown how the equation can be used to ascertain the surface area and the orientation of molecules in the surface region of the binary solution.

ADMINISTRATIVE INFORMATION

This research is a product of the IR/IED Program, Subproject Z-R011 01 01, Task 05600, NAVSHIPRANDLAB Annapolis Assignment A821 147.

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NAVAL SHIP RESEARCH AND DEVELOPMENT LABORATORY

APPLICATION OF SHERESHEFSKY'S THEORY OF BINARY SOLUTION SURFACE TENSION

By
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INTRODUCTION

Equation [1], based on thermodynamic considerations and on an assumed model of the surface region of a binary solution, has been derived by Shereshefsky.[1]

$$[1] \quad \frac{N_{21}}{\Delta\sigma} = \frac{1}{\Delta\sigma_o} e^{-\Delta F_{12}/RT} + \frac{N_{21}}{\Delta\sigma_o} \left(1 - e^{-\Delta F_{12}/RT}\right)$$

where $\Delta\sigma = \sigma - \sigma_1$

$\Delta\sigma_o = \sigma_2 - \sigma_1$

$\Delta F_{12} = (\sigma_1 - \sigma_2)\, A_{2s}/t$

N_{21} = solute mole fraction

σ = surface tension of solution

σ_1 = surface tension of solvent

σ_2 = surface tension of solute

A_{2s} = molecular surface area of solute

t = thickness of adsorbed layer

T = absolute temperature

R = gas constant

Accordingly when $\frac{N_{21}}{\Delta\sigma}$ is plotted versus N_{21} a straight line should result and -

$$[2] \quad \text{slope} = b = \frac{1}{\Delta\sigma_o} \left(1 - e^{-\Delta F_{12}/RT}\right)$$

$$[3] \quad \text{intercept} = a = \frac{1}{\Delta\sigma_o} e^{-\Delta F_{12}/RT};$$

$$[4] \quad \Delta\sigma_o = \frac{1}{a + b}$$

$$[5] \quad \Delta F_{12} = RT \ln\left(\frac{b}{a} + 1\right)$$

Consequently, if surface tension as a function of concentration is known for a particular binary system, $\Delta\sigma_o$, ΔF_{12}, and A_{2s} can be calculated. The purpose of this paper is to demonstrate the general applicability of the derived equation with surface tension data from a number of widely different types of binary systems.

BINARY SOLUTIONS OF LIQUIFIED GASES

Surface tension data for cryogenic binary solutions of carbon monoxide, of methane, and of argon in nitrogen,[1a] and solutions of argon, of carbon monoxide,[1a] and of krypton[2] in methane were analyzed. A least square fit of mole fraction divided by $\Delta\sigma$ plotted versus mole fraction was made for each system, and $\Delta\sigma_o$, ΔF_{12}, and A_{2s} were calculated by applying Equations [1-3]. Results are summarized in Figures 1-3 and in Table 1.

Table 1

Surface Energy and Surface Area Constants for Several Binary Systems of Liquified Gases

System Solute-solvent	T	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF_{12} erg/m x 10^{-9}	A_{2s} (calc.) $\mathring{A}^2$	A_{2s} (dens.)[3] $\mathring{A}^2$	t (layers)
N_2 - Ar	83.82	(5.48) 5.98	5.97	(4.60) 3.06	(14.0) 8.5	16.2(78°K)	2
N_2 - CO	83.82	1.62	1.60	4.30	4.5	16.2(78°K)	4
N_2 - CH_4	90.67	10.91	11.80	10.6	17.4	17.0(90°K)	1
Ar - CH_4	90.67	6.14	6.13	5.58	14.4	14.4(90°K)	1
CO - CH_4	90.67	10.00	10.13	7.87	16.8	16.8(90°K)	1
CH_4 - Kr	116.00	4.25	4.24	3.96	15.5	18.1(133°K)	1

In Table 1, $\Delta\sigma_o$ (calc) was obtained by using Equation [4], $\Delta\sigma_o$ (obs.) was obtained from the actual data, A_{2s} (calc.) was calculated with Equation [5], and A_{2s} (dens.) was determined for the same molecule by Emmett[3] from density data and Equation [6]:

$$[6] \qquad A = 1.091 \left(\frac{\bar{V}}{N}\right)2/3$$

where $\bar{V}$ = molar volume

N = Avogadros number.

Two straight lines resulted for the N_2 - Arsystem. The values in parentheses for this system is for the dilute region. $\Delta\sigma_o$ (calc.) corresponds to $\Delta\sigma_o$ (obs.) to within ± 1 erg/cm^2 for all systems, except N_2 - CH_4, which was expected inasmuch as their difference is indicative of the degree of orientation of surface molecules. A_{2s} (calc.) also corresponds closely to an integer multiple of A_{2s} (dens.), except for the CH_4 - Kr system. It is significant that A_{2s} (calc.) for this system is approximately 15.0Å^2, the molecular area obtained from density data for the solidified gas.[3] This implies that methane molecules in the surface region are packed as in a pure solid state. It is also significant that A_{2s} (calc.) obtained for dilute solutions of the N_2 - Arsystem is approximately 14.4Å^2, the A_{2s} (dens.) value for argon,[3] and that the two possible straight lines intersect at 0.5 mole fraction. This implies that a nitrogen molecule in dilute solution is oriented to occupy the exact area of a displaced argon molecule.

BINARY SOLUTIONS OF ORGANIC LIQUIDS

Surface tension data of solutions of isooctane in benzene, in cyclohexane, and in dodecane,[4] of n-hexane in benzene[5] and in cyclohexane,[6] and of n-dodecane in benzene.[5] The data were treated as previously described. Results are summarized in Tables 2-3 and Figures 4-7.

Table 2

Surface Energy and Surface Area Constants for Several Systems of Isooctane in Organic Solvents -- at 30°C

Solvent	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF erg/m x 10^{-10}	A_{2s} (calc.) $\mathring{A}^2$	A_{2s} (dens.) $\mathring{A}^2$
Benzene	9.60	9.64	2.90	49.9	42.6
Cyclohexane	5.84	5.88	2.01	56.7	
Dodecane	6.36	6.58	0.16	4.2	

The nature of the solvent apparently affects the magnitude of A_{2s} (calc.) for the isooctane-benzene and for the isooctane-cyclohexane systems where it approaches $50\mathring{A}^2$, the molecular surface area of a solvent molecule oriented with its ring plane parallel to the surface plane. Apparently the isooctane molecules in these systems orient and pack into the surface so as to displace solvent molecules on a one-to-one basis. A_{2s} (calc.) for the isooctane-dodecane system implies that the surface region is ten molecules thick if random orientation is assumed. However, if the isooctane molecules are assumed to orient with their major axes normal to the surface plane so as to have an effective area of a methyl group, $20\mathring{A}^2$, then the molecular layer is five molecules thick. The same effect can also be obtained by increasing the kinetic energy of the system as evidenced by the results in Table 3 obtained from the n-hexane-cyclohexane system at various temperatures. Both effects are simultaneously displayed in the results obtained from the n-hexane-benzene system in the same table. The n-dodecane-benzene system has an unexpectedly high A_{2s} (calc.) which can be explained by assuming that the dodecane molecule orients with its major axis parallel to the surface plane so as to displace from six to eight benzene molecules oriented with their ring plane parallel to the surface.

Table 3

Surface Energy and Surface Area Constants for Several Binary Systems of Organic Liquids - at Various Temperatures

T°C	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF erg/m x 10^{-10}	A_{2s} (calc.) $\AA^2$	A_{2s} (dens.) $\AA^2$
n-hexane in benzene					
25	10.10	10.21	2.47	40.6	36.3
30	10.07	10.05	2.44	40.3	
35	9.87	9.91	2.43	40.1	
40	9.74	9.76	2.34	39.9	
n-hexane in cyclohexane					
25	6.23	6.28	1.23	32.5	36.3
30	6.35	6.35	1.08	28.2	
35	6.17	6.17	0.97	26.2	
n-dodecane in benzene					
25	3.50	3.46	6.43	305.0	52.4
30	3.15	3.17	6.72	354.6	
35	2.95	2.96	6.53	367.7	
40	2.75	2.72	6.75	408.5	

Data on the surface tension of molten mixtures of silver and tin at various temperatures measured by Lauermann, Metzger and Sauerwald[7] were fitted to the derived equation. Results are summarized in Table 4 and Figure 8. In Table 4, A_{2s} (ion.) is the molecular surface area calculated using the Pauling ionic radius of tin,[8] .71$\mathring{A}^2$ and A_{2s} (cov.) is the molecular surface area calculated using the Pauling covalent radius of tin,[9] 1.412$\mathring{A}^2$. A_{2s} (calc.) closely approximates A_{2s} (ion.) which implies that tin molecules in the surface region are in the ionic state. However, the ratio of A_{2s} (cov.) and A_{2s} (calc.) indicates that the surface region is four layers thick with tin molecules. The latter interpretation is attractive inasmuch as normally covalent metallic bonds are expected to exist in a metal alloy, yet an adsorbed layer four molecules thick is unusually high. In both cases the results predict that the surface of a liquid metal binary system is rich in the component of lower surface tension. Inasmuch as no large migration of atoms is expected upon solidification, the solid surface of the alloy is also expected to be rich in the same component.

Table 4

Surface Energy and Surface Area Constants for Molten Mixtures of Tin and Silver at Various Temperatures

T (°C)	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF erg/m x 10^{-10}	A_{2s} (calc.) $\mathring{A}^2$	A_{2s} (ion) $\mathring{A}^2$	A_{2s} (cov.) $\mathring{A}^2$	t (ion) layers	t (cov.) layers
1000	400.0	399.0	14.93	6.20	6.33	25.1	1.02	4.05
1100	393.0	396.0	15.56	6.56	6.33	25.1	0.96	3.83
1200	383.0	386.0	14.48	6.28	6.33	25.1	1.00	4.00

BINARY MIXTURES OF MOLTEN SALTS

Surface tension data for various molten mixtures of alkali metal nitrates, alkali metal nitrates and silver nitrate, and alkali metal chlorides and sulfates obtained by Bertozzi and Sternheim[10] and Bertozzi and Soldani[11] were analyzed. The results are shown in Tables 5-7 and Figures 9-11.

Table 5

Surface Energy and Surface Area Constants for Molten Mixtures of Silver and Alkali Nitrates at 350°C

Solute	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF erg/m x 10^{-10}	A_{2s} (calc.) $\AA^2$	A_{2s} (dens.) $\AA^2$	A_{2s} (ion.) $\AA^2$	t (dens.) layers	t (ion) layers
$LiNO_3$	30.12	29.95	2.53	14.0	16.3	23.2	1.2	1.7
$NaNO_3$	24.56	24.00	3.89	26.9	17.7	30.0	0.7	1.1
KNO_3	29.32	29.15	7.69	43.8	20.2	40.9	0.5	0.9
$RbNO_3$	35.42	35.35	8.36	39.3	21.6	46.2	0.5	1.2
$CsNO_3$	43.78	44.40	8.36	31.3	23.3	54.6	0.7	1.7

Table 6

Surface Energy and Surface Area Constants for Molten Mixtures of Alkali Nitrate at 350°C

Solute	Solvent	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF erg/m x 10^{-10}	A_{2s} (calc.) $\AA^2$	A_{2s} (dens.) $\AA^2$	A_{2s} (ion.) $\AA^2$	t (dens.) layers	t (ion.) layers
KNO_3	$NaNO_3$	5.64	5.18	2.56	75.4	20.2	40.9	0.3	0.5
$RbNO_3$	KNO_3	6.25	6.20	1.76	46.9	21.6	46.2	0.5	1.0
$CsNO_3$	KNO_3	12.94	15.25	4.04	51.8	23.3	54.6	0.4	1.0
$RbNO_3$	$NaNO_3$	12.53	11.35	3.62	48.0	21.6	46.2	0.5	1.0
$CsNO_3$	$NaNO_3$	20.47	20.40	5.42	44.0	23.3	54.6	0.5	1.2

Table 7

Surface Energy and Surface Area Constants for Molten Mixtures of Alkali Chloride and Alkali Sulfate at 1200°C

Solute	Solvent	$\Delta\sigma_o$ (calc.) erg/cm^2	$\Delta\sigma_o$ (obs.) erg/cm^2	ΔF erg/m x 10^{-10}	A_{2s} (calc.) $\mathring{A}^2$	A_{2s} (dens.) $\mathring{A}^2$	A_{2s} (ion.) $\mathring{A}^2$	t (dens.) layers	t (ion.) layers
NaCl	Na_2SO_4	83.5	83.4	3.14	6.7	17.0	41.2	2.5	6.1
KCl	K_2SO_4	64.0	58.3	6.66	20.4	20.8	41.2	1.0	2.0

In Tables 6-7, A_{2s} (calc.) and A_{2s} (dens.) refers to solute molecules. A_{2s} (ion.) in Tables 5-6 is the sum of the ionic surface areas of the cation and the anion comprising the solute molecule. Pauling ionic radii were used for the cations, and 1.22$\mathring{A}$ was used as the radius of the nitrate ion[12] which was assumed to be symmetrical. In Table 7, A_{2s} (Cl^-) is the ionic surface area for a monovalent chloride ion. Surface layer thickness t is labeled according to the molecular surface area employed in its calculation.

In Tables 5-6, t (dens.) is a noninteger which is physically forbidden. This indicates, as expected for an ionic mixture, that the alkali nitrate molecules are not randomly oriented in the surface region. The surface layer thickness t (ion), however, does closely approximate an integer, except for the (K-Na) NO_3 system. A_{2s} (ion), used to calculate t (ion), is based on a surface structure consisting of a cation lying adjacent to an anion as found in the solid state. Consequently, these results suggest that the surface structure of the molten mixture is the same as for the solid state.

In Table 7, t (dens.) for the $NaCl/Na_2SO_4$ system, unlike that for the KCl/K_2SO_4 system, is noninteger. t (Cl^-), however, is approximately an

integer for both systems which indicates that the surface region is comprised of a layer of chloride ions. Since the ionic surface area of a chloride ion is much larger than that of either cation, this result is unsurprising.

SUMMARY

Shereshefsky's equation describing the surface tension of binary systems has been applied to a large number of widely different binary systems. The equation has successfully predicted the behavior of these systems. It has been demonstrated how the equation can be used to ascertain the surface area and the orientation of molecules in the surface region of a binary solution.

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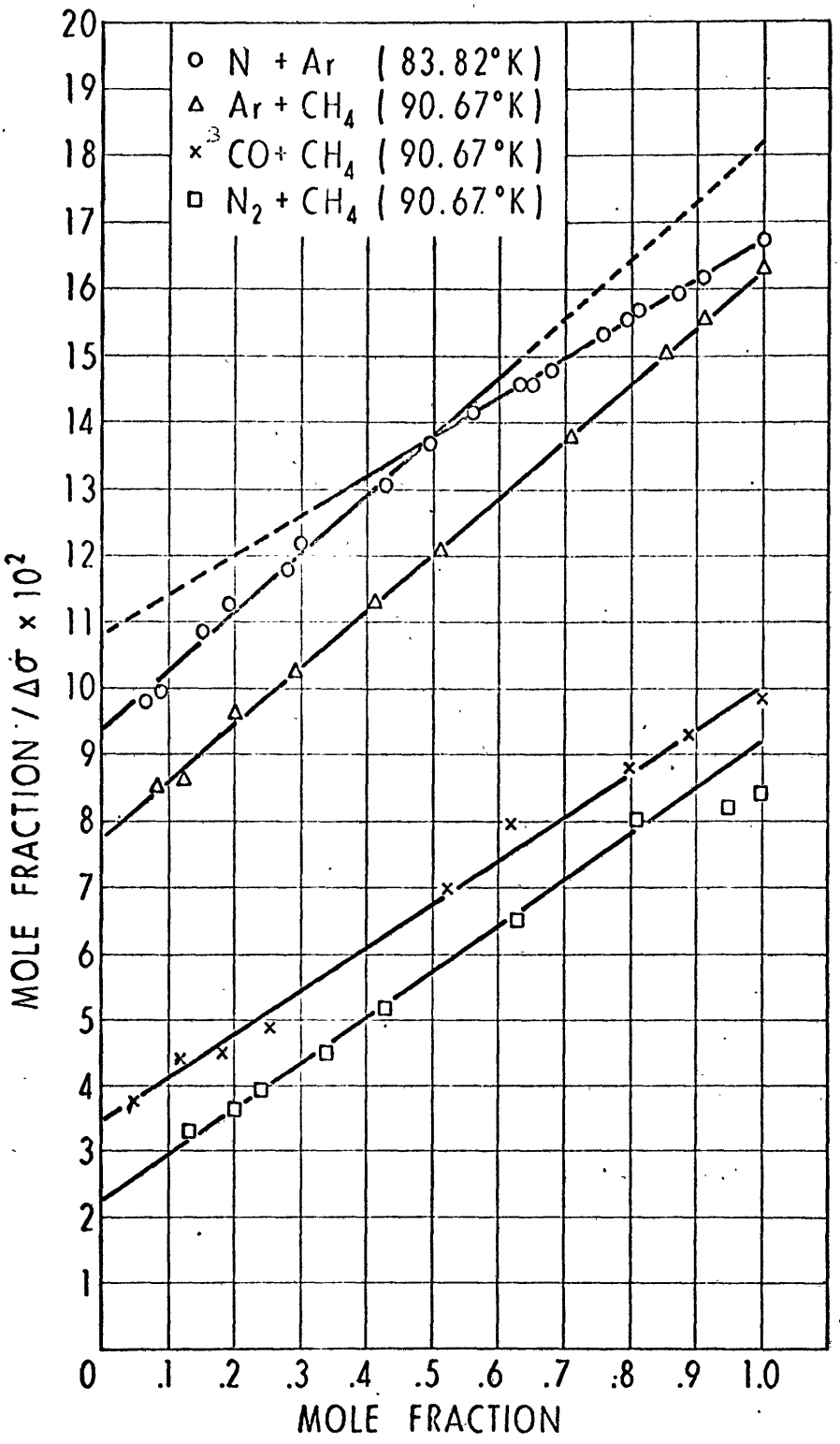

Figure 1
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fractions For Various Solutions of Liquefied Gasses at Various Temperatures

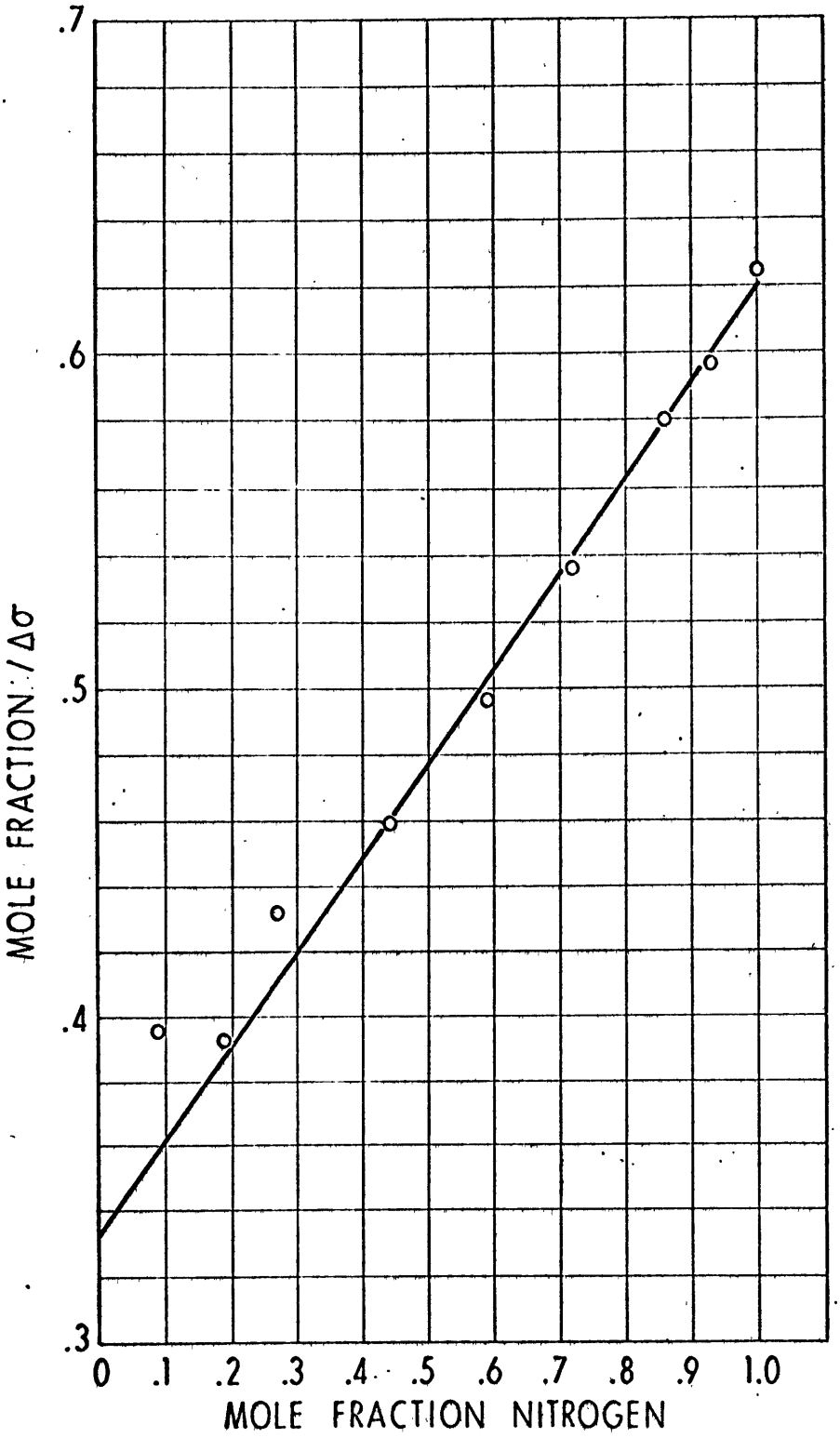

Figure 2
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction For Solutions of Liquid Nitrogen and Carbon Monoxide at 83.82° K

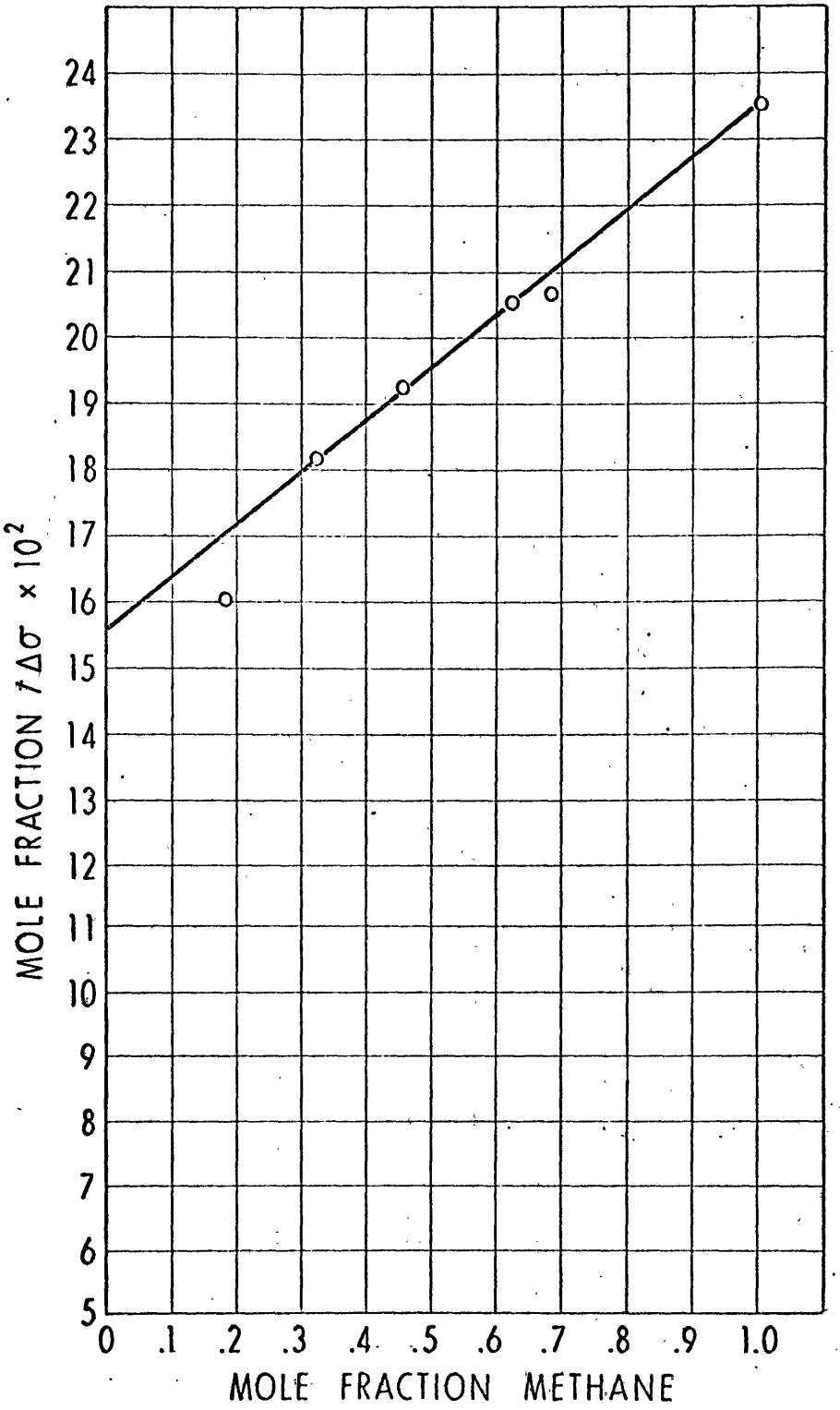

Figure 3
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction For Liquefied Mixtures of Methane in Krypton at 116° K

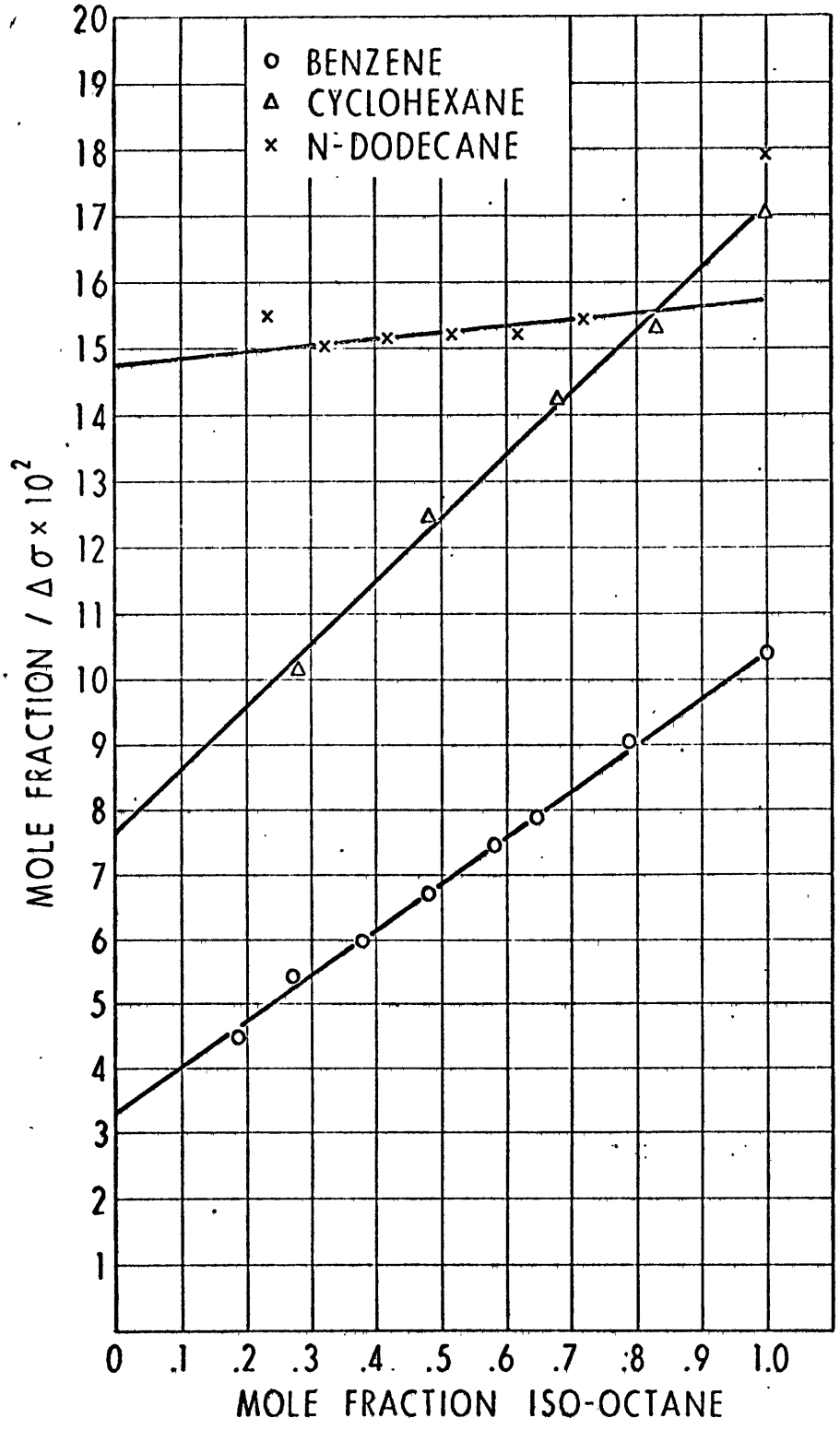

Figure 4
Mole Fraction Divided by Δσ Versus Mole Fraction For Solutions of Iso-Octane in Various Organic Solvents at 30° C

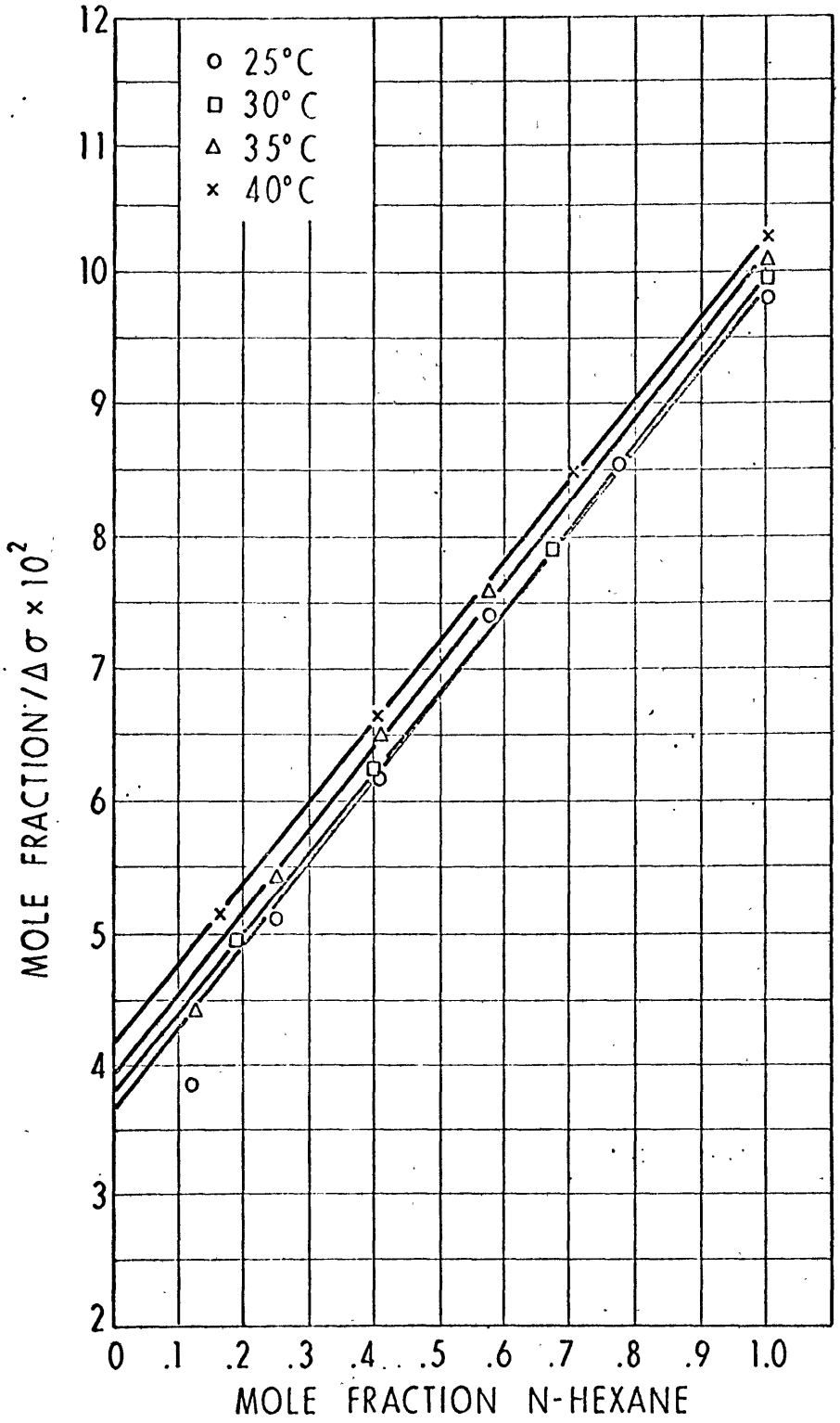

Figure 5
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction For Solutions of N-Hexane in Benzene at Various Temperatures

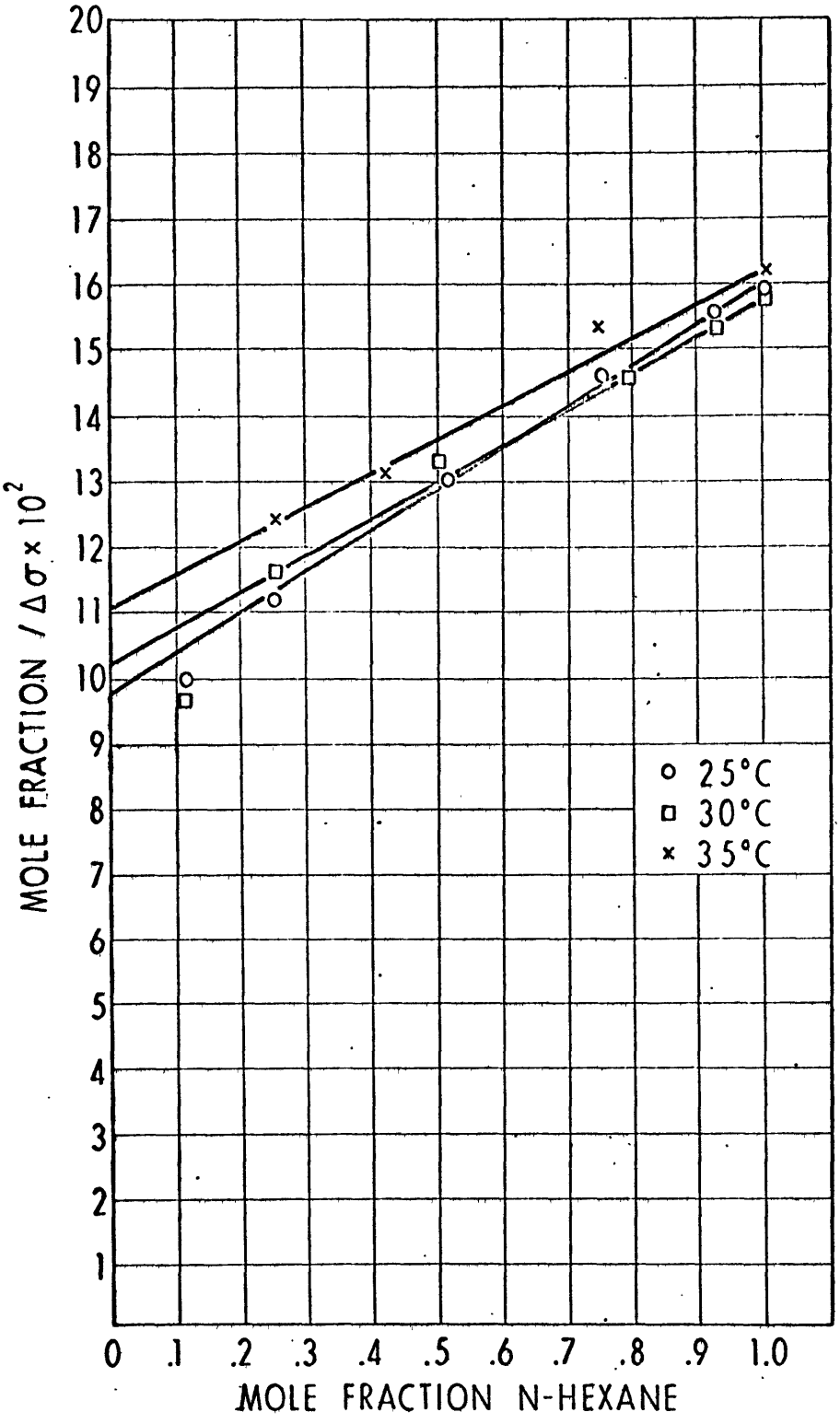

Figure 6
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction for N-Hexane-Cyclohexane Solutions at Various Temperatures

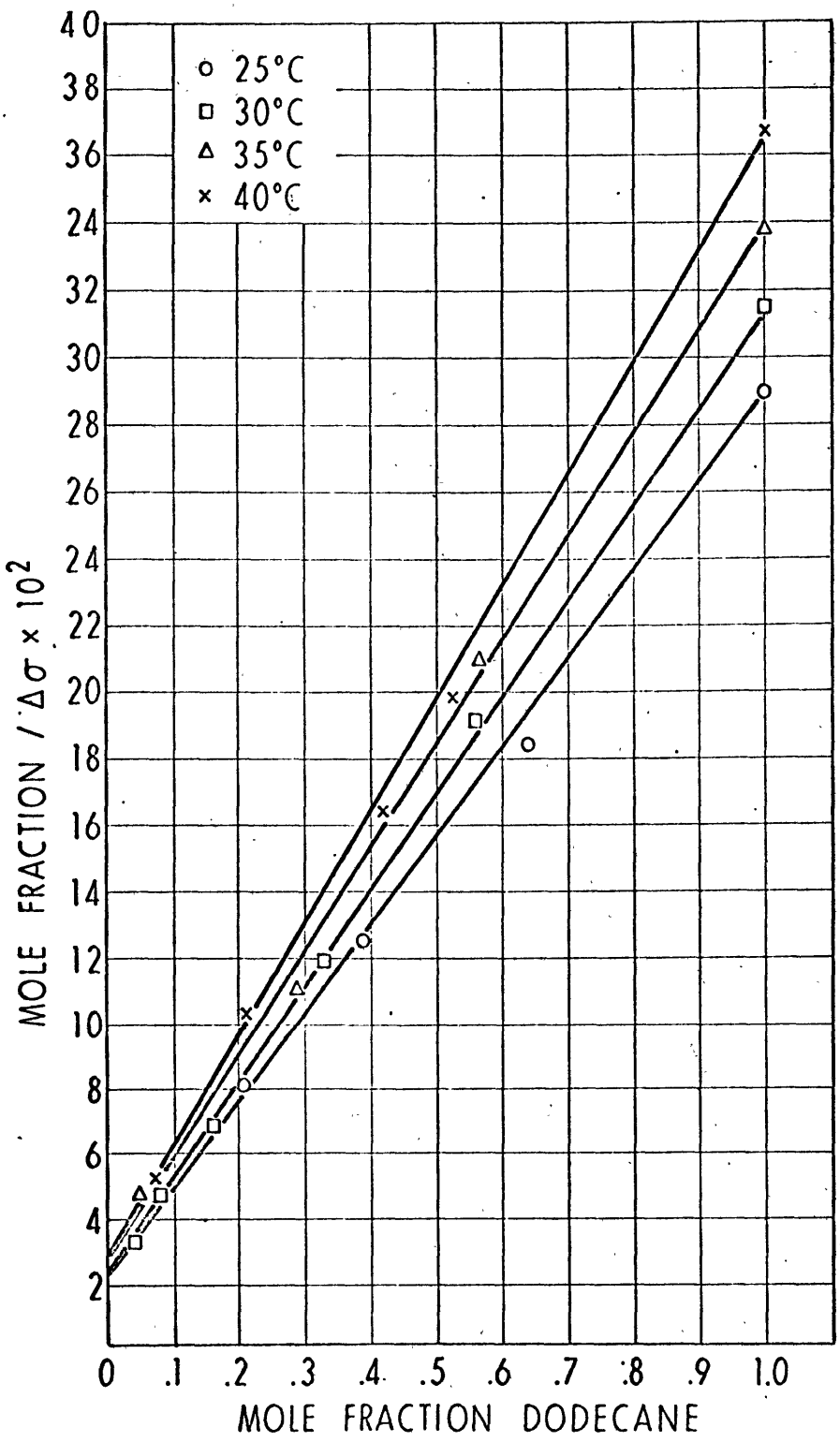

Figure 7
Mole Fraction Divided by Δσ Versus Mole Fraction For Solutions of Dodecane in Benzene at Various Temperatures

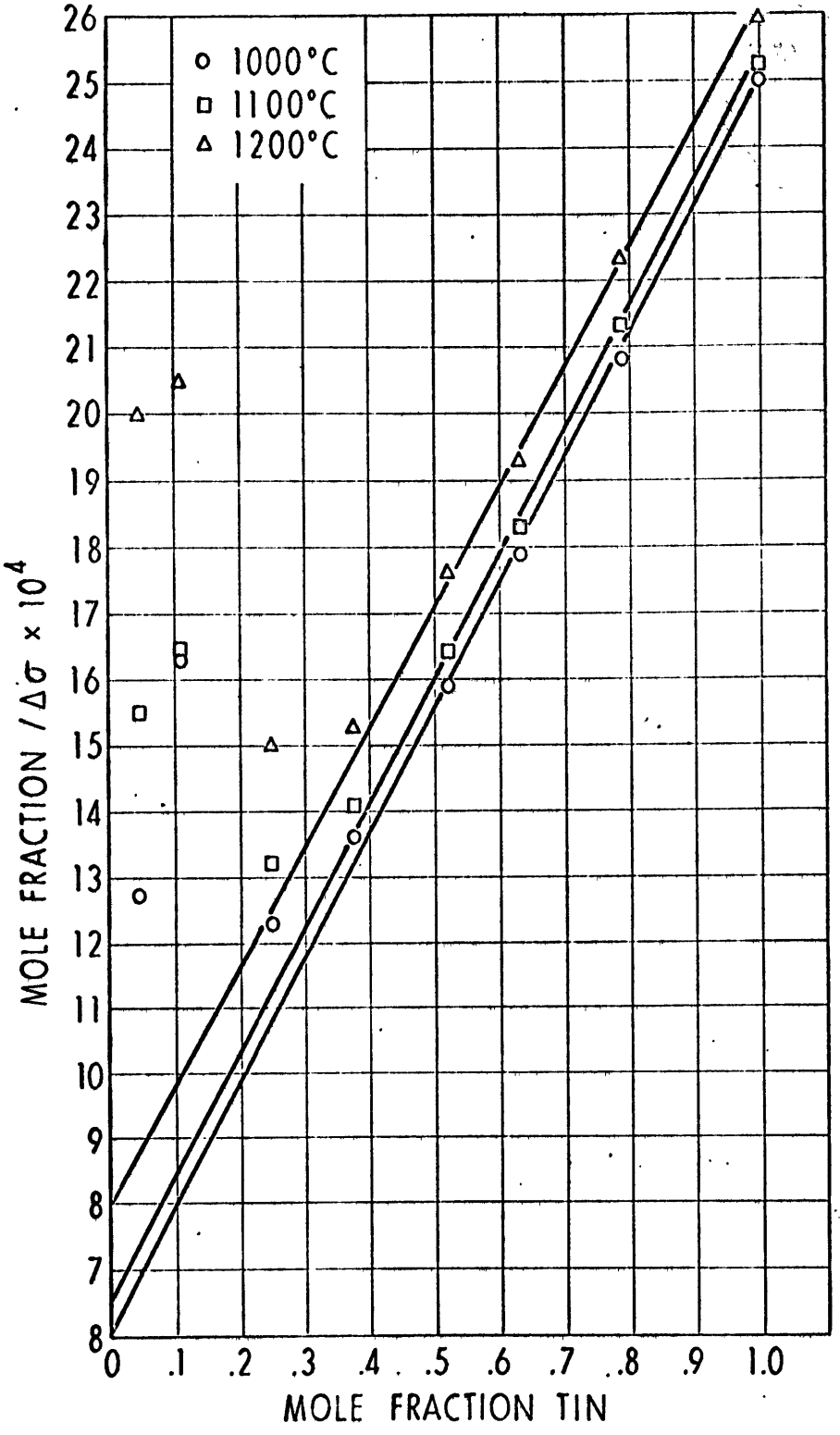

Figure 8
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction For Liquid Mixtures of Tin and Silver at Various Temperatures

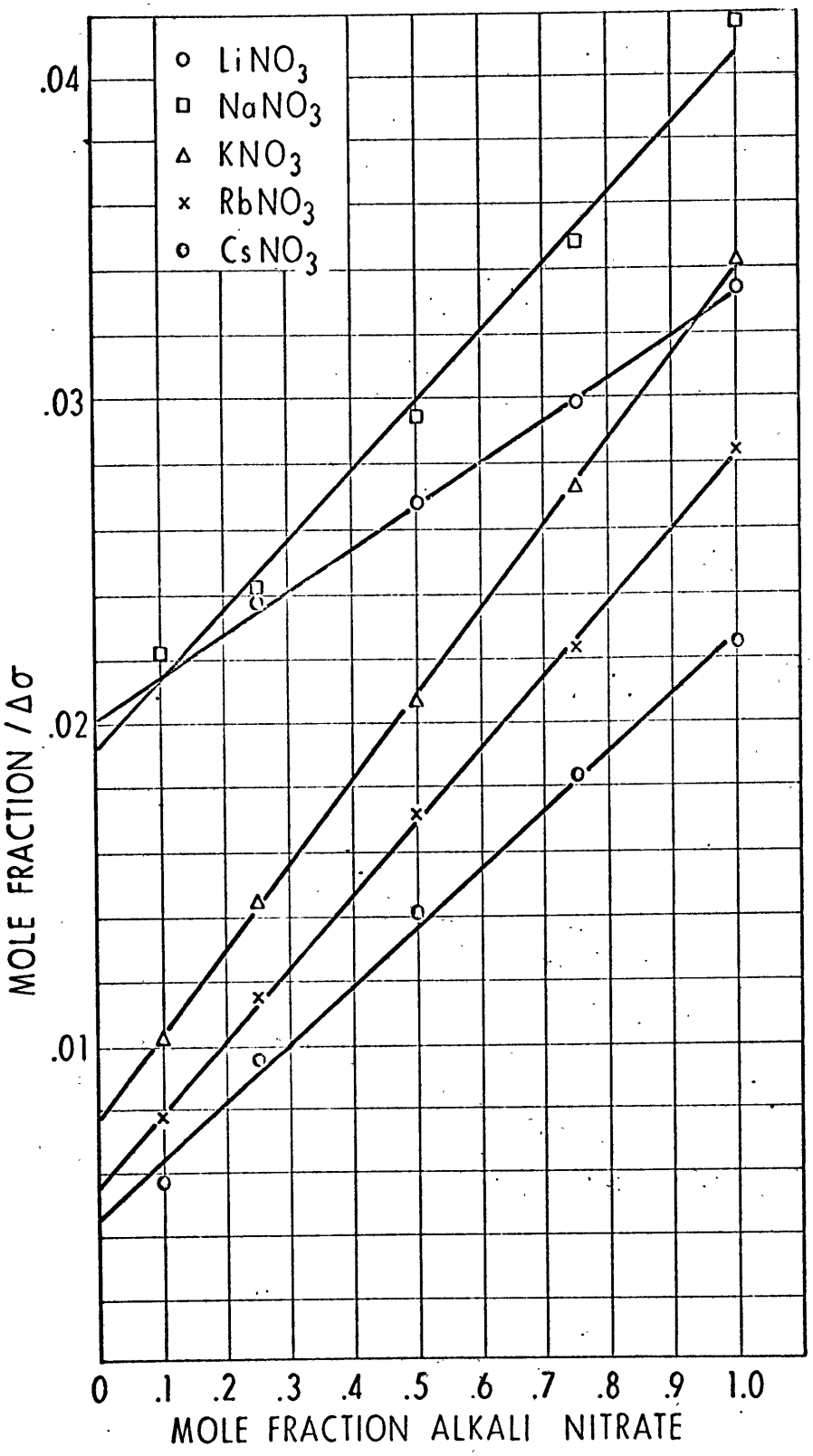

Figure 9
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction
For Liquid Solutions of Various Alkali Nitrates
Dissolved in Silver Nitrate at 350° C

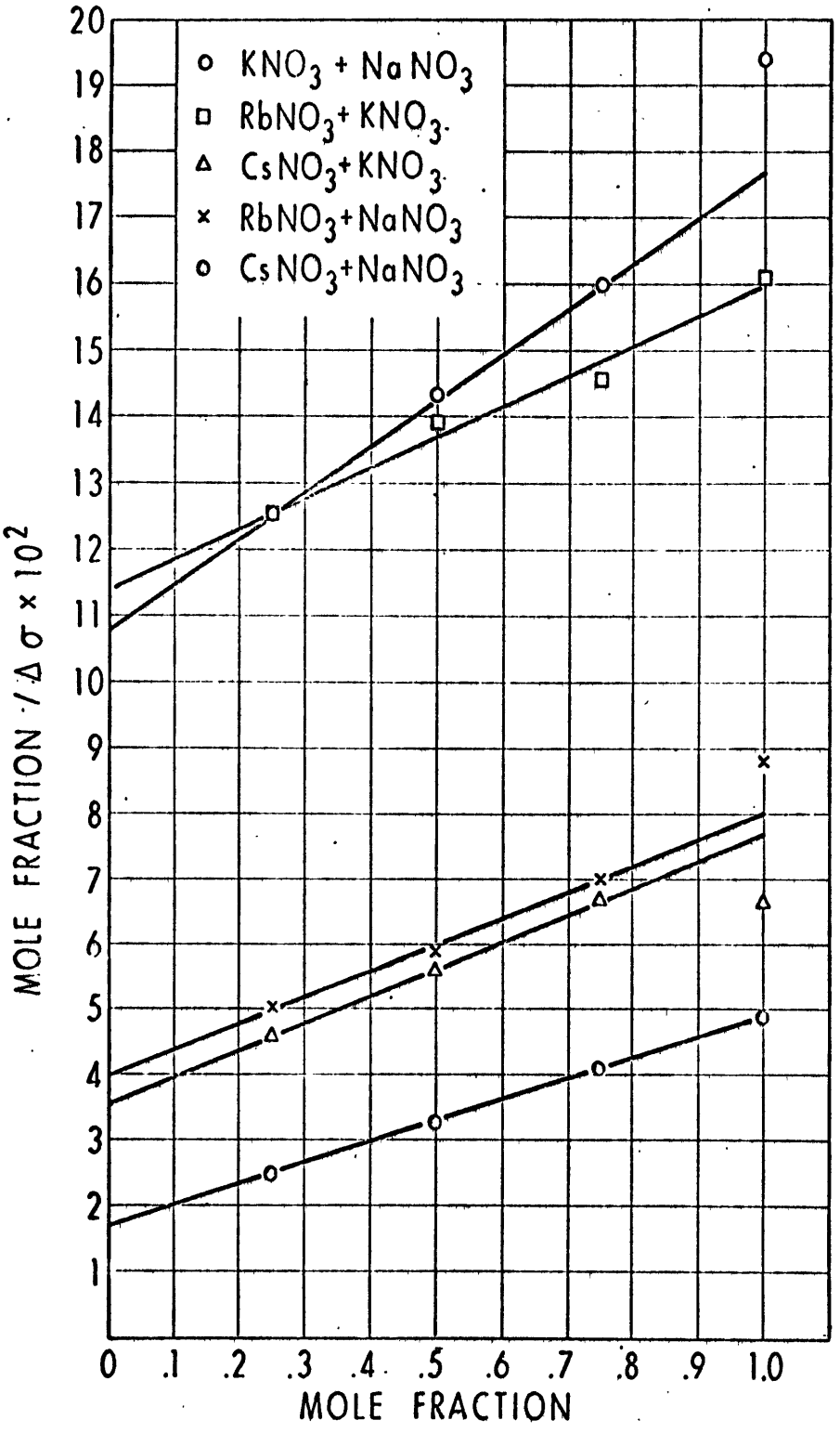

Figure 10
Mole Fraction Divided by Δσ Versus Mole Fraction For Liquid Mixtures of Various Alkali Nitrates at 350° C

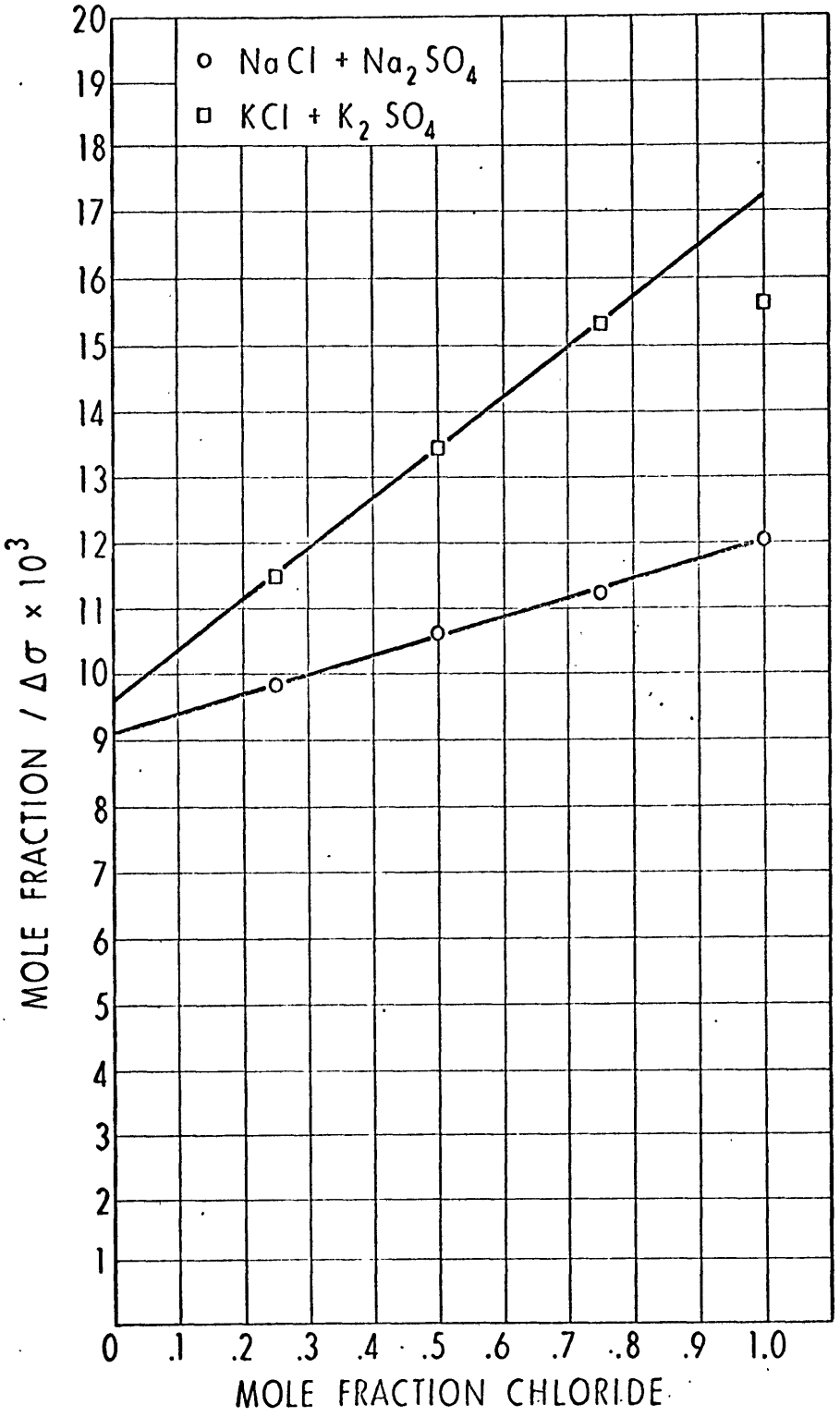

Figure 11
Mole Fraction Divided by $\Delta\sigma$ Versus Mole Fraction For Liquid Mixtures of Various Alkali Chlorides and Sulfates at 1200° C

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3 ABSTRACT

Shereshefsky's equation describing the surface tension of binary systems has been applied to cryogenic liquid, metal alloy, organic, and molten salt binary systems. It has been demonstrated that the equation successfully predicts the behavior of these systems in terms of the physical properties of their components. It has been shown how the equation can be used to ascertain the surface area and the orientation of molecules in the surface region of the binary solution.

(Author)

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Binary solutions						
Thermodynamics						
Surface tension						
Alloys						
Molten salt						
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Molecular area						

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